Ph.D. Dissertation

Alexandra Jakab-Nácsa

2024

Supervisor Recommendation

for the PhD Thesis titled

A Data-Driven Modelling Approach for Heterogeneous Catalysis Research

by Alexandra Jakab-Nácsa

Alexandra started her PhD studies as a development engineer at Wanhua BorsodChem within the Kerpely Antal PhD School at the University of Miskolc. The aim of her research is to design an industrially applicable catalyst qualification system that will help the company's production and development processes to operate more efficiently.

During the 4 years of the doctoral program, she carried out the data collection and data processing work completely independently, and after a critical analysis of the data, she started to develop the basis for the MIRA21 system. As her research progressed, she demonstrated increasing independence and exhibited creativity in manually verifying developed methods, as well as in organizing and documenting tasks associated with their application to specific systems.

Following the outstanding publication of the MIRA21 model, as an independent task in the Master of Chemical Engineering program, Alexandra conducted the application of the model to more than a dozen different heterocyclic processes with students in independent work. She also prepared the corresponding training materials. The educational utilization of the research results underscores the candidate's versatility.

The data processing phase of the research has opened new frontiers in the application of artificial intelligence. Collaborating with an experienced colleague, Alexandra produced remarkable analyses in a short time, and I am confident that this aspect of her PhD work will guide future developments.

The results reported in the publications and in the PhD dissertation, which Alexandra has included in her thesis, are her own findings and support her independent scientific work. Through intensive collaboration, I have come to regard

Alexandra as a motivated, persistent, and conscientious individual. Her exceptional work ethic and precision have been evident throughout the entire PhD period.

Based on her performance, I highly appreciate her research and recommend her to others. **Alexandra Jakab-Nácsa is suitable for independent research and I recommend that she be awarded the PhD degree.**

Miskolc, 2024.02.19.

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A Data-Driven Modelling Approach for Heterogeneous Catalysis Research

PhD dissertation

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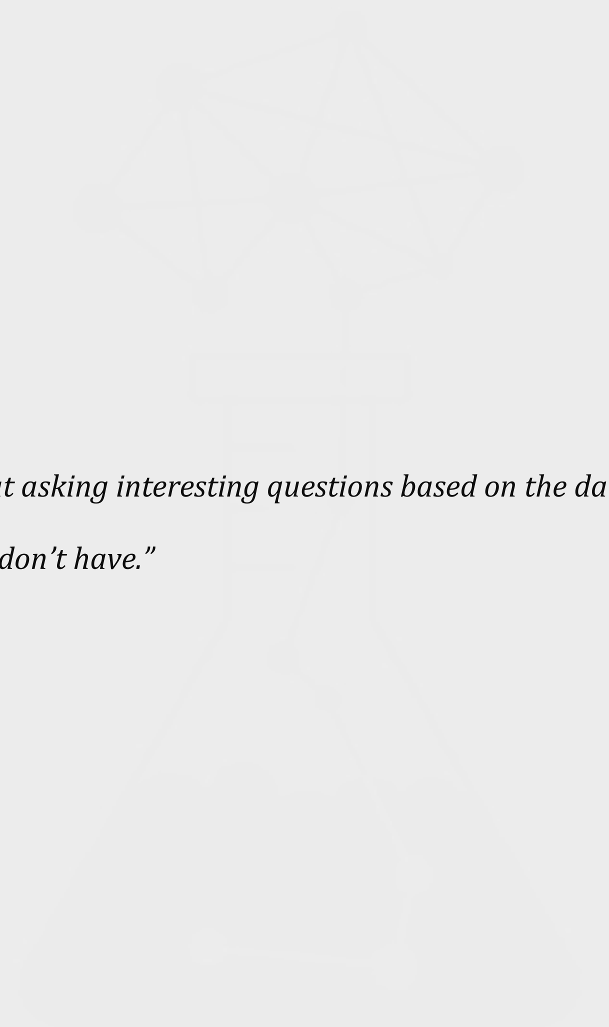


"Data science is all about asking interesting questions based on the data you have

—or often the data you don't have."

SARAH JARVIS

CONTENTS

1. ABBREVIATION

AREMO	Alkali Rare Earth Metal Oxide
BET	Brunauer–Emmett–Teller method
CAT	Catalyst
Class	Classification
COND	Reaction conditions
DFT	Density Functional Theory
DNT	Dinitrotoluene
EDA	Exploratory Data Analysis
HNB	Hydrogenation of Nitrobenzene
ID	Identification
IQR	Interquartile ranges
KKD	Knowledge Discovery in Databases
MCDM	Multi-Criteria Decision Making
MIRA 21	Miskolc Ranking 2021
ML	Machine Learning
MOF	Metal organic frameworks
N-BCNT	Nitrogen-doped Bamboo-like Carbon Nanotube
PER	Catalyst performance
PVP	Polyvinylpyrrolidone
RMSE	Root-mean-square error
SBA-15	Mesoporous silica - Santa Barbara Amorphous-15
SoS	Support on Support
STY	Space-Time Yield
SUS	Sustainability
TDA	Diaminotoluene
TMO	Transition Metal Oxide
TOF	Turnover Frequency
TON	Turnover Number

2. INTRODUCTION

2.1. AIM

Looking at the centuries-old history of the science of catalysis, the hope of fully recognising and understanding it seems to be slipping away. Despite today's technological tools, which allow a more detailed study of the phenomenon, the infinite pile of accumulated data creates confusion in clear vision. However, artificial intelligence has opened new horizons on data transformation into information and into knowledge[1]. Catalyst design is an area in which computational chemistry and machine learning techniques lead to outstanding results[2].

My doctoral dissertation aims to emphasize the data-driven catalyst design. The objective is to elevate the catalyst basic research results to the next level on the path to industrial application by using them in a practice-oriented way.

My research work is based on two pillars, whose database was built from more than 15.000 data points published in scientific literature. The first pillar is the establishment of MIRA21 (**MI**skolc **RA**nking 20**21**) model, a functional and practical mathematical model of catalyst characterization and exact comparison of each other (Figure 1). The second pillar of our research is the application of EDA (Exploratory Data Analysis), which refers to preliminary work on predicting catalyst composition through machine learning.

Figure 1 *Denomination of the model*

In the first chapter of this study, the phenomenon of catalysis is reviewed. The most important properties of catalysts, their classification, and the possibilities for catalyst characterization are summarized. The state of data analysis will be also presented briefly. The methodology of the MIRA21 model, as well as the presentation of the data analysis, are detailed in the chapter of the methods. In the next section, the first results of the MIRA21 model for the hydrogenation of nitrobenzene are presented. To investigate the extension of the application, the qualification of catalysts suitable for the hydrogenation of dinitrotoluene was also discussed here. In the discussion chapter, the results to date are reviewed and conclusions regarding catalyst design are drawn. The database has been expanded, and the MIRA model has been revised and modified. Finally, the points of thesis are determined.

2.2. INTRODUCTION TO CATALYSIS

Catalytic science is constantly evolving and plays an important role in many areas such as sustainable energy sources, environmental protection or drug development[3]. Design and development of catalysts enable many applications, such as increasing the efficiency of industrial processes, solving environmental problems, and creating new materials.

Rational catalysis design requires a complete understanding of the catalytic process, from the phenomena of catalysis, through the most important properties and their relationships, to the classification of the catalyst. Structure-activity relationship needs deeper and detailed knowledge of the above topics. This knowledge enables scientists and engineers to develop unique catalysts that improve efficiency and selectivity of chemical reactions.

Prior to proceeding, it should be noted that the relationship between academic research and industrial research is significant in this study. As a development engineer in the chemical industry, the practicality of the industrial environment in the research process must be considered, and, therefore, the industrial aspect is always mentioned in my dissertation in addition to the scientific approach.

2.2.1. PHENOMENON OF CATALYSIS

The history of chemistry development is closely intertwined with the phenomenon of catalysis[4]. The word is Greek consist of the words "*cata*" and "*lysein*", meaning down + tear or break, so that the catalyst can degrade the forces that inhibit the reaction. G. C. Bond, one of the well-known catalytic researchers, is humorous in his book, but he points out that although the word catalysis is used in different senses in different periods of history, they still have something in common[5]. Whether these inhibition forces are dismantled with a chemical substance or a marriage broker – because the Chinese term "*tsoo mei*" for a catalyst also means a marriage broker – the essence is to bring together the different parties[6].

The phenomenon of catalysis was already well-known to the ancient Greeks, but it was recognized only in the 19th century. Swedish chemist *Berzelius* was one of the pioneers in the field of chemistry. He has defined the phenomenon of catalysis, among many other discoveries, in 1836, and based it on the fact that at that time, substances that apparently did not participate in reactions but were necessary to make them occur, were already consciously used. Berzelius called the catalysis, according to its original meaning "*in a state outside the law*", and substances with such properties were called "*catalysts*"[7]. The concept of catalysis has been modified and clarified several times over the centuries, as a result of the development of science[8].

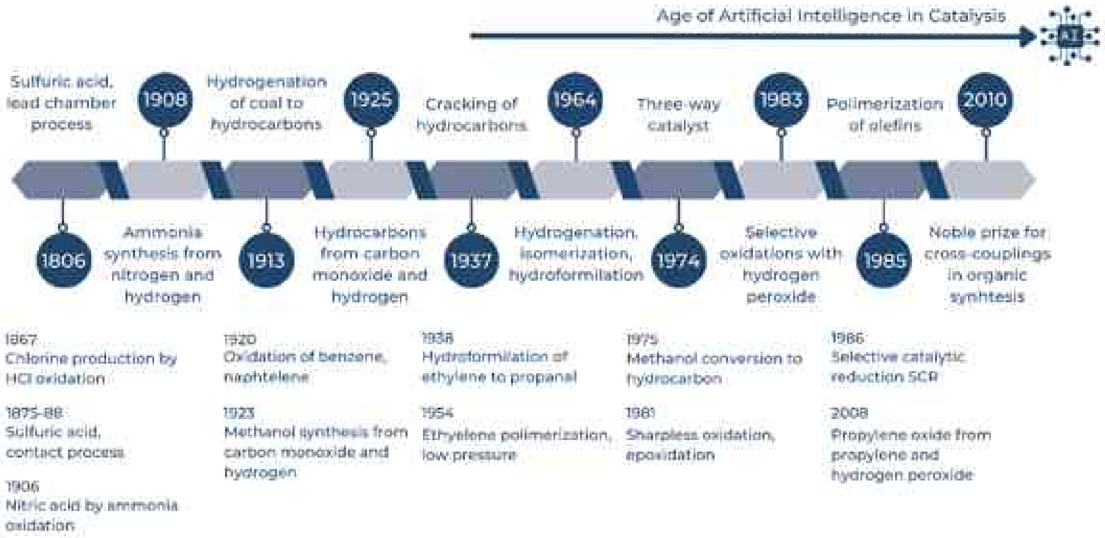


Figure 2 *Short timeline of catalysis research* [9]

The advancement of catalysis research has been on the rise for more than 200 years and many milestones can be highlighted (Figure 2)[9]. The use of catalysts has achieved outstanding results in chemical industry processes such as ammonia synthesis, methanol synthesis, polymerization processes, and the operation of motor vehicles [10–12]. The further technological development of the chemical industry is based on the latest scientific results, aimed at the production of specific, multi-component catalysts associated with extraordinary performance[13–15]. Artificial intelligence science was already present in the 1940s, but only the technological developments of the last 20–30 years have enabled the real evolution of artificial intelligence in several fields such as catalysis chemistry[16].

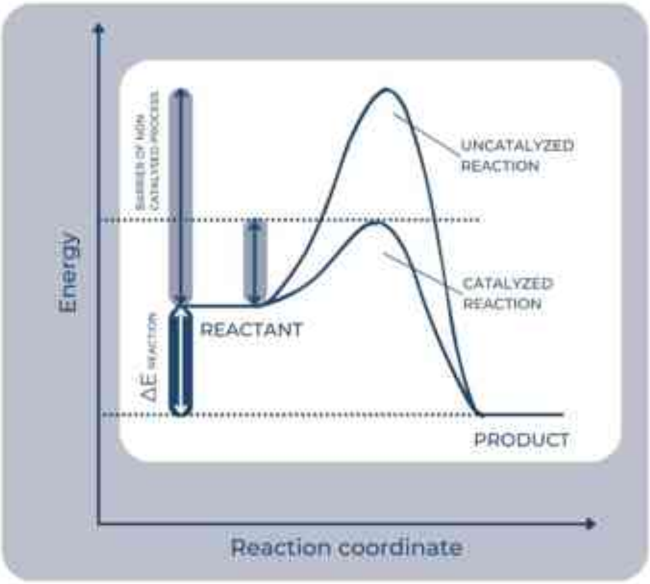


Figure 3 *Energetic presentation of catalyst role*

However, what are catalysts? Catalysts promote processes that are kinetically impossible, require extreme conditions, take infinite time, or do not occur at all (Figure 3)[9]. Catalysts reduce the activation energies of reactions, which allow to open new reaction paths and thus increase the reaction rate. However, the catalyst alters the speed of the reaction, but not its thermodynamics and not its stoichiometry. The process can be described as a catalytic cycle in which the catalyst remains "theoretically unchanged" and would not consume[9]. Indeed, it is recognized this is a much more complex process. In practice, catalytic technology can also be considered the cornerstone of green chemistry.

Many expectations are held regarding catalysts properties. In comparison to this, it seems that the number of parameters that influence them is "always one more". This could be predicted by a complex set of parameters, but the task in most cases, remains undetermined.

2.2.2. CATALYST CHARACTERISTIC FEATURES

In general, the most important properties of catalysts can be said to be given mainly by a triumvirate: activity, selectivity, and stability (Figure 4, inner circle). The importance of the three listed characteristics is indisputable, regardless of whether it is academic or industrial research. The characteristics most frequently discussed together are activity and selectivity. The question of stability, which is more interesting from the perspective of economic and sustainability issues, complicates the picture. Other parameters associated with these properties can be read in the outer circle.

Figure 4 *Most important catalyst qualification factors*[15] *(TON – Turnover Number, TOF- Turnover Frequency)*

ACTIVITY

The catalyst has a significant effect on the rate of reaction. Activity is a term used to describe the rate of a reaction. The catalyst activity is thus manifested in the fact that it significantly increases the speed of the reaction, which can be described in several ways. Table 1 highlights the most frequently used concepts for characterizing activity. The table contains the metrics used to characterize the activity, their basic calculation method, and the measurement units.

Although conversion (Table 1, 4th line) is in fact the most common catalyst performance characteristic, it must be clarified when defining it. The conversion of a reaction is described by a ratio, in which the denominator is the amount of starting substance, and the numerator is the amount of substance consumed. In order to properly operate this metric number, the conversion must specify the exact component to be monitored as well as the time conversion is connected to. The definition of instantaneous conversion as well as a total conversion is possible[17]. The calculation of the conversion also depends on whether the conversion is a batch, semi-batch or continuous process from an operational point of view[18].

Table 1 *Summary of properties describing the activity of the catalyst*[9]

NAME	EQUATIONS	UNIT
Reaction rate (r)	$\frac{\text{converted amount of substanse of a reactant}}{\text{volume or catalyst mass} * \text{time}}$	$\frac{\text{mol}}{\text{l} * \text{h}}$ or $\frac{\text{mol}}{\text{kg} * \text{h}}$
Kinetic activity (simple irreversible reaction)	$\frac{dn_A}{dt} = k * V * f(c_A)$, where n_A is molar amount of component A, k is the rate constant, and f (c_A) is a concentration term	$\frac{\text{mol}}{\text{s}}$
Space velocity	$\frac{\dot{V}_0}{m_{cat}}$, where $\dot{V}_0$ is the volume flow rate, $m_{cat}$is catalyst mass	$\frac{\text{m}^3}{\text{kg} * \text{s}}$
Conversion	$\frac{n_{A,0} - n_A}{n_{A,0}}$, where $n_{A,0}$ is starting molar amount of A component, n_A is molar amount of A component	$\frac{\text{mol}}{\text{mol}}$ or %
Space-time yield (STY)	$\frac{\text{Desired product quantity}}{\text{Catalyst volume} * \text{time}}$	$\frac{\text{mol}}{\text{l} * \text{h}}$
Turnover frequency (TOF)	$\frac{\text{volumetric rate of reaction}}{\text{number of centers/volume}}$	time^{-1}
Turnover number (TON)	TOF $*$ lifetime of the catalyst	-

Another performance feature is STY (space-time yield) extraction, which provides a basis for comparison already during the size increase, in the way of industrial application.

It is necessary to highlight the characteristics of TON and TOF in the table. Although the definitions of this metrics have long been used, they are not clear and uniform[19–22]. These concepts can be derived from the science of biocatalysis and enzyme kinetics and from there they were adopted by heterogeneous and homogeneous catalysis research[23]. The study by Kozuch et al. provides a detailed overview of these concepts and their interpretation[24]. Research efforts have raised several important questions, such as the combination of the two concepts, the relationship between TOF value and reaction concentration or the effect of circumstances, which makes these metrics difficult to determine. Kozuch et al. defined the standard TOF value, and the TON value associated with it.

SELECTIVITY

One of the 12 principles of green chemistry prefers selective catalysts instead of stoichiometric agents, since the main objective is to minimize unnecessary waste during production[25]. From a sustainability point of view, as well as an economic point of view, the use of catalysts with high selectivity is beneficial.

In fact, the selectivity is a parameter that characterizes the quality of the product formed during the reaction. Selectivity is an issue that can be approached from several angles and, therefore, can be interpreted and detailed in several ways.

According to the IUPAC the term can be used in two different ways:

"It sometimes refers to the discrimination shown by a given reactant A when it reacts with two alternative reactants B and C, or in two different ways (e.g. at two different sites) with a reactant B."[26]

"The term also sometimes refers to the ratio of products obtained from given reactants. This meaning is of importance for catalysts, which can have a wide range of selectivities. Selectivity is quantitatively expressed by ratios of rate constants for the alternative reactions, or by the decadic logarithms of such ratios."[27]

In practice, it can be said that it catalyses only one of the several thermodynamic pathways possible. Based on this, chemo-, regio-, and stereo-selectivity can be discussed[28]. In this case, the ratio of the resulting products is usually calculated. Another formula focuses on the proportion of the original material converted to the desired product[9].

Selectivity is one of the most useful parameters to measure and control during catalysts. Somorjai et al. have compiled a systematic study summarizing the molecular factors affecting catalytic selectivity in heterogeneous reactions[29]. In their research summarizing catalytic selectivity, seven molecular factors are determined in addition to reaction conditions.

One of the factors influencing catalyst selectivity is reaction conditions such as temperature, pressure, catalyst composition, or reaction type. Another group is composed of molecular factors, which are intermediate products of reaction, surface structure and composition, oxidation state, charge transport, intermediates, and induced restructuring by adsorbate.

STABILITY

Since catalyst stability has a serious economic impact, this parameter must be considered in the design of catalysts.

The lifetime of catalysts is determined by chemical, physical and thermal stability concepts[30–32]. In industrial reactions, the activity and selectivity of catalysts decrease after a certain period and are replaced. Through the examination and monitoring of these properties, the stability of the catalyst is described. Deterioration of catalyst stability can also be caused by catalyst poisoning, coking, loss of active catalyst due to evaporation or decomposition caused by overheating[33].

The concept of stability is closely related to the deactivation of catalysts. The 2015 review study by Morris et al. describes in detail, among other things, mechanisms, types, possible causes and methods of minimization of deactivation and also methods of regeneration[34]. According to their study, the cause of deactivation consists essentially of three kinds: chemical, mechanical, and thermal. Most catalyst deactivation processes can be avoided through the prevention process. Regeneration of catalysts is a process which can be carried out under certain conditions, in particular to restore the loss of activity [35–37].

Stability of catalysts is generally measured by measuring the time dependence of factors characteristic of catalyst activity. Vylder et al. designed a laboratory-scaled liquid solid (LS)2 plug-flow reactor with a fixed-bed catalyst to investigate the deactivation of heterogeneous catalysts by determining the turnover frequency[38].

LABORATORY TEST

During catalyst development, catalysts produced are tested for the first time in the laboratory. Thus, the above-mentioned performance indicators of catalysts are valid under certain experimental conditions.

Figure 5 *Most important experimental variables for laboratory development in academic and industrial research*[39]

Figure 5 summarizes the variables associated with the laboratory experiments[39,40]. Input parameters and output parameters are present. Input parameters can be independent measurable parameters, controlled parameters, or uncontrolled external parameters. Independent variables include reaction conditions such as temperature or pressure, catalyst parameters or, for example, quantity of starting materials. Output parameters are the parameters that can be analysed to characterize the performance of the catalyst.

ECONOMY AND SUSTAINABILITY

Over the past decades, the design of chemical catalysts has experienced a paradigm shift towards the priority of economy and sustainability. Heterogeneous catalysis plays a vital role in various processes, from petrochemical production, through fine chemical industry to environmental remediation[41]. Due to the increasing concern about the impact on the environment and depletion of natural resources, it is important to develop environmentally friendly and cost-effective catalysts.

Economic and sustainable considerations in catalyst design include optimizing catalyst efficiency, selectivity and stability to minimize raw materials consumption and maximize product yield[42–44]. Various catalyst cost estimation programmes or guidelines have already been developed, indicating the economy[45–48].

Furthermore, sustainable efforts include minimising waste generation and the use of environmentally friendly materials, including nanomaterials that enhance catalyst activity[49–51]. These developments are consistent with efforts to combat climate change through carbon capture and use and the production of renewable energy.

2.2.3. CLASSIFICATION OF CATALYSTS

Catalysts can be classified according to several classification criteria. They can also be classified based on state, structure, composition, or application. In Figure 6, the different clustering of homogeneous and heterogeneous catalyst was emphasized. On some points, the groups found in the literature differ as well as depending on the catalysts for which the categorization of the reaction group contains[52–56]. The classification of catalysts provides the basis for building a database, the central element of which can be the grouping of the catalyst according to different aspects and the examination of the catalytic performance of the groups formed in this way.

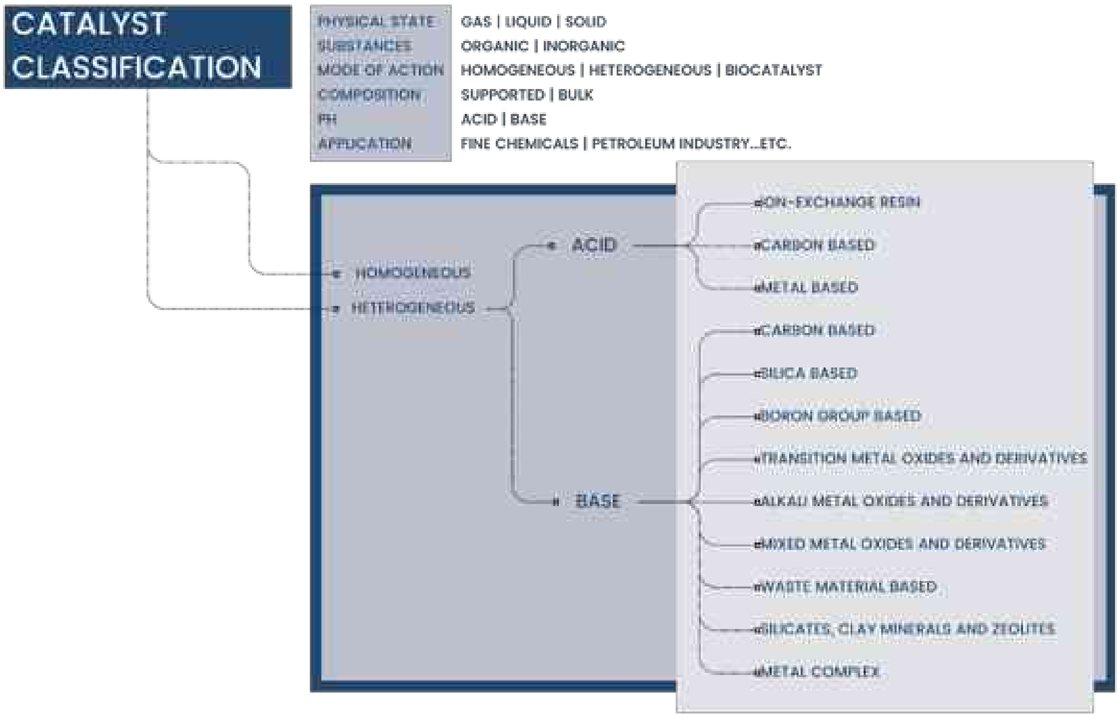


Figure 6 *Classification of catalysts*[52–54]

Homogeneous and heterogeneous catalysis are two distinct types of catalytic processes that differ in the phase of the catalyst relative to the reactant. In heterogeneous catalysis, catalyst and reaction agents are in different stages[57]. In heterogeneous reactions, compared to homogeneous catalysis, reactions are performed at higher temperatures and at higher reaction rates, with lower selectiveness. However, due to the simple separation and recyclability of catalysts, it is used more frequently in industry than in homogeneous catalysis.

Although heterogeneous catalysis research is promising and essential for industrial applications, there are some challenging aspects faced by researchers[58]. Heterogeneous catalysts often have complex structures and nanoscale active sites, requiring complex analytical methods. The analysis and characterization of catalysts under industrial reaction conditions can be particularly difficult because these conditions differ significantly from normal laboratory conditions. The catalyst design itself is a very time-consuming and energy-consuming process. The development of catalysts to maximize their activity, selectivity and stability requires deep knowledge of material science, surface chemistry, reaction kinetics and computational chemistry. It is difficult to determine the exact reaction mechanisms on heterogeneous catalyst surfaces due to the transitional and dynamic properties of the adsorbed species. Heterogeneous catalysts may deactivate over time due to factors such as contamination, poisoning or decomposition, leading to reduce in catalyst activity and lifetime[59]. Furthermore, mass transfer limits can hinder the access of the catalysts to the active site and affect the overall performance of the catalyst. Another difficulty and challenge at the same time is the issue of scale-up, reproducibility, recovery process, real-time monitoring and so on.

Despite all challenges, research efforts continued with the support of advanced experimental techniques and computational methods to understand heterogeneous catalysis and to continuous innovations in chemical industry.

2.3. THE ERA OF BIG DATA IN CHEMISTRY

Scientific research has undergone major changes in recent years as data growth has increased exponentially. The amount of data in scientific research is growing rapidly, which brings both benefits and challenges. On the one hand, this amount of data allows researchers to make new discoveries and better understand things. For example, in field like new energy power and energy storage, and chemo and bioinformatics or management system, big data has led to significant progress[60–62].

However, it is not easy to handle such a large amount of data. Researchers have difficulty managing, processing, and analysing all data and information. Sometimes, they cannot use all the data they have. Furthermore, sharing data between different research groups can be difficult due to different formats, rules, and encryption.

Over the past few decades, the number of published articles in the field of scientific publications has increased exponentially, which constitute the source of data for various disciplines[63]. Although this level of growth in scientific literature represents progress, it also poses challenges in terms of access and overload of information.

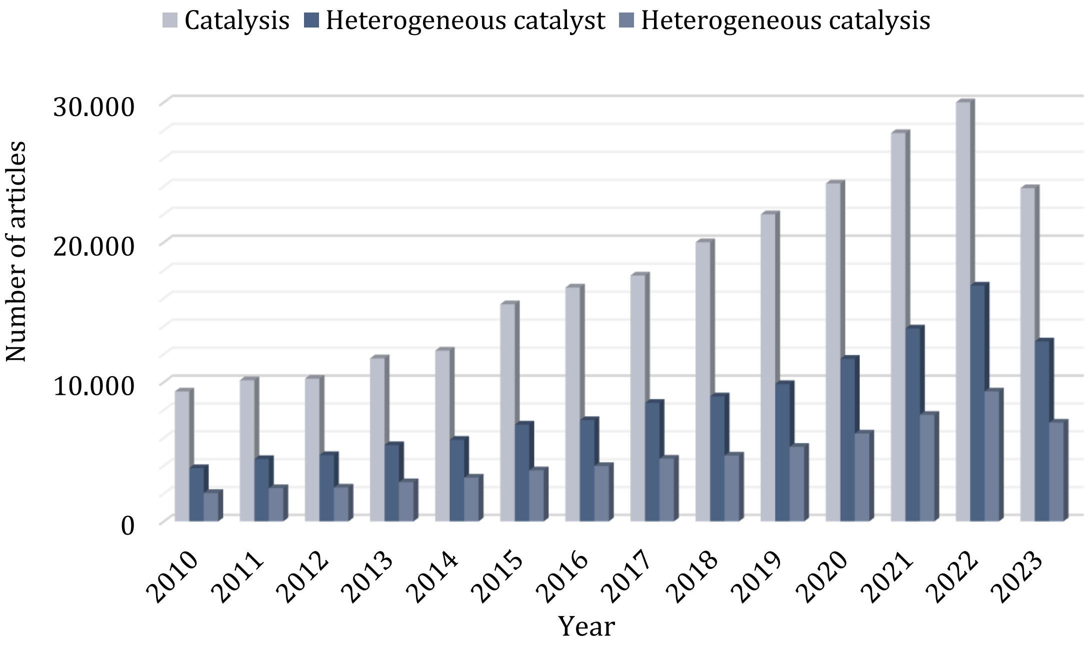


Figure 7 *Change in the number of publications over the past period [searching criteria: catalysis, heterogeneous catalyst and heterogeneous catalysis keywords, 2010-2023, review articles and research articles]*[64]

The increase in scientific publications is due to several factors. Technological advances and digitization have made it easier for researchers to share their findings with a global audience. Free-access journals are becoming more common, enabling the rapid dissemination of research results. Furthermore, inter-disciplinary research and international cooperation have contributed to a wide range of published works.

ScienceDirect is a leading source of scientific, technical, and medical research in the world. Based on the keyword searchers, the database contains more than 1 million research articles and reviews in the field of chemistry and approximately half a million in the field of catalysis[64]. Figure 7 illustrates the scientific trend in publications over the past few years based on the defined keywords, such as 'heterogenous catalysis'. By contrast, while doctoral students can read and process hundreds of scientific articles during four-year doctoral education, thousands of new scientific publications are produced every year on heterogeneous catalysis.

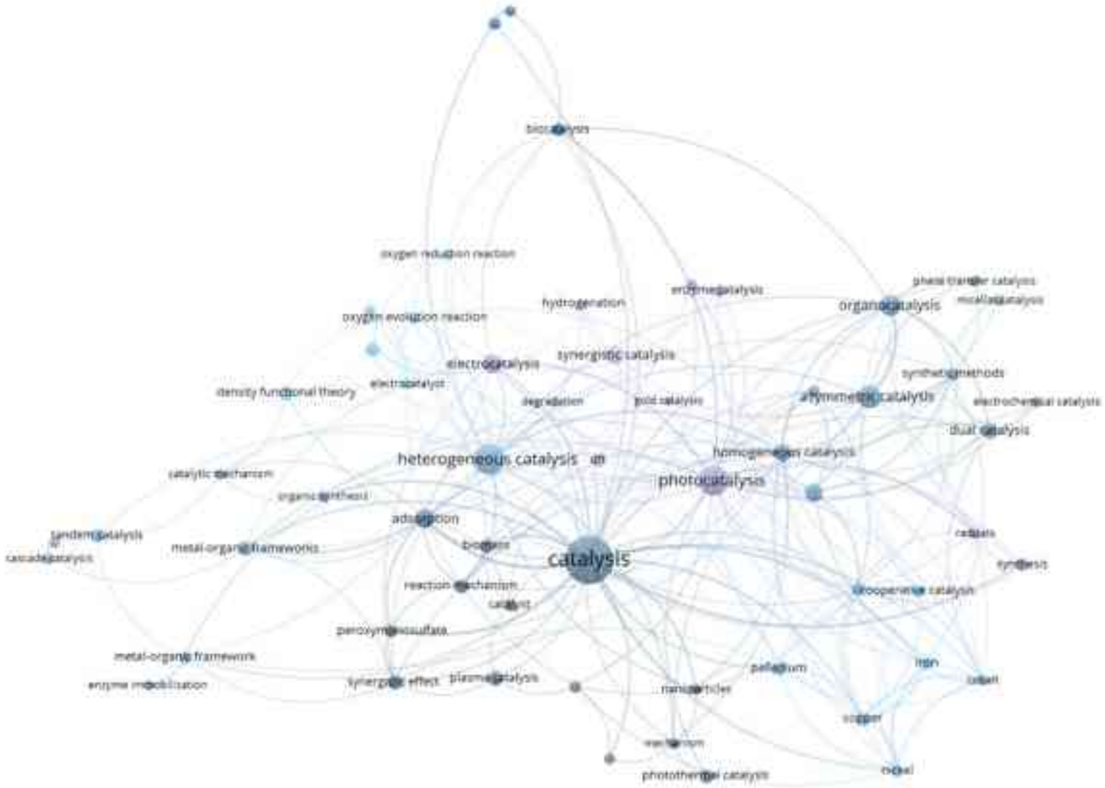


Figure 8 *Visualization of bibliometric networks in case of catalysis keyword*[65] *[searching criteria: catalysis keyword, 2010-2023, relevance, first 200 articles]*

Science of chemical catalysis is continuously evolving, researchers explore new boundaries, and respond to critical challenges. Figure 8 presents a bibliometric network of catalysis based on the most relevant 2000 articles from 2010 to 2023[64,65]. The objective of visualizing the bibliographic network is to provide a clear and comprehensive representation of the relationship between scientific publications, authors, or research areas[66]. The different colours represent the connection point between terms and clusters generated by the *VOSviewer* software program. Based on visual data processing, recent research focuses on photocatalysis, heterogeneous catalysis, organocatalysis, homogeneous and asymmetric catalysis. From this figure, the most studied topics in the field of heterogeneous catalysis are metal-organic frameworks, cooperative catalysis, electrocatalysis, synergistic effects and reaction mechanisms. Density Functional Theory (DFT) appears among the keywords as a theoretical method for the study of chemical reactions, and palladium also appears, as one of the most used noble metals in heterogeneous catalysis.

2.4. DATA ANALYSIS IN CATALYSIS

The focus of this doctoral work is to collect, systematize, analyse, and interpret data accumulated during catalysis research, which is difficult to use directly to support the development of catalysts. Catalysis plays an important role in accelerating chemical reactions and has a major impact on various industrial processes, energy production and environmental protection. As the field of catalysis evolves, the importance of data analysis to understand the complex mechanism of catalytic processes becomes increasingly apparent[67].

In modern catalytic research, many data are collected from experiments, simulations, and theoretical calculations. Data analysis enables researchers to extract valuable information from these high-dimensional data sets and reveal patterns, relationships and trends that would otherwise remain hidden[68–70].

Data analysis plays an important role in identifying the active sites of the catalyst surface. The relationship and interpretation of activity data with structural and composition information provides insights into the structure-activity relationship of catalysts[71,72]. It can also be an important step for researchers in the field of kinetic analysis and reaction mechanism. Through data analysis, experimental data can be combined with kinetic models, to determine activation energies and identify reaction transition states[73,74]. This knowledge is very important in the design of practical catalysts and in the development of more efficient selective catalysts.

In addition to many other possibilities of application, the age of big data has an important impact on catalysis research. Machine learning algorithms are capable of processing large datasets, recognizing hidden patterns, and generating prediction. In catalysis, machine learning technology is used to find new catalysts, predict catalytic performance, and optimize reaction conditions. These data-driven approaches can accelerate catalyst research and facilitate the discovery of new catalytic systems[16].

2.5. MACHINE LEARNING IN CATALYSIS: A SYNERGISTIC PARTNERSHIP

Machine learning (ML) is a field of artificial intelligence that enables computers to learn from experience (with mathematical data models) and improve their performance continuously without the need for special programming[75]. Algorithms have been developed to search for patterns and rules in large data sets, to predict, decide and solve problems.

Machine learning can be seen as the latest tool in catalytic research, as it provides researchers with the opportunity to effectively model and interpret modern catalytic systems containing many variables and complexity[16,76–79]. These studies build algorithms from experimental data or literature data using mathematical models to predict processes through ML and gain new knowledge of certain processes.

The more commonly used term is catalysis informatics, an interdisciplinary field that lies at the boundary between catalysis and informatics. The main goal of this field is to promote and accelerate the research, development and optimization of catalytic processes using information technology methods and technologies[80,81].

Information technology allows modelling, simulating, speeding up quantum chemical simulations, and collecting data on catalytic reactions. With computer tools, data can be extracted and analysed, which helps relationships, and correlations related to catalytic processes. This enables a better understanding of the correlation between catalytic systems, reaction mechanisms and the properties of the catalytic process describing the performance of the catalyst.

2.6. DATA PROCESSING IN LITERATURE RESEARCH: COMPARISON OF CATALYSTS

The process of designing catalysts also requires literature research, in most cases involving catalyst mapping, which is developed and published by different research groups.

Information literature research is superficial, resulting in a large amount of data available, but it is difficult to see by us more. A deeper literature search, which already deals with processing and interpretation of data, helps researchers to understand more complex relationships.

However, in this regard, comparison of catalyst in catalysis research is a challenge, as many factors make it difficult to handle data properly.

The first problem is the lack of data unification. Various research groups use different methods and conditions to examine catalysts and test them in catalytic reactions. Different data collection methods, reaction conditions and properties make it difficult to compare. In addition to different measurement methods, the quantitative and qualitative characteristics of catalysts are not standardized. Since catalysts are usually complex compounds, and their effects on catalytic systems are numerous, it is not trivial to interpret and compare data. The specificity of the reaction of the catalyst makes comparison even more complicated. The process and performance of catalyst are also influenced by environmental factors such as temperature, pressure, and type of reaction. The inclusion of such variables makes the comparison task even more difficult. Not to mention that many data are still in the dark because of possible encryption of innovation processes.

Taking all these into consideration, thorough processing and understanding of the data collected during literary research create the opportunity to accelerate innovative studies.

2.7. OBJECTIVE OF THE DOCTORAL STUDY

Figure 9 *From data to knowledge – schematic of PhD study*

My doctoral thesis summarizes my research in the field of data-based catalyst design and contributes to new methods of catalyst research. The goal is to build a hierarchy of data-information-knowledge from the data point through various methods and achieve catalyst design strategies in advance (Figure 9). The main objective of catalyst design is to optimize catalyst composition through analysing literature and experimental data, using various mathematical models and computer software to predict and apply catalyst composition.

During literature research, the structure of specialized literature publications, how research results are discussed, and the data content of publications became known. It was determined which parameters are useful for extracting information from publications and which are appropriate for the characterization of catalysts.

The literature contains inconsistent data that are difficult to compare each other directly. Since data sets are difficult to handle in this way, in collaboration to the University of Miskolc Catalyst Group, a database was created and then standardized with the MIRA21 model into a single quantifiable data per catalyst. The catalysts have thus become rankable and classable. In the next step of information collection, data were cleaned up and parameters selected, and the correlation between parameters and factors influencing the composition of optimal catalysts were investigated. As the process of exploring data analysis continued, data appeared to be very well used by artificial intelligence, especially in machine learning. Thus, catalyst design guidelines and machine learning data sets were created as a result of data analysis processes before machine learning.

Throughout my doctoral studies, new knowledge was gained in the field of catalysis informatics, which helps to promote the use of artificial intelligence in the design of catalysts. Given our considerable accumulation of knowledge on the semantic aspects of publications based on human learning, this research also forms a solid basis for utilizing the potential of semantic searches.

2.8. THE CHOSEN TEST REACTIONS

The first catalyst design reactions are the catalytic hydrogenation of nitro-aromatic compounds such as nitrobenzene and 2,4-dinitrotoluene, known chemical reactions with important industrial applications.

Aromatic nitrogen compounds are widely used in explosives, pesticides, fertilizers, dyes, pharmaceuticals, plastics, resin, and fuel additives. The growing demand for these industrial sectors has an impact on the market for nitrobenzene, especially nitrobenzene, which was estimated at $10.38 billion in 2023[82].

The hydrogenation of aromatic nitrates has been widely studied[83–86]. Nitrobenzene is the raw material for the production of nitric acid[87]. The reduction of nitrobenzene by Bechamp is the oldest technology of aniline synthesis, which uses iron in the presence of hydrochloride, while modern industrial aniline production processes are performed by catalytic hydrogenation of nitrobenzene in gaseous or liquid phases and in the presence of metal catalysts[88]. The following reaction equation shows the general reaction of the hydrogenation of nitrobenzene to alanine (Figure 10).

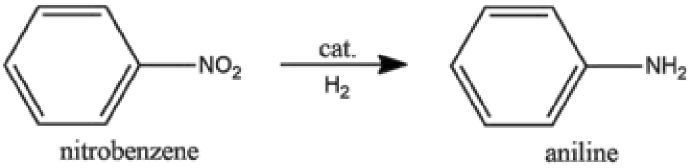


***Figure 10** General reaction of nitrobenzene to aniline by hydrogenation*

Toluene-diisocyanate is produced in a continuous process of three steps (Figure 11). Dinitrotoluene is produced in the first step by toluene nitration. The second and decisive step is the catalytic hydrogenation of dinitrotoluene into toluene diamine. In the last step, toluene diamine forms toluene diisocyanate.

The second stage of the industrial process is catalyst hydrogenation of dinitrotoluene to toluene diamine using high-pressure and high-temperature solid catalysts (100-150°C, 5-8 bar). This step was previously done with iron fillers and water hydrochloric acid but is hydrogenated today using Ra-Ni or Pd/C catalysts[89]. Figure 11 shows the general reaction equation with the main product of the hydrogenation of 2,4-dinitrotoluene.

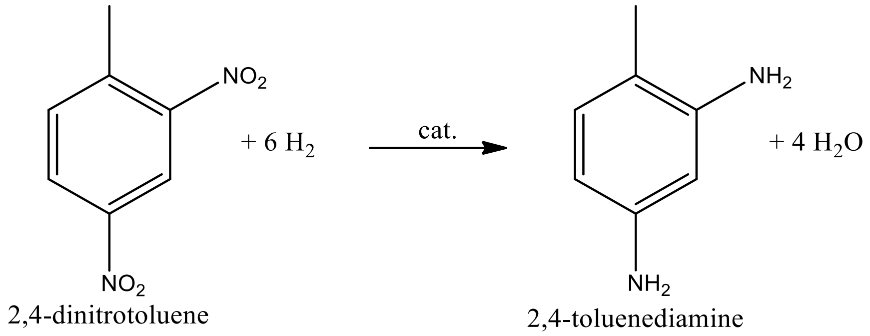


Figure 11 *General equation of dinitrotoluene hydrogenation with the main 2,4 isomer*

Since my doctoral dissertation contains a complete literature survey and data analysis of these two reactions, it is not discussed in detail in this paragraph.

3. METHODS

3.1. MIRA MODEL

The Miskolc Ranking 2021 (MIRA21) methodology is a multi-step process for comparing catalysts employed in a specific reaction and identifying new patterns among parameters characterizing catalytic processes. This is a systematic approach to create a simple, general parameter from parameter vectors for the comparison and analysis of catalysts data.

3.1.1. METHODOLOGY

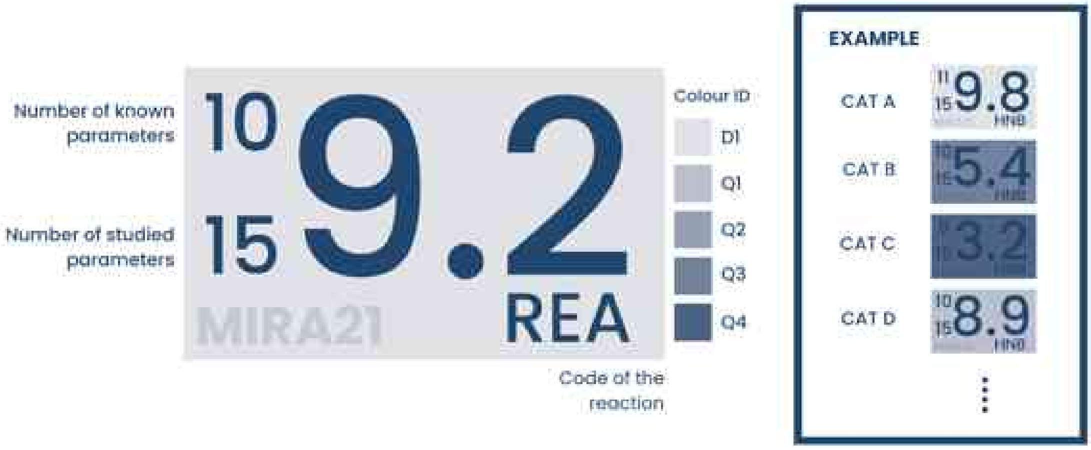
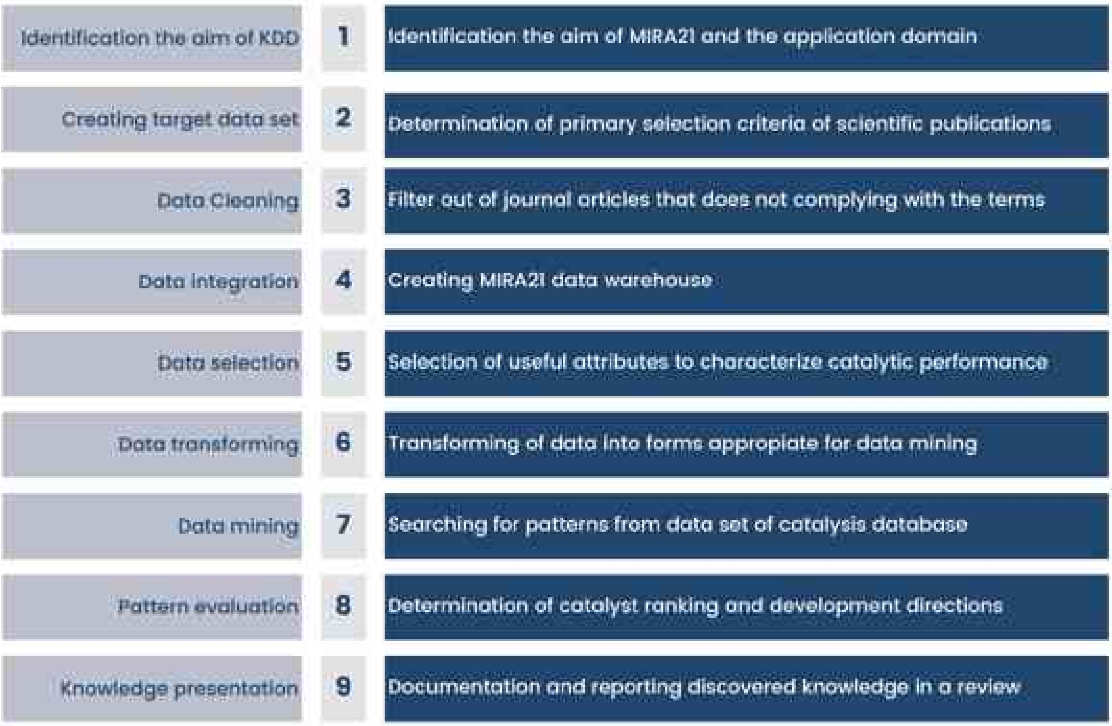


Figure 12 *Visualization of quantitative description of a catalyst by MIRA21 number with corresponding classification and colour code: D1 (top10%), Q1(0-25%), Q2(25-50%), Q3(50-75%), Q4(75-100%), the deepening of the colour indicates the rank*

Figure 12 shows the formation of MIRA21 number assigned to each catalyst tested. Furthermore, this number provides two indexes. The subscript represents the number of attributes that were studied (number of parameters studied) and the superscript indicates the number of attributes available in the scientific publication (the number of known parameters). These two parameters provide additional information on the detail level of published data available. If only a few known parameters are available in the publication, the characterization of the catalyst according to the MIRA21 model is less robust. The acronym HNB (**H**ydrogenation of **N**itro**B**enzene) shows the type of the catalytic reaction. The D1 classifies corresponds to the top 10% of the catalyst. Quartiles are also defined, using four classes: Q1 (first), Q2 (second), Q3 (third), Q4 (fourth).



Identification the aim of KDD	1	Identification the aim of MIRA21 and the application domain
Creating target data set	2	Determination of primary selection criteria of scientific publications
Data Cleaning	3	Filter out of journal articles that does not complying with the terms
Data integration	4	Creating MIRA21 data warehouse
Data selection	5	Selection of useful attributes to characterize catalytic performance
Data transforming	6	Transforming of data into forms appropiate for data mining
Data mining	7	Searching for patterns from data set of catalysis database
Pattern evaluation	8	Determination of catalyst ranking and development directions
Knowledge presentation	9	Documentation and reporting discovered knowledge in a review

Figure 13 *MIRA21 methodology through KDD steps*[90]

The establishment of MIRA21 methodology based on the so-called Knowledge Discovery in Databases (KKD) process[90]. KDD is a methodological process used in data extraction and machine learning to extract valuable and previously unknown information or patterns from large-scale data sets. Figure 13 demonstrates the methodology of the MIRA21 model according to KDD process.

The main purpose of the MIRA21 model is to provide a standard for assessing the “goodness” of a catalyst with objective numerical data and to compare and classify it accordingly. The classification promotes the efficient selection of appropriate links relevant to support the design of a new catalyst or improve existing ones. The comparison of special catalysts for a reaction allows the monitoring of trends in research and development. Standardization of access data in MIRA21 will also promote accurate and consistent data in future publications.

The application field of the model has been reduced to catalytic reactions, mainly heterogeneous catalytic reactions. This methodology was developed using the hydrogenation reaction of aromatic nitrogen compounds described above.

The literature sources determine the validity and reliability of research. When choosing literature, reliance on the sources written by recognized authors, published in high-quality journals, and used in the past to obtain reliable information is worthwhile.

Figure 14 *Selection criteria*[21,91]

Figure 14 summarizes the three steps of selecting the processed scientific articles. The primary criteria were determined based on the quartiles of the journals. The value of Q index was examined based on the latest results of the Scimago Journal&Country Rank[92]. As a priority, the research results of journal that received Q1 and Q2 certification were accepted. The second criteria for selecting are the date of publications, since new research is usually related to current scientific and technological developments, which helps to keep up with the latest discoveries and promote the process of innovation. Afterwards, a rapid informational analysis of literature is also required, resulting in articles that have a strong lack of data or unrelated content being filtered.

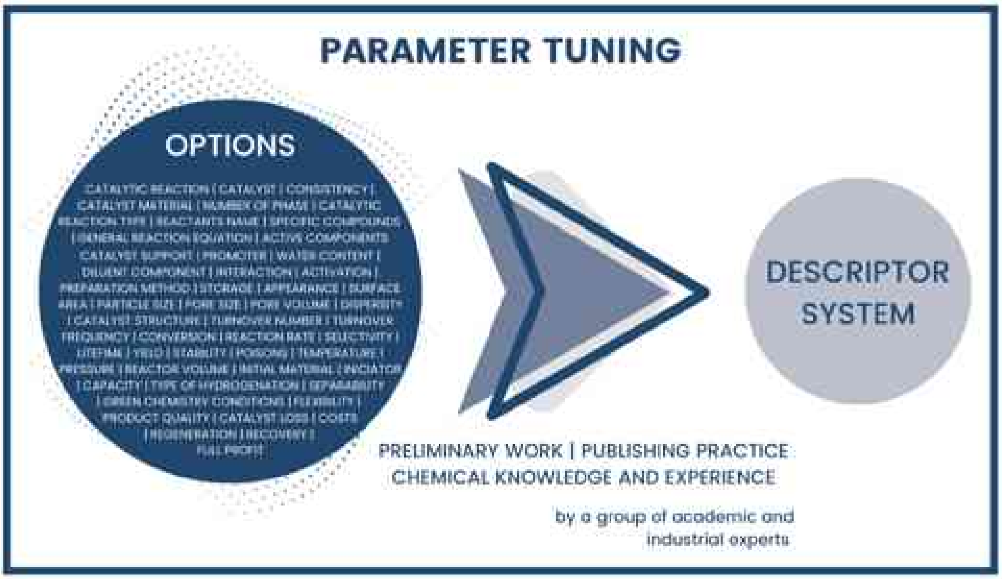


Figure 15 *Establishment of descriptor system by a critical rigorous method*

It is quite difficult to find suitable descriptors for the catalytic process or catalyst, as the activity of solid catalysts is a function of many variables. Descriptors are used in several field in different senses[93–95]. In this case, they refer to concepts or characteristics that describe, identify or mark catalyst. The design of the descriptor system was preceded by the compilation of the initial list of parameters (Figure 15). The second pillar is literature research practice, because publication habits clearly limit the types of descriptions to be used. The final element of the process is to select a system composed of 15 parameters based on the professional knowledge and experience of the Chemical Institute's Catalytic Hydrogenation Group.

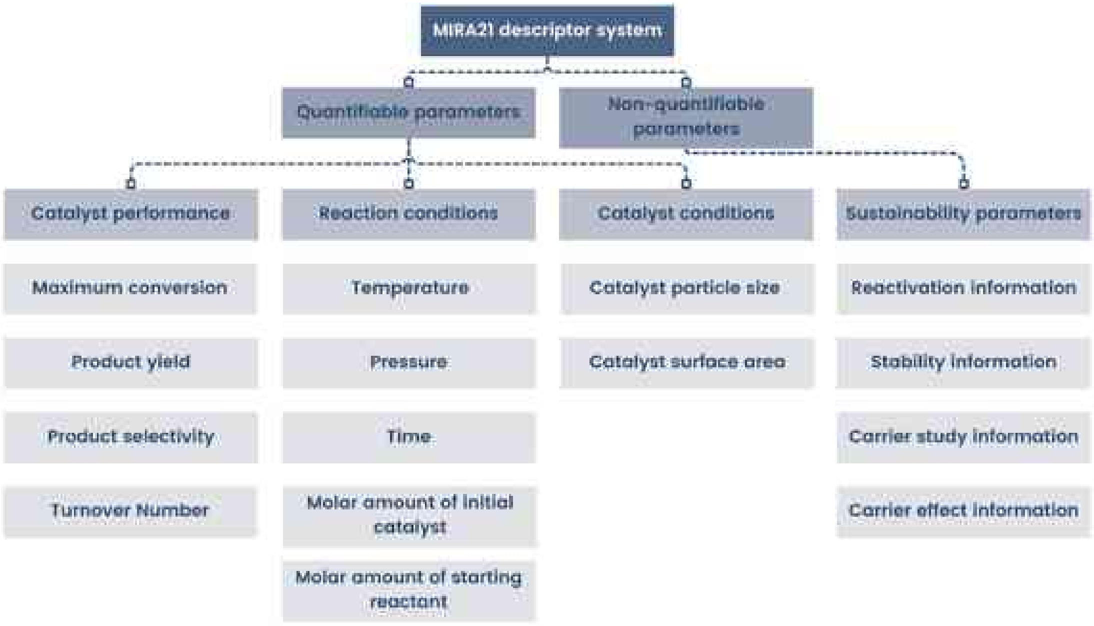


Figure 16 *Classification a hierarchy of parameters and descriptors*

The model descriptor system can be divided into 4 classes as shown in Figure 16. The quantifiable classes are the performance of the catalyst, the reaction conditions, and the catalyst conditions. Sustainability parameters include the non-quantifiable class.

During the development of the methodology, the independence of parameters was not evaluated. Table 2 demonstrates in detail the descriptor system of the 15 parameters, with their notation, unit, and definition.

Table 2 *Descriptor system of MIRA21 model for heterogeneous catalytic reactions*

Descriptor system							
Categories			**No.**	**Notation**	**Name**	**Unit**	**Definition**
Quantifiable parameters	Catalyst performance	I.	1.	X_{REAmax}	Maximum conversion	n/n%	Maximum reactants conversion achieved on a given catalyst
			2.	Y_{PR}	Product Yield	n/n%	Product yield for maximum conversion
			3.	S_{PR}	Product Selectivity	n/n%	Product selectivity for maximum conversion
			4.	TON_{PR}	Turn Over Number	-	Number of moles of product formed per 1 mol active metal when the maximum conversion reached
	Reaction conditions	II.	5.	$Tmax_{conv.}$	Temperature	K	Reaction temperature for maximum conversion
			6.	$P_{max.conv.}$	Pressure	atm	Reaction pressure for maximum conversion
			7.	$t_{max.conv.}$	Time	min	Time required to reach maximum conversion
			8.	$n_{cat.}$	Molar amount of initial catalyst	mol	The molar amount of the active metal involved in the reaction - in case of several metals, the sum of molar numbers
			9.	n_{start}	Molar amount of starting reactant	mol	The initial amount of starting reagent involved in the reaction
	Catalyst conditions	III.	10.	CPS	Catalyst Particle Size	nm	Average particle size of the catalyst
			11.	CSA	Catalyst Surface Area	m^2/g	Catalyst (active metal + support) surface area
	Does the publication contain information about these subjects?						
Non-quantifiable parameters	Sustainability parameters	IV.	12.	Rea	Information about Reactivation	-	Reactivation means the physical process by which the activity of the catalyst used returns to or near the original activity level.
			13.	Stab	Information about stability of catalyst	-	Stability means preservation of catalytic activity
			14.	Care	Information about catalyst carrier effect	-	Carrier effect means that the catalyst support influences the catalytic reaction
			15.		Catalyst carrier effect	-	Nature of the effect (positive, no effect, negative)

The maximum conversion, X_{PRmax} obtains the highest priority in the data analysis. All additional data depend on the conversion. The determination of the maximum conversion is based on the following equation (Eq. (1)):

$$\mathrm{X_{REAmax}} = \frac{\text{consumed } \mathrm{n}_{reactant}}{\text{initial } \mathrm{n}_{reactant}} * 100 \tag{1}$$

The following equation was used to calculate the yield of the product, Y_{PR} (Eq. (2)):

$$\mathrm{Y}_{PR} = \frac{\text{synthetized } \mathrm{n}_{dproduct}}{\text{theoretical } \mathrm{n}_{dproduct}} * 100 \tag{2}$$

The product selectivity, S_{PR} of the catalyst was calculated as follows (Eq. (3)):

$$\mathrm{S}_{PR} = \frac{\text{synthetized } \mathrm{n}_{dproduct}}{\text{consumed } \mathrm{n}_{dproduct}} * 100 \tag{3}$$

Where $\mathrm{n}_{dproduct}$ the molar amount of the desired product, and $\mathrm{n}_{reactant}$ the molar amount of the reactant.

In general, the conversion, yield, and selectivity necessary to describe catalytic performance can be found through literature research. Publications usually contain only some of these three data or pair data. Therefore, using all three attributes in ranking even though their independence was not examined.

Several publications have been published regarding the explanation of TOF (turnover frequency) and TON (turnover number)[90,96,97]. The study of Kozuch et al. wrote a detailed discussion about the definition and application of these metric numbers and use of TOF in connection of heterogeneous catalysis[97].

Although Boduart first defined the frequency of rotation in the 1960s, the concepts of TOF and TON numbers in heterogeneous catalysis are still different[98]. The experience of literacy has shown that these metric numbers are used in a variety of ways to communicate the results of scientific research. Most of the data in articles were found as the TON number.

To standardize the data, TON was determined according to the following definition: “turnover number specifies the maximum use that can be made of a catalyst for a special reaction under defined reaction conditions by the number of molecular reactions or reaction cycles occurring at the reactive centre up to the decay of activity”[99].

According to this explanation, the following equation (Eq. (4)) was used to calculate the Turnover number, $\mathrm{TON_{PR}}$:

$$\mathrm{TON}_{PR} = \frac{\text{synthetized } \mathrm{n}_{dproduct}}{\mathrm{n}_{\mathrm{catalyst}}} \tag{4}$$

where $\mathrm{n_{catalyst}}$ is the corresponding molar amount of catalytically active metal.

It was found catalysts tested in a tube reactor. In cases where the residence time could not clearly determine, data was uniformly recorded for 60 minutes. These cases are marked “*” in the data set.

The second class consists of data about the reaction conditions: reaction temperature, reaction pressure, reaction time to reach maximum conversion, the molar amount of the catalyst, and the molar amount of the reactant. In the industrial process, the hydrogenation reactions are performed at high temperatures and pressures. However, in the MIRA21 model, temperature and pressure were scaled and measured from a thermodynamic economics point of view. The higher temperature and pressure, the more expensive the reaction from an energy point of view is.

If more than one maximum conversion data point is defined, the second priority is the reaction time. The calculation of the molar number of the catalyst was defined as the molar number of the active metal involved in the reaction. If more than two active metals are used, the total catalyst quantity is the sum of the molar quantity.

The third class of characteristics is the particle size and the specific surface area of the catalyst. These parameters are important to note because the surface of the catalyst and the support differ, and they apply to the prepared catalyst.

Normally, the size of the particles and the specific surface area are clearly correlated[100–102]. The optimal surface that the reactant can contact varies depending on the type of the reaction. Consequently, if the reaction agent is inaccessible, it is necessary to increase the specific surface are. For this reason, both properties were included in the descriptor system. On the other hand, the size of particles can be decisive parameter from an industrial point of view, not only for the reaction but also for the separation technique. The distribution of particle size is another key parameter in case of catalyst characterization[103–105]. However, the practice of publishing has shown that the distribution of particle sizes is not standardized and rarely described by researchers, so this parameter has been omitted from the system.

The last class of the descriptors contains non-quantifiable parameters. The objective of the descriptor group is to investigate catalyst from the sustainability point of view. Almost all catalysts studied were self-made and are therefore not commercially available. In the evaluation of the appropriateness of a catalyst, economic and environmental sustainability considerations also arise. Although there may be several indicators of stability, the descriptors are determined by the data available in the publications. Data on catalyst reactivation methods, catalyst stability, and catalyst carrier effect were included.

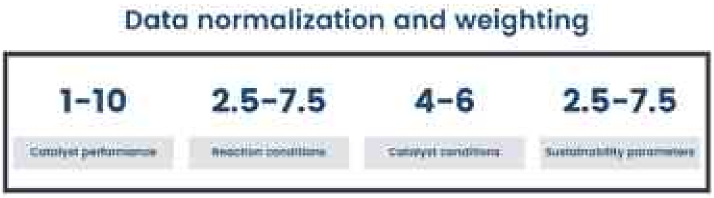


Figure 17 *Initial weighting system of MIRA21 model*

The collected data must be converted into appropriate forms for data mining. Our model applies data normalization and weighting techniques. In the first step, the initial weights of the four classes were determined (Figure 17). The purpose of the weighting method is to provide a usable numerical value that also distinguishes the four classes with minimal distortion.

The first class characterizes the catalytic performance in a scale of 1 to 10, and 10 is the highest performance. The second type of parameters for the reaction conditions are the temperature, pressure, time, and quantity of the initial materials. They considered their 50% less weight than the first class. The third class consists of the physical parameters of the catalyst. These parameters have 80 per cent less weight than the first class. The fourth category deals with sustainability. Most scientific publications do not include studies on sustainability, even though sustainability is one of the most pressing issues of our time. This is why this category has a greater weight than the third, which supports the importance of sustainability.

Non-quantifiable data were scored at 2.5 or 7.5 points, except carrier effects. In case of carrier effects, the nature of the effect was also examined. If the studied catalyst carrier had a positive effect on catalytic performance, it got 7.5 points. If the catalyst support has no effect on the reaction, it received 5 points. The catalyst that negatively affected the hydrogenation reaction, received 2.5 points.

Frequently, the values of conversion, yield, and selectivity were given inaccurately. Authors often use the context of “above 99%” or “more than 99%” to characterize catalytic performance. Therefore, there is no difference between 99% and 100% and a maximum score were given to conversions for values of 99% or more.

The following equation was used to normalize the data from quantifiable parameters (Eq. (5)):

$$\mathrm{A^t} = \mathrm{MIN} + \frac{(\mathrm{MAX} - \mathrm{MIN}) * \mathrm{A} - \mathrm{min_A}}{\mathrm{max_A} - \mathrm{min_A}} \tag{5}$$

where A is the value of the attribute, A^t is the transformed attribute value,min_A and max_A are the corresponding calculated minimum and maximum values of the attribute in the data set, MIN is the minimum scoring point and MAX is the maximum scoring point. As the data sets change, the minimum and maximum values of the specified attributes also change, meaning that the scale depends on the current data set. Outliers can also significantly affect the scaling and scoring system.

The normalized data was summed in a multiplication function, and the logarithm to the base ten of the resulting sum was taken. The following equation shows the formulation of the MIRA21 number (Eq. (6)):

$$\mathrm{MIRA21} = \log \prod_{\mathrm{i}=1}^{\mathrm{n}} \mathrm{A_i^t} \tag{6}$$

where i=1…15 the number of attributes, and A^t is the transformed value of an attribute between the corresponding scores. The value can be specified with a decimal point due to the logarithmic calculation. However, the second decimal point was also indicated in the tables of ranked catalysts because of ranking clarification.

Each catalyst can be characterized by MIRA21 having a maximum value of 13.43. The ranking is based on the calculated MIRA21 numbers. In addition to establishing the ranking, the results were divided into quantiles. The upper limit of the D1 class score was calculated by the following equation (Eq. (7):

$$\mathrm{ScorelimitofD1class} = \mathrm{MAXrank} - \left(\frac{\mathrm{MAXrank} - \mathrm{MINrank}}{10}\right) \tag{7}$$

where $MAXrank$ is the highest and $MINrank$ is the lowest score of MIRA21 ranking. Quartiles were also defined according to this principle, wherein the first decile and four quartiles: D1, Q1, Q2, Q3, Q4 were used. The first quartile is made up of Q1 and the top 10% (D1).

3.1.2. DATABASE DESIGN

Data processing was performed in Microsoft Excel from the data collection to the data normalization and evaluation.[106] (Appendix 1). In Excel, a separate spreadsheet was created for the descriptor system, where you can change the parameter weight. Data storage of journal articles, data storage of catalyst composition, raw data collection based on MIRA21 model, determination of data normalization, calculation of MIRA21 number, ranking, classification, and evaluation are performed on a separate spreadsheet.

During the processing of data, three types of data were generated. The first data type provides explicit data about the attribute called available data. The second is extracted data, which can be calculated from known information. The third is the graphical data called the readable data. Derivative and readable data are also marked in the data set. Calculations were made in a predetermined way. Origin was used to read the figures of journal articles[107].

Although a descriptor system is available, reading, and interpreting literature is often subjective because the researchers themselves have different backgrounds, interests, and experiences. In other words, the scientific results described can be interpreted differently depending on the context of the text and focus on different aspects. Therefore, it is thought that three curators work together during data processing, checking each other, comparing results, and discussing the points concerned. In the event of conflicting data, the entry of data with the participation of several researchers were occurred.

The application and review of the MIRA21 model methodology can be found in the results and discussion section of the doctoral thesis.

3.2. EXPLORATORY DATA ANALYSIS

The aim of the study is also to investigate how to obtain conscious data analysis from the collection of data guided by chemical intuition and to obtain results suitable for the construction of machine learning algorithms.

Exploratory data analysis (EDA) is the application of several statistical techniques aimed at investigating, describing, and summarizing the nature of data[108]. This allows us to identify possible errors, reveal the existence of an outlier, check the relationship between variables (correlations) and their possible redundancy, and conduct a descriptive analysis of data using graphical representations and summaries of the most important aspects[109]. EDA of previous catalytic data reveals the exploration of correlations between the physicochemical properties and performance of catalysts[110–116].

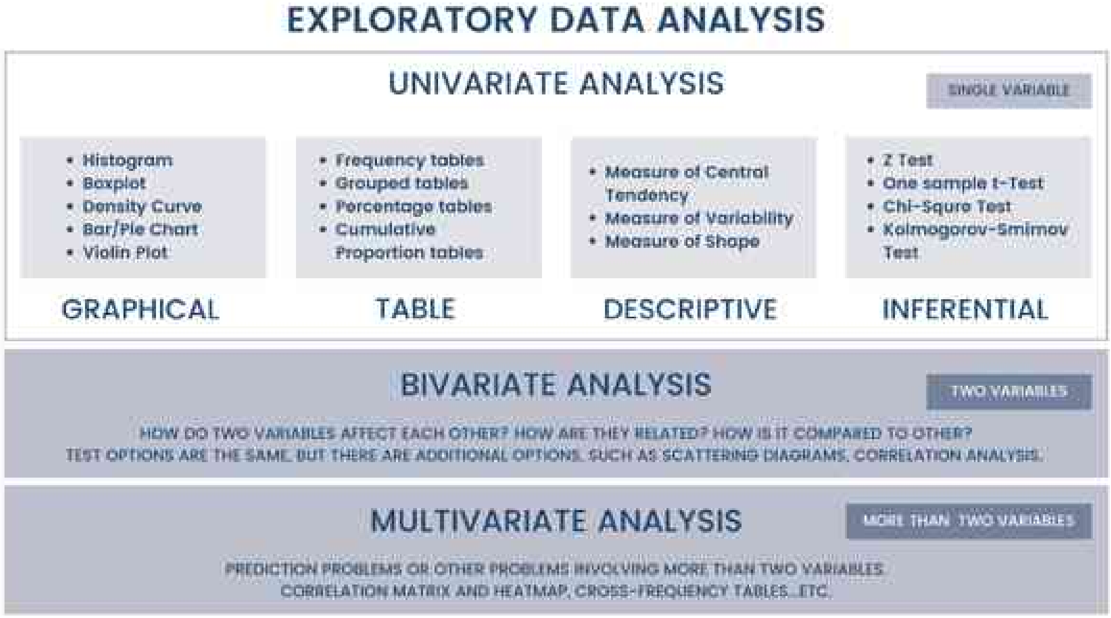


Figure 18 *Types of EDA*[117,118]

Before performing data analysis for statistical or prediction purposes, for example using machine learning, it is inevitable to understand the raw data in question. Understanding and evaluating the quality of data is necessary to detect and treat atypical or incorrect points to avoid possible errors that may affect the results of analysis.

One way to carry out this pre-processing is through exploratory data analysis. EDA can be divided into three parts[119] (Figure 18):

- Univariate analysis
- Bivariate analysis
- Multivariate analysis

Figure shows the most typical tool for the given type of analysis.

3.2.1. UNIVARIATE ANALYSIS

Univariate analysis is a type of statistical analysis that focuses on a single variable at a time. Its aim is to explore and summarize the characteristics of given variables to gain an understanding of its distribution and characteristics[120].

A single-variable analysis technique group is graphical, table, descriptive and inferential statistics[117].

Figure 18 presents the types of univariate data analysis in detail. In univariate analysis, frequency tables and bar graphs can be used to investigate variable distribution, histograms and box diagrams can be used to investigate continuous variables. Descriptive statistical analysis such as mean, median, mode, range, variance, standard deviation is also related.

Data visualization is a critical step in data science process. To enhance understanding, hybrid plots are used which combine the strengths of different chart types and helps to avoid losing valuable information.

To visualize the distribution of the dataset and summarize statistics, the combination of violin and strip plots were applied. Violin plot is a combination of a box plot and a kernel density plot[117]. The strip plot represents an implementation of a scatterplot that shows exactly the inner structure of distribution, its sample size, and the location of the individual observations.

3.2.2. BIVARIATE ANALYSIS

A bivariate correlation analysis whether and how two variables are linearly consistent, i.e. if one variable change linearly as the other variable changes.

Correlation is a bivariate analysis that measures the strength and the direction of the relationship between two variables[121]. Regarding the strength of the relationship, the values of the correlation vary between +1 and –1. A value of ±1 indicates the perfect relationship between two variables. When the correlation coefficient values go towards 0, the relationship between the two variables becomes weaker. The direction of the relationship is indicated by the coefficient symbol, + symbol represents a positive relationship, and – symbol represents a negative correlation. In general, in statistics, four types of correlations are measured: Pearson's correlation, Kendall's correlation, Spearman's correlation, and Point-Biserial's correlation. Pearson's correlation analysis is the most common method.

In the correlation analysis the first step is to make sure that the following five assumptions are met for calculating the Pearson's coefficient[122]:

- Level of Measurement: The two variables should be measured at the interval or ratio level.
- Linear Relationship: There should exist a linear relationship between the two variables.
- Normality: Both variables should be roughly normally distributed.
- Related Pairs: Each observation in the dataset should have a pair of values.
- No Outliers: There should be no extreme outliers in the dataset[123].

If the data does not match the normal distribution, another correlation analysis must be performed. The second most used type is Spearman correlation, because it relies on nearly all the same assumptions as the Pearson's correlation, but it does not rely on normality, and the data can be ordinal as well and thus, it is a non-parametric test.

Spearman's coefficient (ρ) is used to measure the monotonic correlation between two variables[124]. A monotonic function is a function of one variable which is either entirely increasing or decreasing. The Spearman correlation coefficient is defined as the Pearson correlation coefficient between the rank variables[125] (Eq. (8)).

$$\rho = 1 - \frac{6\sum d_i^2}{n(n^2-1)} \tag{8}$$

where d is the difference between the values of rank x_i and rank y_i, n is the number of observations.

The correlation coefficients between different variables are visualized in correlation heatmap that is a graphical representation of a correlation matrix.

To visualize the relationships between each variable, joint and pair plot were used[126]. It produces a matrix of relationships between each variable in your data for an instant examination. It can also be a great starting point to determine types of regression analysis to use. The plot is supplemented with kernel density estimate (KDE) along the diagonal, which provides the distribution of the data. There is a categorical variable within our data frame, and it can be used to visually enhance the plots and see trends and distributions for each category by coloring.

3.2.3. PROCESS OF EDA IN THIS STUDY

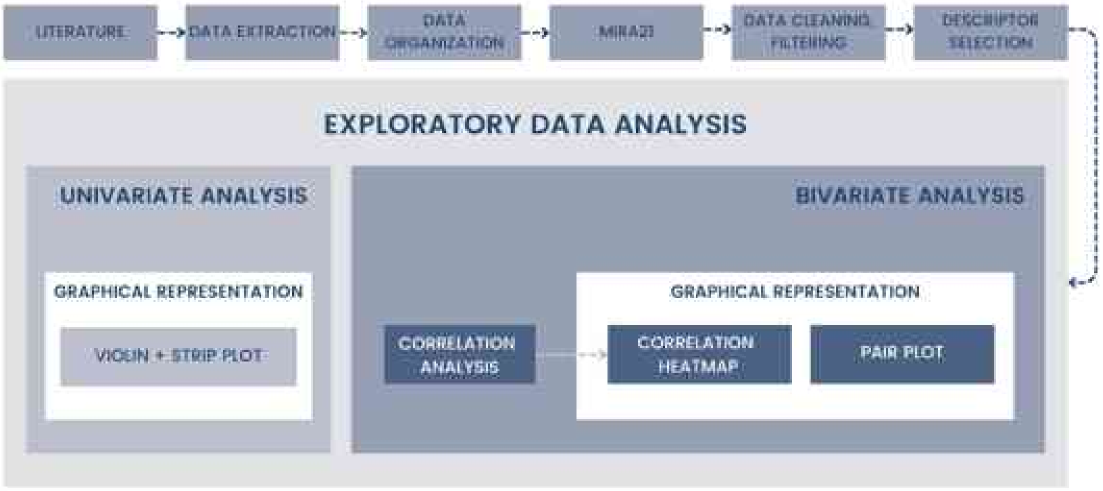


Figure 19 *Process of our exploratory data analysis*

Data analysis can be described in a multi-step process, as a result of which the necessary database is prepared for machine learning and a catalyst design strategy can be complied (Figure 19).

The next step is to clean and filter data based on the MIRA21 model database. During the data cleaning, the data were examined by descriptive statistical tests and then the parameters of the descriptor system were selected which exploratory data analysis were also occurred. After the univariate analysis process was performed, which examined the data sets of each parameter by performing violin and strip diagrams and determined the filters for the filtered data sets during the bivariate analysis. The normal distribution examination determines the correlation analysis to be performed. Subsequently, correlation analysis and pair analysis were performed. The data analysis results and conclusions are contained in the results and discussion section.

The EDA was carried out in python programming environment by using NumPy, Pandas, Seaborn, and Matplotlib libraries[127–130].

4. RESULTS AND DISCUSSION

4.1. MIRA21 STUDY OF NITROBENZENE HYDROGENATION REACTION[131]

MIRA21 model was initiated for use with catalysts suitable for nitrobenzene hydrogenation. The chemical reaction chosen is well-known in industrial and academic research.

BorsodChem manufactures toluene diamine by using aniline. In recent years the production of aniline itself has also been carried out in the company. In my R&D work, I also participate in the research into the hydrogenation process of aromatic nitro compounds, so this process is close to me. The industrial experience and approach contribute to correct interpretation of model-based analysis.

The university's László Vanyorek group is engaged in the development of catalysts for the catalytic hydrogenation of nitrobenzene and dinitrotoluene[132–134]. The group's experiments and results provide an opportunity to test and refine the model in-house and to take advantage of the application possibilities. In addition, theoretical chemical calculations are also carried out at the University in connection with TDA and MDI production, so this is a well-known reaction from the side of computational chemistry[135].

4.1.1. CATALYSIS LIBRARY

The Chapter describing the method of the MIRA21 model discusses which sources are used to build a catalyst database. As described above, the criteria for selecting literature were determined.

Scientific publications with a Q1 or Q2 rating (according to Scimago Journal Rank) in 2019 are included in the database[91]. Since hydrogenation of nitrobenzene is a regularly studied topic, only the results of the latest papers were collected. Consequently, only with articles written after 2000 were selected. The articles that met the first two criteria were then reviewed in terms of data content by quickly reading them.

At the time, a check was conducted whether the selected articles relating to the development of catalysts belonging to the corresponding reaction type, on the one hand, and whether the data could be collected from the articles by understanding the descriptor system. Many papers were excluded from the study because, although suitable for research conditions, they describe a more kinetic result or examine the effects of conditions, and do not focus on catalyst research.

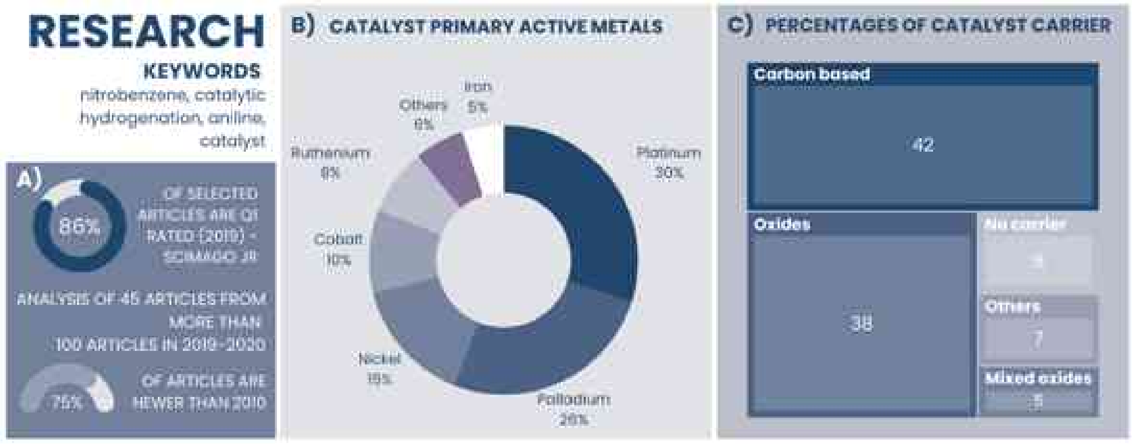


__Figure 20__ Summary of research database information [Box A) general information about the search is provided (publication year, Q index), box B) the distribution of the active metal components of the catalytic collection is presented in a donut diagram, box C) the distribution of the carriers is presented]

Figure 20 shows the filter conditions for collecting data and the first information collected from the database. The literature research was occurred based on the following keywords: ‘nitrobenzene, catalytic hydrogenation, aniline, catalyst’. 86% of the publications studied were classified as Q1, and 75% were published after

2010. In general, catalysts are supported systems that include one or two active components.

85% of catalysts are monometallic, while 15% of catalysts consist of two active components. A large percentage of prepared and applied hydrogenation catalysts contain palladium or platinum as active metal (Figure 20). Many studies have also been conducted on the catalysts of Ru, Rh, Co, Fe, Au and Cu. The distribution of active metals is slightly distorted. The occurrence of an active metal increased not only with the number of objects tested, but also with the number of catalysts tested.

Further examination of input data shows the variability of catalyst composition. Carbon based supports are usually used as catalyst carrier. Most of the carbon-containing catalysts are activated carbon due to their low cost, high performance, easy regeneration a reusability. Moreover, many experiments have been conducted to develop carbon nanotube catalysts. The group of oxide catalyst carriers is much more heterogeneous, for example, there are silicon, titanium, cerium, and aluminium oxide catalysts of this class.

During the process, two PhD colleagues and I reviewed and collected the scientific publications. The data processing was also carried out by three of us and the data was then checked. The construction of the first database of the MIRA21 model was an extremely time-consuming task, as human subjectivity had an important role to play in collecting data in literary processing. Therefore, initially, individual publications were managed in such a way that they were discussed together to reach an agreement on issues relating to data management. Such a question was, for example, how to enter conversion data semantically.

As a result, a catalyst library was created, consisting of 45 articles and 154 catalyst data sets, including about 4,500 data points [136–179].

4.1.2. RESULTS

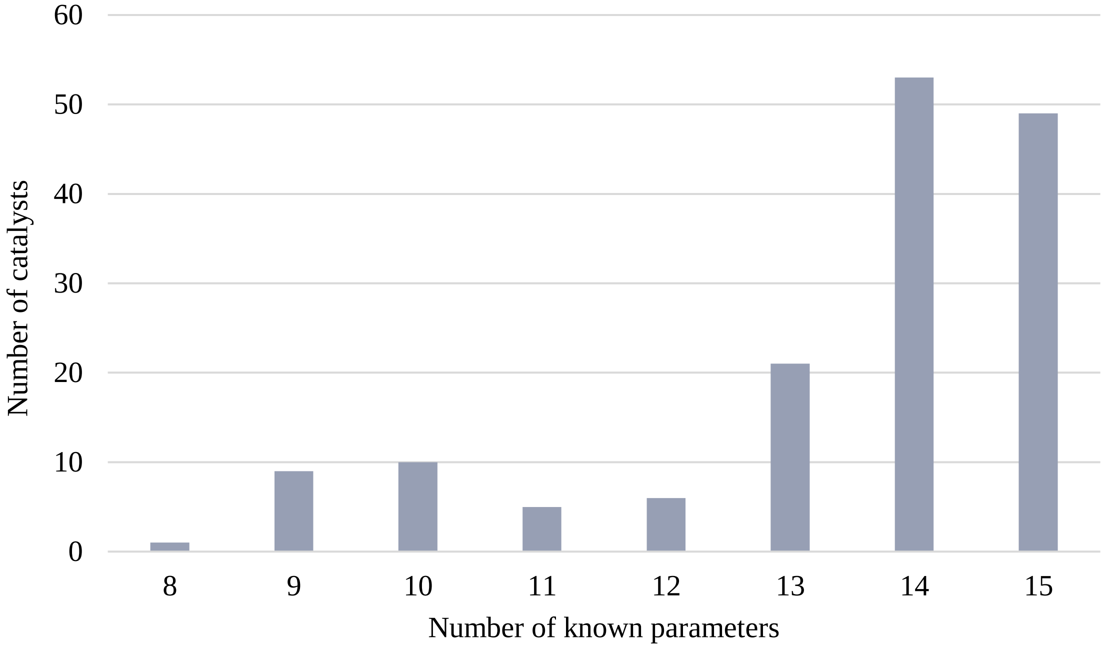


Figure 21 *Information content of studied scientific publications*

After the database was completed, the descriptor system was studied by univariate analysis.

Based on the data quality and frequency (Figure 21), catalysts can be well characterized with the determined parameters. The figure indicates that in 80 % of cases, catalysts can be characterized by at least 13 parameters. The minimum characteristics due to sustainability parameters are 4. Furthermore, most catalysts are characterized by 14 parameters. Based on the data processing, it was observed that the content of information differed even between catalysts examined within an article, as in many cases only the results of catalysts considered the best were described in detail or only this was further investigated.

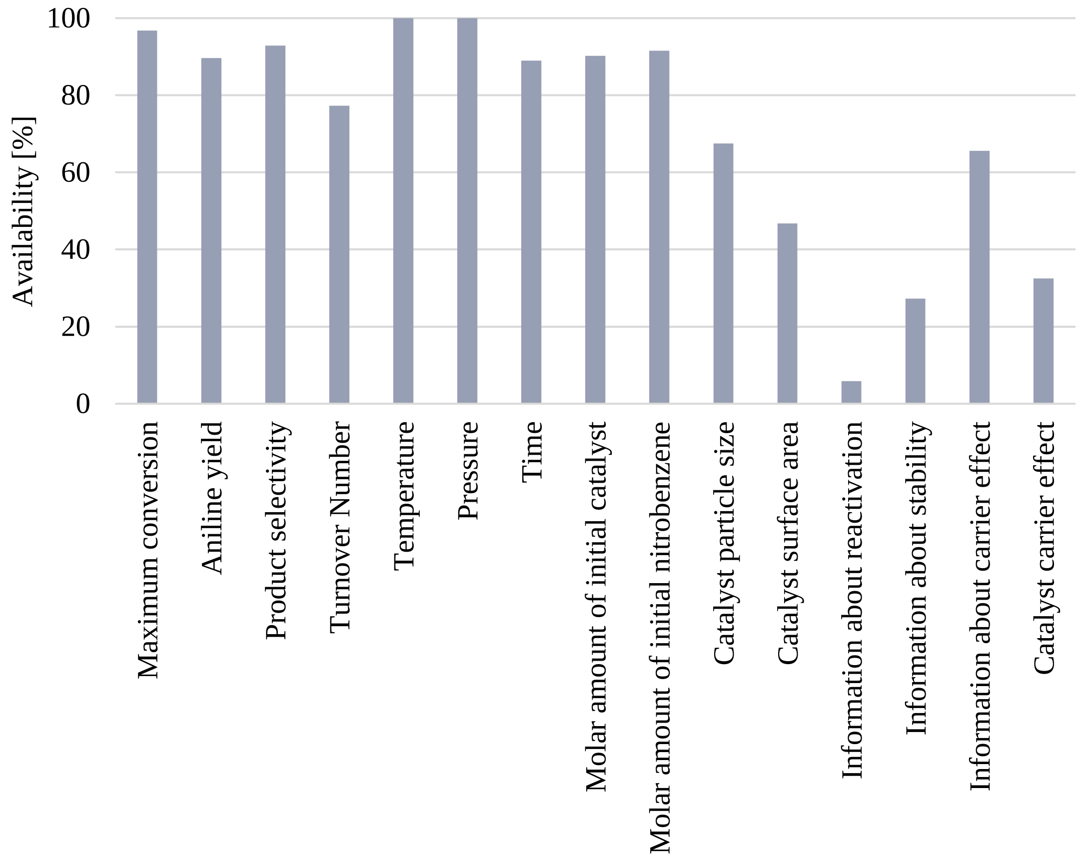


Figure 22 *Relative frequency of data occurrence according to MIRA21 descriptors*

Figure 22 summarizes how the data availability for the different descriptors has changed. The parameters that characterize catalyst performance and the parameters that describe catalyst experimental conditions are high-quality data available. In most cases, the extraction data was not available and so it had to be calculated. The two essential characteristics of chemical reactions are temperature and pressure. These indicators are always present in scientific publications, as was the case with us. However, in several cases, no response time has been given. In these cases, conversion rates and selection data were also missing or there was insufficient data to determine their calculation (flow reactor systems). More than 90% of the molar amounts are available from articles or can be deduced from other data. An interesting observation is that in some cases the molar amount of catalysts was not possible to calculate because the required data were not clearly presented. Furthermore, in some experiments, the initial molar amount of the original substrate was so low that it was difficult to interpret the performance of the reaction.

A small amount of information was available about the physical properties of catalysts. The most applied analytical measurements were the Brunauer–Emmett–Teller (BET) method and electron scanning microscopy analysis. The selected physical parameters are not often found in the article. In this case, the sustainability data indicate whether there is information on the subject in the publication.

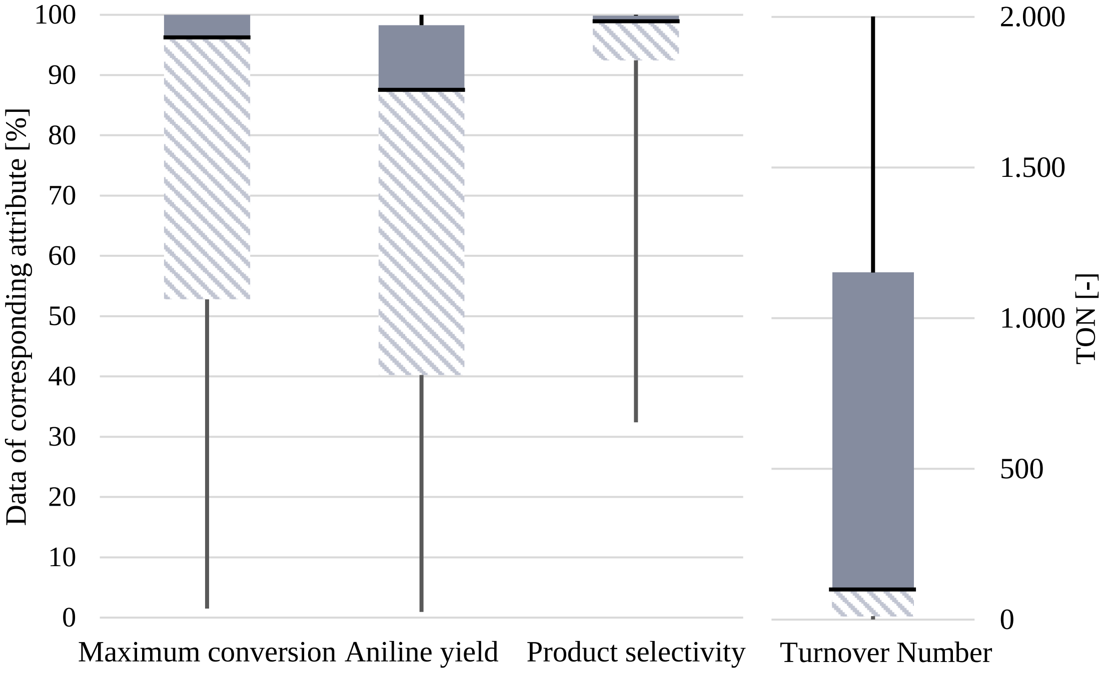


Figure 23 *Data distribution of catalyst performance descriptors on a box and whisker chart: maximum conversion, aniline yield and selectivity data in percentage (left), TON [-] (right) [On each box plot, the central black line indicates the median, and the solid pat of the column shows the first quartile, and the striped part shows the third quartile]*

According to the data analysis, it was found that the distribution of data by catalytic performance descriptors varies (Figure 22). The next box-and-whisker chart shows the minimum, maximum, Q1-Q3 range (contains the Q1-Q2-Q3 quartiles), median values of the conversion, yield, and selectivity values (Figure 23). The central black line shows the median. The end of the vertical black lines shows the minimum and maximum values of attributes. The solid column shows the first quartile, the striped column shows the third quartile. The meeting of solid and striped columns shows the median value.

Among the parameters describing catalyst performance, the selectivity can be highlighted, because as shown in the figure, the data range is much smaller than the other range, and the range between Q1 and Q3 is between 92 and 99 %. Outliers can be seen in all cases, but in data sets, the TON value has a significantly different maximum value. The TON value is displayed between 0 and 2000 in the diagram, but the maximum value is not displayed in the diagram, which indicates an outlier. Examining the data set, the highest TON was about 35 000 in two cases. The main reason for this difference is the different amount of catalyst used in hydrogenation test experiments.

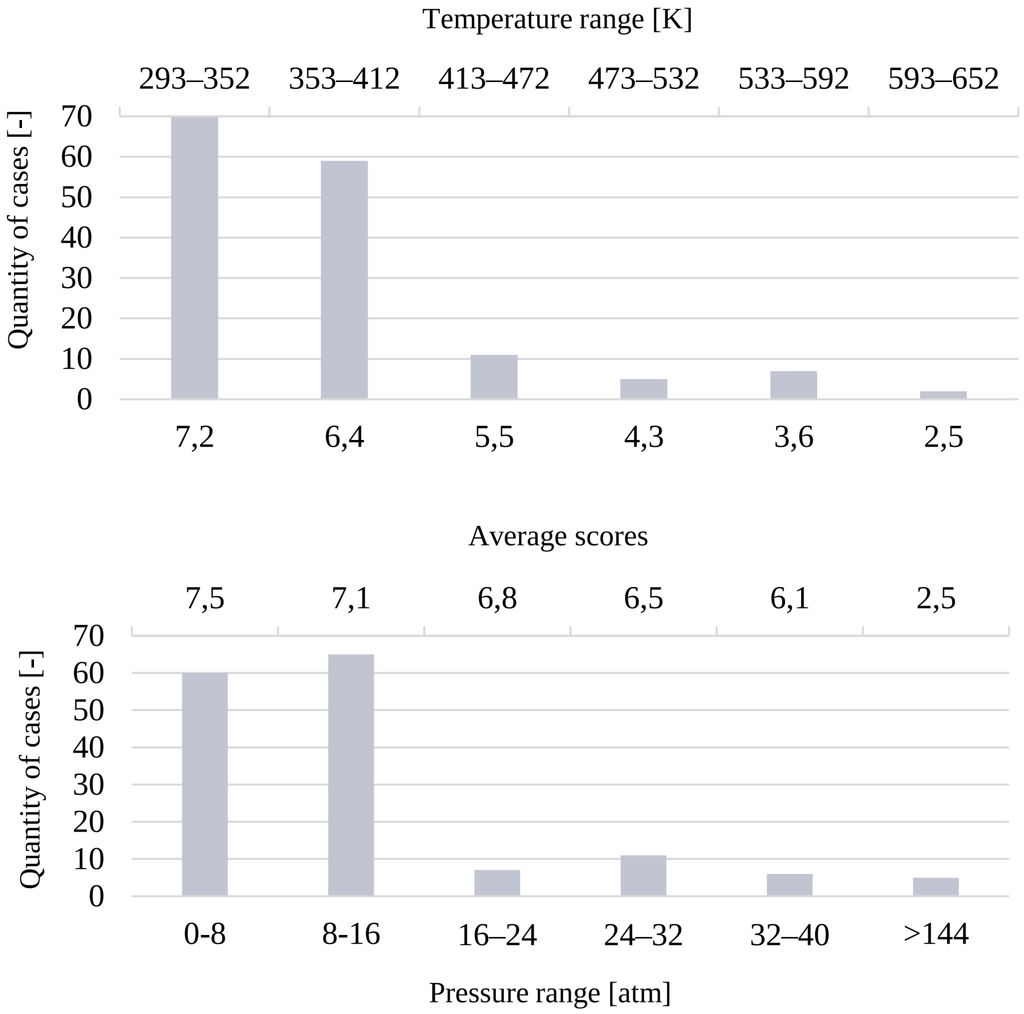


Figure 24 *Applied reaction conditions (top – temperature, bottom – pressure) [First x-axis shows the ranges, and the second x-axis shows the average scoring of ranges in MIRA21(According to Eq. 5 and Eq. 6).]*

As shown in Figure 24 , the catalyst used for hydrogenation has been tested in a wide range of reaction conditions. Compared to industrial conditions, laboratory experiments are usually carried out at lower temperatures. Catalysts are tested at an average temperature of 90°C, the general range was 293-412, but industrial aniline production occurs at 230-250 °C.

A normally used reaction pressure was less than 16 atm for hydrogenation of nitrobenzene under laboratory conditions. In our database, there is an outlier in terms of pressure. Zhao et al. studied the hydrogenation reaction of supercritical carbon dioxide[171]. They studied the influence of pressure, solvent and particle size on this reaction catalysed by Pt/C. It was shown that conversion increased as CO_2 pressure increased to 10 MPa but declined at pressure greater than 14 MPa.

The reaction time of the test experiments also differs greatly. In some catalyst tests, the hydrogenation time was several hours. The reason for multi-hour observation could be mainly to achieve maximum conversion that conversion increased over time. The shortest reaction time is 7 minutes for a carbon nanotube-based platinum catalyst under elevated conditions.

The last description evaluated the sustainability of catalysis. These questions can be answered for all catalysts. The catalysts examined in terms of reactivation, stability and catalyst support received a "+" rating. Furthermore, the catalytic whose support favoured the reaction received an additional "+" rating. Descriptors of regeneration defined for physical techniques, but regeneration procedures that contain chemical treatments were also included due to experience in data analysis. Zhang et al. studied the stability of cobalt catalysts and their recycling potential [77]. After each reaction, the catalyst was washed with ethanol and dried. The catalyst was reused at least five times without loss of activity. In a publication on cerium-based catalysts, centrifugation was used to separate the catalyst to reuse it [62].

Stability was reported in 27% of the samples. An important requirement for the catalyst to have a long life. During the stability test, the number of cycles the catalyst performs before the catalyst's activity begins to decline is determined. In articles in which several catalysts have been tested, only the best of these catalysts has been examined or described for stability. This may be one of the reasons for the low amount of data on the subject. the stability of the context was encountered as the recyclability or reusability of a catalyst in the articles. For example, Qu and others have investigated the stability of AuPd/TiO2 catalysts for solvent-free hydrogenation of nitrobenzene to aniline [79].

In several cases, the article focused on the effect of catalyst support. In a large proportion of studies, type of catalyst carriers was the catalyst's innovative part. For this reaction, researchers used various supports, including conventional carbon-based support systems, carbon nanotubes, cerium oxides and porous organic polymers. Gao et al. prepared several types of carbon hybrids based on N and S co-doped carbon for catalytic hydrogenation and investigated the role of cobalt salt [55]. These catalysts showed that they could effectively reduce functionalized nitro aromatic compounds to corresponding amines.

4.1.3. OVERALL RANKING

After the establishment of the data base, the MIRA21 rankings were implemented. Data scaling was done according to data normalization. The changes to the scale are already contained in the method description.

If the occurrence of data according to the descriptors was examined, information on reactivation, stability, surface of catalyst, carrier effect and particle size was the least available data. This experience correlates with the scoring of descriptors, except in the case of the last class of descriptor, but this was deliberate. We expected that these parameters could be used to differentiate the catalysts. This is the reason for the greater weighting of the sustainability questions.

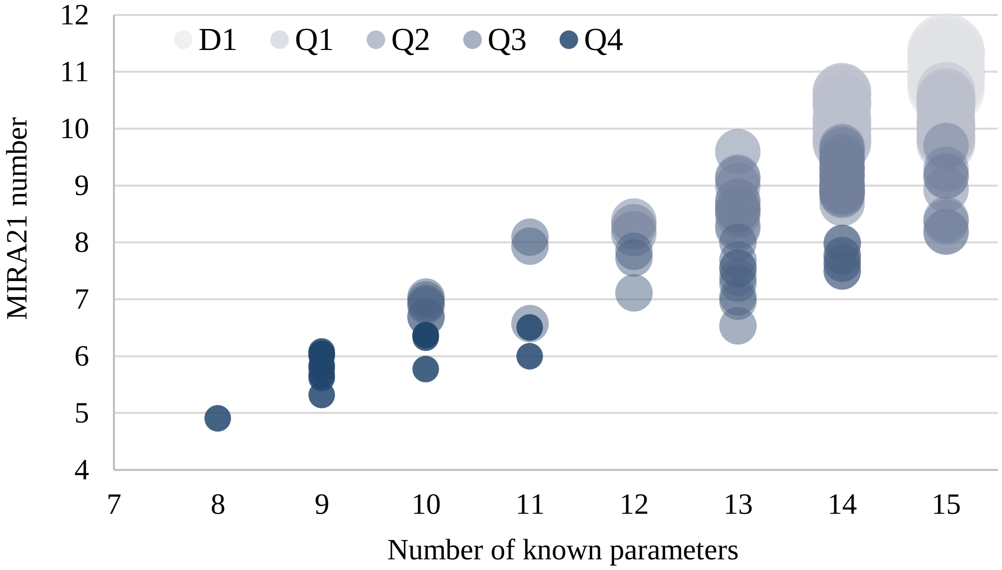


Figure 25 *Effect of increasing number of known parameters for MIRA21 number*

To provide evidence for our previous statement, Figure 25 highlights one of the bases of the number MIRA21. The more information about catalyst given, the higher the number MIRA21 that can be assigned to the catalyst. Furthermore, as the amount of data about the catalyst increases, the difference between the catalysts also increases. Reduced parameters were also studied. If the number from 15 to 10 was reduced, a much more unclear picture of the relative quality of catalysts was experienced.

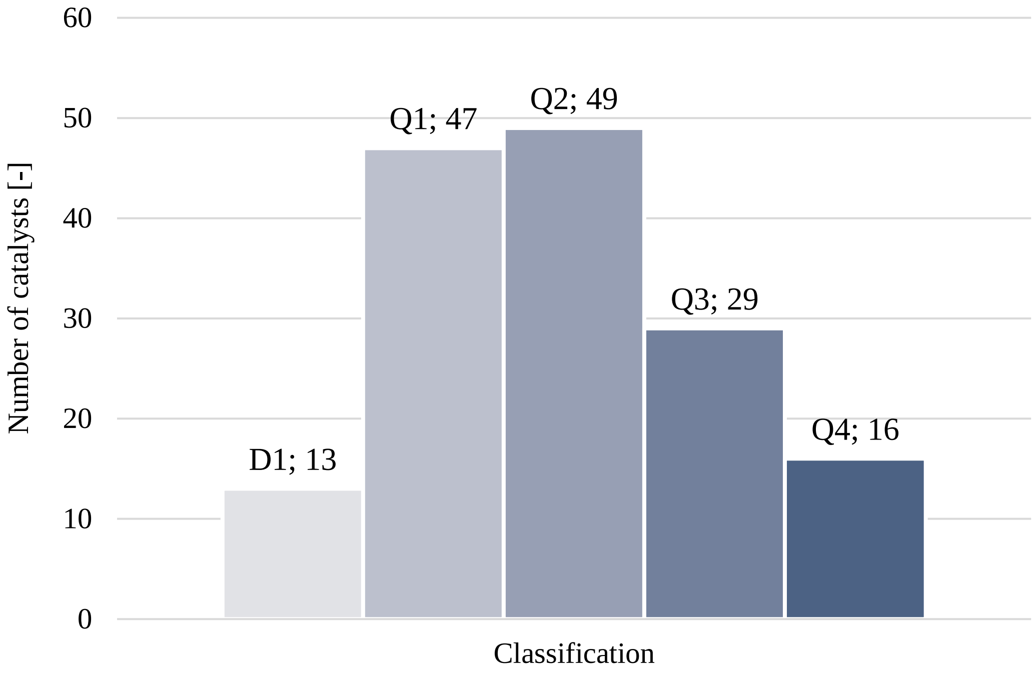


Figure 26 *Distribution of catalysts according to classification (D1, Q1, Q2, Q3, and Q4) system*

Figure 26 shows the distribution of the catalyst classifications. The total Q1 is made up of the Q1 and D1 categorises. According to the MIRA21 model, 12 catalysts were placed got D1 category. 60 of 154 catalysts were placed in the first quartile and 16 were placed in the last quartile.

The appendix contains catalysts from the catalyst data library (Q1-Q4), which provides catalyst identifiers, catalyst name, MIRA21 number, number of known parameters and classification (Appendix 2). Catalyst ID consists of reaction type, author address ID, year of publication, and the serial number of the catalyst.

Table 3 *Identification of D1 classified catalysts according to MIRA21 model*

RANK	CATALYST ID	CAT. Name in Journal	KNOWN parameters	MIRA21 number	Class.
1	HNB_XIA2021_1	0.07%Pt/@-ZrO_2/SBA-15	15	12.22	**D1**
2	HNB_BRA2015_1	Pd/C	15	12.22	**D1**
3	HNB_TAI2017_3	Co@NMC-800	15	12.13	**D1**
4	HNB_MIS2019_1	Pd/N-BCNT	15	12.04	**D1**
5	HNB_SHA2015_1	Pt/CMK-3	15	11.92	**D1**
6	HNB_MIS2020_1	5w/w% Pd-CC	15	11.84	**D1**
7	HNB_SHA2020_1	Pt/meso-Al_2O_3	15	11.83	**D1**
8	HNB_BEI2013_2	Pd/MWCNT-SA-4.3	15	11.79	**D1**
9	HNB_GUA2017_4	Pd/N@CNTs-1.5 h	15	11.77	**D1**
10	HNB_GUA2020_2	Pt CeO_2-R-600	15	11.72	**D1**
11	HNB_HAN2013_1	Pt@MIL-101	15	11.69	**D1**
12	HNB_LAN2020_3	γ-Fe_2O_3/NPC-800	15	11.65	**D1**

By calculating the number of MIRA21, determining the classification and classification of catalysts, the best catalysts based on our descriptor system were specified. Table 3 presents the D1 catalysts with known parameters, MIRA21 number and classification. The experimental results of the D1 catalyst were published after 2013.

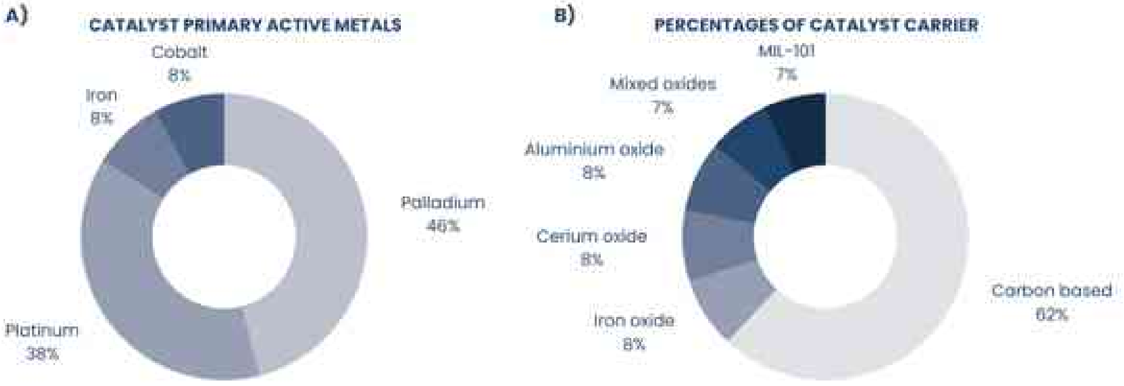


Figure 27 *Distribution of active metals (A) and type of catalyst carriers (B) in D1 classified catalysts*

Figure 27 summarizes the catalyst composition according to active compounds and catalyst carrier. The highest – rank monometallic catalysts included palladium or platinum as active compounds which are also supported by catalysts that have been classified Q1, so the situation is also similar. The results show that palladium and platinum can achieve similar good results, but from an economic point of view, it is not important which catalyst is used, it is also worthwhile optimizing the catalyst for cost. The results of catalyst support are no longer so uniform. The range of suitable types of carriers is quite varied. Among carriers, however, carbon-based carriers can be distinguished. For carbon-based substrates, the latest nanotube solutions can be highlighted from the database. From the D1 category, it can also be seen that it is difficult to compete with traditional Pd/C catalysts, as it is also one of the best.

The D1 catalyst experiments were carried out at temperatures ranging from 20 to 80 °C. There were much larger differences in the reaction pressure. The catalytic performance parameters of the D1 catalyst are almost identical, except for the TON value. Experiments with the D1 catalyst were conducted between 20 and 80°C. There were large differences in the content of professional information in the articles, whether it was a study of reactivation, regeneration, support effect or stability.

Y. Zhang et al. prepared hybrid 0.07% Pt/@-ZrO_2/SBA-15 nanostructure catalyst which demonstrated 100% conversion and 100% aniline selectivity at 40 °C, 7 atm in 50 minutes [173]. The high activity, selectivity, and stability of the catalyst can be attributed to the special structure of the catalyst and the synergistic effect within it. The authors compared this catalyst to many other catalysts from different study.

Research of Turákova et al. focused on the mechanism of the liquid phase hydrogenation of nitrobenzene [174]. They used a conventional palladium catalyst with activated carbon support. At 70 °C and 30 atm the nitrobenzene conversion was almost 100% after 40 minutes. This high pressure is commonly used in industrial applications.

The D1 category of MIRA21 also includes non-noble metal catalysts for nitrobenzene hydrogenation. It should be noted that most of the hydrogenation catalysts developed contain precious metals. In contrast, researchers from Taiwan developed an Co-based N-doped mesoporous carbon catalyst that demonstrates high catalytic activity and chemo selectivity for various nitro aromatics at 80 °C and 1 MPa with only 2 mol% of cobalt[169]. The synthesis of the catalysts facilitates the simultaneous optimization of porous features and cobalt nanoparticles. The research F. Zhang et al. have developed a new approach to improve the catalytic activity by the formation of an embedded cobalt-based catalyst with N- doped mesoporous carbon.

Another new trend in hydrogenation catalyst development is the “support on support” (SoS) type catalyst that contains a nitrogen-doped bamboo-like carbon nanotube (N-BCNT) on the surface of zeolite[137]. Vanyorek et al. developed a SoS system to improve the effectiveness of aniline production. Examination of various noble metal catalysts has shown that Pt/N-BCNT-zeolite was the most active (at 50 °C, 5 atm). However, in the case of Pd/N-BCNT only one main by-product was formed.

Li et al. prepared and investigated a platinum nanoparticle containing catalyst with a CMK3 ordered mesoporous carbon support[173]. They examined its catalytic activity for the hydrogenation of nitrobenzene and its derivatives in ethanol. According to the study, the performance of the Pt/CMK-3 catalyst was excellent within a very short time. A reusability test of the catalyst was also done. The Pt/CMK-3 catalyst could be recovered easily and could be reused more than 14 times with no loss in activity.

Researchers from the University of Miskolc examined another type of catalyst. They prepared a carbonized cellulose catalyst support and used palladium nanoparticles as active metal for hydrogenation[132]. The temperature dependence of the catalytic reaction was examined. The catalyst developed by Prekob et al. reached 100% conversion at 323 K and 20 atm in 240 minutes.

Nie et al. synthetize a mesoporous Al_2O_3-supported platinum catalyst[179]. They developed a special, solvent-free, rapid, and generalized method for catalyst preparation by ball milling. The catalyst performed well in the selective hydrogenation of nitrobenzene at 40 °C and 20 atm.

The research of Z. Wang et al. yielded a mild, green, and sustainable preparation method of a Pd/CNT catalyst[179]. The focal point of the process is that the implementation occurred in aqueous solution and at room temperature. The catalyst has shown high performance within 15 minutes under mild conditions. According to the recyclability test, the catalyst could be used three times without demonstrating a loss in activity and selectivity.

Experiments of Dong et al. focused on metallic impurities in carbon nanomaterials[179]. They demonstrated the deactivation effect of residual growth of N-doped carbon nanotubes for hydrogenation. This effect was examined through carbon nanotubes supported by palladium nanoparticles with controllable iron contamination. Only tens of ppm of iron contamination had a significant negative impact on the catalytic performance.

Cerium-oxide is a less commonly used catalyst support. According to a study, cerium-oxide supported platinum catalyst demonstrated a high level of aniline productivity. Q. Zhang et al studied the effect of the shape of the support and the key role of additional cerium ions sites[155]. The shape effect was attributed to exposed crystal planes on CeO_2 with different reducibility. High temperature reduction has improved the performance of the catalyst by providing additional Ce^{3+} sites on the surface. They prepared Na-containing cerium-oxide support because they found that Na^+ could help stabilize the Ce^{3+} surface.

Metal-organic frameworks have gained attention in recent years. Du et al. fabricated homogeneously dispersed platinum adatoms in an ordered mesoporous meta-organic framework[157]. Pt@MIL-101 catalysts also demonstrated a high catalytic activity under relatively mild conditions (20 °C and 10 atm). The high efficiency of the catalyst was attributed to the homogeneous deposition of platinum particles in the carrier.

The research of Lv et al. dealt with a preparation of iron oxide modified N-doped porous carbon catalyst derived from porous organic polymers[177]. The great advantage of the catalysts is that it can be easily recycled with a magnet and reused at least 10 times without reducing the catalytic activity and selectivity, according to the experiments.

4.1.4. CONCLUSION OF THE FIRST TEST SYSTEM

The aim of this research is to demonstrate the functioning of the MIRA21 model by studying the catalytic hydrogenation of nitrobenzene. The first section contains general information on the selected reaction, describes the MIRA21 ranking model, and compares 154 catalysts from 45 articles published over the past 20 years (2000-2020).

The aim of MIRA21 is to characterize the effectiveness of each catalyst with clear, objective, and minimal distortion of numerical data. A mathematical equation containing 15 factors was derived to classify catalysts. According to literature analysis, 15 parameters were found sufficient and necessary to distinguish between catalysts. However, due to the possible correlation between the descriptors, revisions to our descriptor system are justified. Outliers were identified during data processing. These exclusions have a significant impact on scale and score.

Using of MIRA21 facilitated the collection of information, because it determined the focus of the articles during the data processing. According to the model, the developed catalysts became more comparable. The ranking helps the researchers work by showing a simple number, which characterizes the hydrogenation catalyst. The ranking model can be flexibly applied to other catalytic reactions.

As the results of data processing, the experience was that the information found in articles are difficult to use due to the non-standardized data within them. Unclear wordings do not help the reader understand the main points of the publication. A pivotal point among the attributes describing the catalysts is the TON number. Furthermore, few studies would guide catalyst development on the path of industrial application. Based on the research, exploring knowledge about the sustainability of catalysts is beyond the focus of most research.

The ranking of the catalysts enabled the new development trends and directions to be mapped. According to MIRA21 model, the conclusions of this review about nitrobenzene hydrogenation to aniline are as follows.

Monometallic-supported catalysts are the most suitable for the hydrogenation of nitrobenzene, bimetallic or multimetallic catalysts did not show significant advantages. However, based on the results of the newest catalysts with the highest MIRA21 number, the metal content of the support has a positive impact on the catalyst performance. It is also observed that in the case of platinum catalyst, various transition metal oxides promote the hydrogenation properties of platinum. It turned out that small amount of platinum combined with transition metal oxides can be an effective competitor for palladium-carbon catalysts.

Precious metals are most used for aniline production, especially palladium or platinum, but there are some non-noble metals used as catalysts with excellent activity and selectivity, such as iron and cobalt. Development of novel carbon materials dominates activated carbon as a catalyst carrier, as the application of carbon nanotube carriers became more common due to their good catalytic performance. Classified catalysts D1 are composed of special compositions such as platinum adatoms in ordered mesoporous metal-organic frameworks or iron oxide modified N-doped porous catalyst derived from porous organic polymers.

4.2. MIRA21 STUDY OF DINITROTOLUENE HYDROGENATION[180]

4.2.1. AIM

The model's application has two directions: one is the revision of the descriptor system by statistical methods, and the other is the application of the model to other reactions. Hydrogenation of 2,4-dinitrotoluene to 2,4-toluene is an important technological step in the polyurethane industry. The technological process, the reaction mechanism and the reaction kinetics have been studied and generally accepted, but much remains to be learned about the catalysis of this process. Therefore, the map of the current state of the development of catalysts also facilitates the development of scientific research. However, the review of the literature on catalysts used in TDA synthesis does not provide sufficient information to achieve this objective. The comparison of the catalysts studied so far provides a much more complete picture of the latest developments in their effectiveness. The MIRA21 model was therefore used to conduct the characterization, comparison, ranking and classification of catalysts[131].

4.2.2. CATALYST LIBRARY

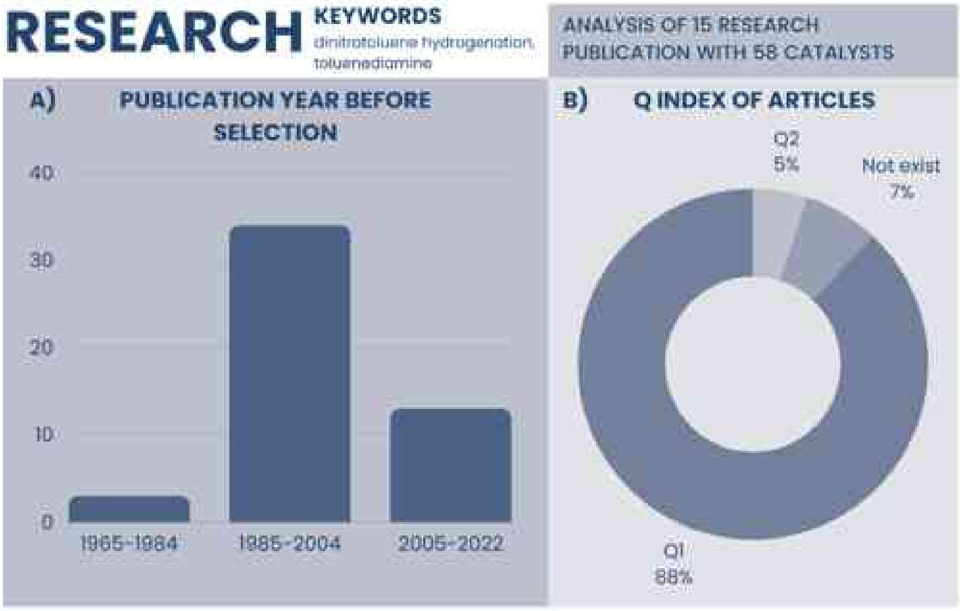


Figure 28 *Publication year distribution of 56 articles after first selection (Box A), and Q index distribution of 15 articles after second selection (Box B)*

The results of the literature research are surprising because there are relatively few published scientific results about the dinitrotoluene hydrogenation process. They were mostly prepared before the 2000s. Based on Google Scholar searches for the keywords ‘dinitrotoluene hydrogenation’, 2210 matches were given, however if ‘toluenediamine’ was added there were only 212 hits. 92 pieces of these included scientific results obtained after 2010. To demonstrate this, the keyword ‘kinetic’ was added to the initial search, which then yielded 120 articles. Overall, only a few research groups have studied TDA synthesis and have prepared catalysts for this reaction. On one hand, a smaller database reduces the reliability of the MIRA21 results. On the other hand, a smaller dataset makes it easier to delineate the possible research pathways on the topic.

After the first selection 56 articles remained. During the data analysis, it was justified to change the publication year selection criteria (after 2000). Figure 28 A) shows the distribution of scientific publications according to publication date. B) part of the figure presents the studied articles based on its Q-index in 2021 after the primary article selection (relevance, publication year, Q-index). The figure shows that the data used to analyse the catalysts mainly came from Q1 articles. A few publications whose publisher has since ceased to exist were also included in the analysis, because they had previously provided space for the publication of high-quality scientific works.

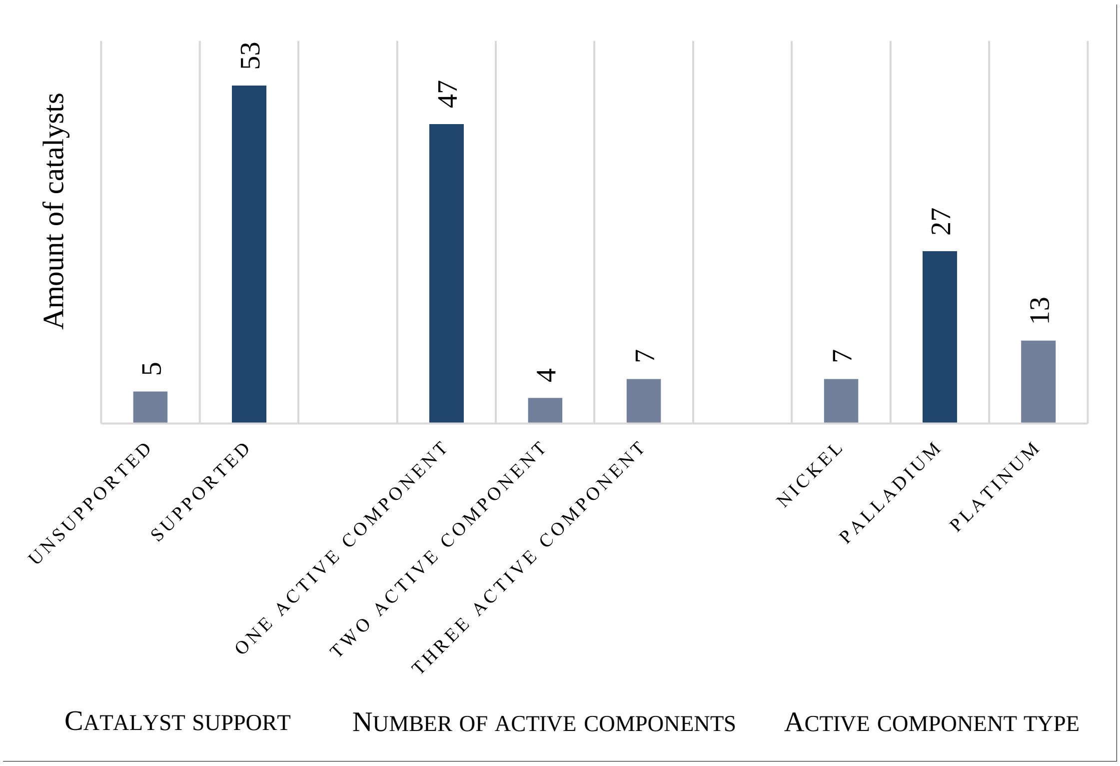


Figure 29 *Composition of 58 studied catalysts according to support and active component*

Selected 58 qualified catalysts of 15 articles are mostly supported catalyst (Figure 29 left)[143,144,146,147,149,157,169,181–196]. Most of the produced catalysts contain one active component on the support (middle of the figure). The catalysts with two active components generally apply palladium-platinum, palladium-iron combinations. Catalytic systems containing three active components are composed of either iridium-manganese-iron, iridium-iron-cobalt or nickel-lanthanum-boron. The frequency of the active metal components is in the order of Pd > Pt > Ni. In addition to palladium and platinum, nickel can also be seen, which is used as a common catalyst in industrial practice (Figure 29 right). Regarding the catalyst carrier, metal oxides (zirconium, chromium, titanium, aluminum, and silicon), ferrites, maghemites, zeolites, and activated carbon as typical in the chemical industry were identified. Occasionally, PVP-based catalysts were also investigated[196].

4.2.3. OVERALL RANKING

Based on catalyst library available data, it is difficult to get a consistent picture of DNT hydrogenation catalysts. However, these catalysts can be well qualified and comparable according to MIRA21 model. A total of 58 catalysts from 15 articles reporting research results were successfully analysed.

The catalyst is detailed because 10 or more known parameters (of 15) can be collected in each case. Test reaction conditions are 295-393 K and 1-50 atm, except in two cases (95-150 atm). The rime required to convert maximum amount ranged from a few minutes to a one-day interval, indicating a relatively large standard deviation. The average reaction time for conversion of 100% in 60 minutes.

The amount of initial dinitrotoluene was in the range of 0.002 and 0.3 mol. The amount of active metal in the catalyst also showed a large deviation from $5.13*10^{-7}$ mol to 0.034 mol. Despite the low amount of catalyst as was mentioned above, 100% conversion was achieved[191]. The increased amount of material was typical for nickel-type catalysts.

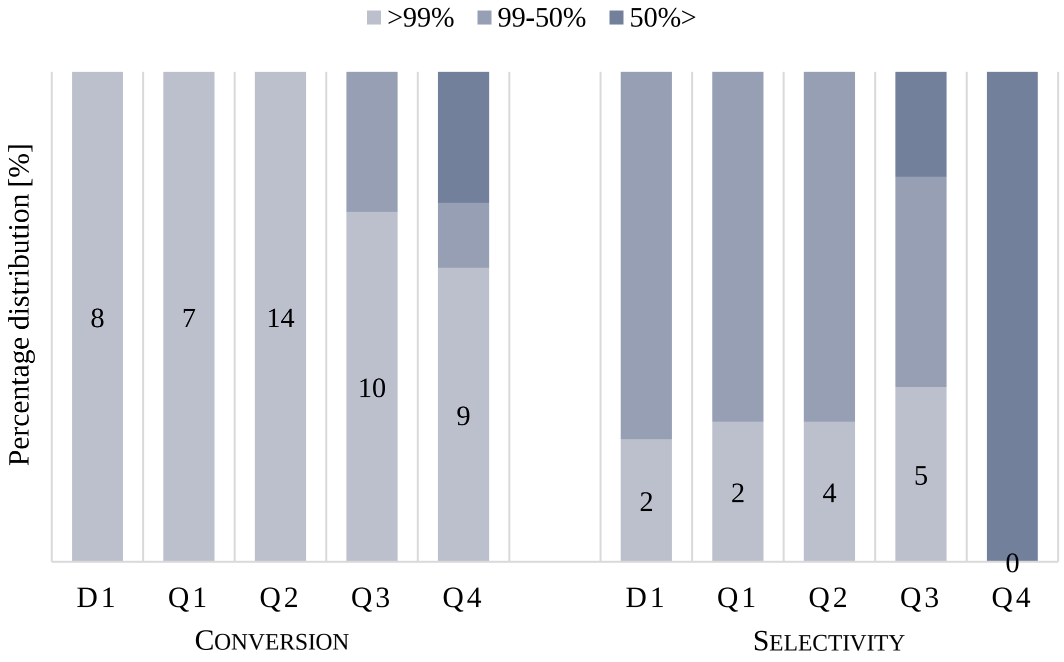


Figure 30 *Catalyst performance of catalysts – conversion and selectivity depend on classification*

Figure 30 shows the catalytic performance results of selected, studied, characterized, classified catalysts. The conversion rate of the catalyst studied in the

D1-Q1-Q2 class is over 99 n/n%, but the selectivity is much more differentiated. Based on these results, it can be said that achieving a pure TDA product produced during hydrogenation is a serious challenge for researchers. The worst catalysts(Q4) performed at less than 50 n/n%.

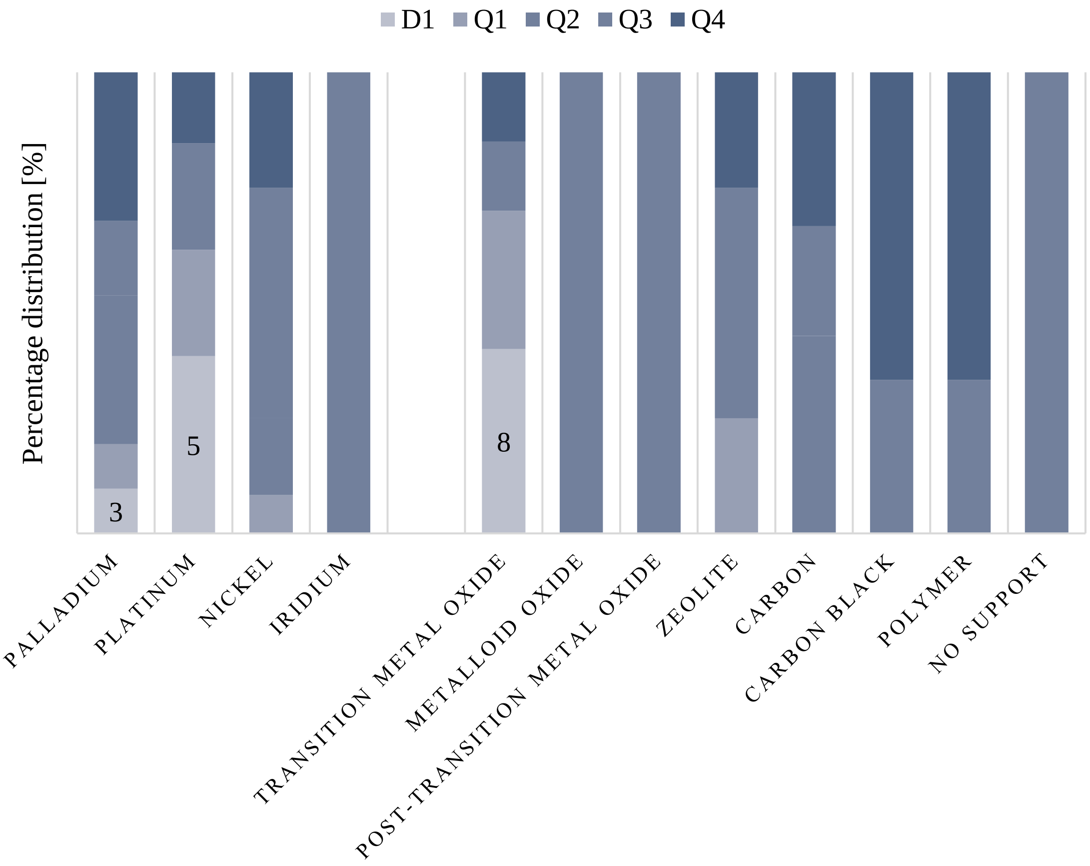


Figure 31 *Distribution and active components of catalysts according to MIRA21 ranking and classification (D1-best, Q4-worst qualification – according to MIRA21 colouring)*

The catalyst composition changes according to the MIRA21 model classification. Figure 31 shows the active components and support types of catalytic system based on their classification. The best-performing catalysts (class D1) consist of palladium or platinum and transition metal oxide support. Although nickel is more widely used in the industry, these types of catalysts are at the bottom of the list. Iridium, an active component of catalyst also got relatively good number of MIRA21s. Most of unsupported catalysts, carbon black, aluminium oxide, silicon oxide is in the lower half of the ranking. Practically, the catalyst carrier of the system differed according to the MIRA21 classes. It can be found in Q2 and Q4 classes mainly activated carbon supports. Catalysts with transition metal supports are at the top of the ranking.

Table 4 *TOP8 catalyst of DNT MIRA21 ranking*

No.	CATALYST ID	Catalyst name	Support	Active component	Number of known parameters	MIRA21 number
1	HDNT/MIS/2021/2/2	Pt/CrO2	Chromium (IV)-dioxide	Platinum	15	11.50
2	HDNT/MIS/2021/2/1	Pd/CrO2	Chromium (IV)-dioxide	Palladium	15	11.49
3	HDNT/MIS/2021/3/1	Pd/NiFe2O4	Nickel ferrite	Palladium	15	11.45
4	HDNT/TIA/2020/1/3	15Pt/ZrO2-300	Zirconium-dioxide	Platinum	13	11.44
5	HDNT/TIA/2020/1/4	15Pt/ZrO2-400	Zirconium-dioxide	Platinum	13	11.43
6	HDNT/TIA/2020/1/2	15Pt/ZrO2-200	Zirconium-dioxide	Platinum	13	11.42
7	HDNT/MIS/2021/1/2	Pd/maghemite	Maghemite	Palladium	15	11.35
8	HDNT/TIA/2020/1/5	45Pt/ZrO2-300	Zirconium-dioxide	Platinum	13	11.06

The 8 best D1 classified catalysts are listed in Table 4. These columns include the ID code, catalyst name, type of catalyst support, active component, known parameters, calculated MIRA21 number. Additional results are included in Appendix 3.

The best DNT hydrogenation MIRA21 catalysts consist of an active component and a transitional metal oxide support. Based on these results, the catalysts containing platinum produced better results than their competitors. The synergistic effect of the combination of active components is difficult to assess because there is not enough information. Class D1 includes catalysts that are studied based on sustainability considerations such as stability and reactivation capability, too.

When compared to the catalyst classification analysed for nitrobenzene hydrogenation reactions, the best catalyst in the MIRA21 classification was similar to the first-class Pt/ZrO_2 catalyst, one of the most effective catalysts. Zhang et al.

developed hybrid nanostructure catalysts $Pt/ZrO_2/SBA\text{-}15$ that had excellent catalytic performance at 313 K, 7 atm and 50 minutes for hydrogenation of nitrobenzene to aniline[173]. They found that ZrO_2 dispersion in the SBA-15 improved the catalyst performance due to its mesoporous structure. Consequently, it would be useful to try this catalyst for TDA synthesis as well.

The work of Hajdu et al. focused on the development of new magnetic catalysts for the hydrogenation of DNT to TDA[189,193,197]. One of the catalysts is $Pd/NiFe_2O_4$ that has achieved 99 n/n% TDA yield at 333 K and 20 atm. In this work, they synthetized nickel ferrite spinel nanoparticles to solve the problem of separating the catalyst from the products by magnetization. Another magnetic catalyst with good catalytic performance is Pd/maghemite, which is made by a combustion method with a sonochemical step. Palladium on a maghemite support resulted a high catalytic activity for TDA synthesis at about 60 minutes and under the same reaction conditions as ferrite hydrogenation.

The first and second position of the MIRA were those of chromium oxide platinum and palladium catalysts. These innovative systems have produced excellent catalysts with performance. It was prepared with chromium (IV) oxide nanowires decorated with platinum and palladium nanoparticles. These catalysts showed high catalytic activity at 333 K and 20 atm. If a catalyst Pt/CrO_2 was used, 304.8 mol TDA would be produced under these conditions, while only 1 mol of precious metal catalyst would be used. If palladium is used, only 60.14 mol TDA is produced, but it is also relatively large quantity. From an industrial point of view, it is important to separate this type of catalyst from the reaction mixture easily due to its magnetic properties. The stability of the catalyst has been studied, and it has been found that the catalyst can be used at least four times without regeneration.

Ren and his colleagues made half of the D1 class catalysts, and these catalytic systems consisted of zirconium oxide supports and platinum precious metal[198]. Ren et al. prepared ZrO_2-supported platinum catalysts with different Pt concentrations and at different reduction temperatures. They found that 0.156% Pt/ZrO_2 catalyst has the highest catalytic performance at 353 K and 20 atm. According to their results, the use of this catalyst reached an initial hydrogen consumption of 4583 mol H_2 mol Pt^{-1} min^{-1}. In this work, they investigated the interaction between the precious metal and the oxide support. It was found that zirconium oxide has the highest adsorption capacity for platinum ions due to its ability to be protonated and deprotonated.

4.2.4. CONCLUSIONS OF THE SECOND TEST

The purpose of this study was to summarize the hydrogenation of dinitrotoluene to toluenediamine and testing the applicability of MIRA model. 58 catalysts were selected from 15 research articles and studied using the MIRA21 model, which covers the complete scientific literature on catalysts of DNT hydrogenation. According to ranking and classification, eight catalysts were ranked in the highest class (D1).

The number of catalysts developed for the synthesis of TDA is low, as scientific research focuses mainly on reaction mechanisms and reaction kinetics. Despite this, many different catalysts have been developed. Based on the descriptor system, the catalysts tested were well characterized.

More than 80 %of the 58 catalysts produced and tested have an excellent conversion, but only 45 % have demonstrated a selectivity above 90 n/n%. More than 80% of the catalysts produced are composed of a single active component. Since the combination of catalysts has not been investigated in large numbers, one suggested direction of research is multicomponent catalysts. The development of catalysts represents a new trend that has led to the creation of many high-performance catalysts. According to the analysed catalysts, catalysts with oxide and/or magnetic supports showed better results under laboratory conditions compared to traditional carbon-based supports. Carbon-supported nickel catalysts are mainly used in industry, but nickel catalysts have not produced the best results. The advantage of a good magnetic catalyst is that it can be repaired, but the economic impact of its industrial application must also be taken into account.

4.3. REVISION OF MIRA21

During the doctoral research, the MIRA21 model was established, which characterized catalysts with quantified values that were comparable and classified. 15 parameters were chosen based on the catalysts were characterized from literature sources. The model was used for catalysts for hydrogenation of nitrobenzene and dinitrotoluene.

The data were normalized and weighted, and the MIRA21 number describing the catalyst was obtained with a calculation formula. Based on the number MIRA21, a ranking and classes were created that facilitate a faster and more transparent classification of catalysts. Using this model, recent research was analysed, qualified catalysts, and found the most promising results. Model descriptor systems and classifications have begun to be used in the Institute of Chemistry, on the one hand as catalyst characteristics formulas, and on the other as publication writing standards.

Research in literature, data collection and correct interpretation, as well as data evaluation, have enabled us to gain experience in order to plan further research.

The study and processing of specialized literature has given rise to experience that reveals difficulties in formulating and interpreting research results. It was recognized that the formal structure of the database must be modified. The frequency and consistent handling of selected descriptors were observed. It is also considered that the scope of data sets and the examination of outliers are justified. It was found that a lack of examination of the relationship between parameter can distort the system. It was noted that, by adjusting the weighting criteria system, a flexible operational classification could be established. This classification proved beneficial in the selection or development of catalysts. It was also found that the model could be applied appropriately to almost the same reaction, but additional research on its applicability is required.

The discussion of the results was extended in two additional directions. One was to analyse the databases built, and the other was to review the model itself. The creation of catalyst databases and data analysis also opened a new research chapter focusing on the possibilities of applying machine learning to catalyst research.

4.4. EXPLORATORY DATA ANALYSIS ON DNT CATALYST LIBRARY[199]

Literature data on 2,4-dinitrotoluene hydrogenation catalysts for the synthesis of toluenediamine were analysed with the MIRA21 descriptor system.

First, the relationship between the different variables was examined, and then patterns were searched in the system regarding the composition of the catalyst. As a result of the correlation examination of properties describing the catalyst, information was collected for the ranking. By mapping the patterns, the composition of those catalysts can be predicted which can provide excellent performance during the hydrogenation of 2,4-dinitrotoluene.

Since there was a relatively small amount of data available for this reaction, the results carried some uncertainty. However, the applicability of exploratory data analysis procedure to other reactions is undeniable.

The study used Exploratory Data Analysis (EDA) to understand the relationship between individual variables such as catalyst performance, reaction conditions, catalyst composition and sustainable parameters. The results would be applicable to catalyst design and would also be possible to use machine learning tools.

Table 5 *Selected parameters for EDA*

Catalyst composition	Catalyst performance	Reaction conditions	Catalyst properties	Sustainability parameters
Active metal content	Maximum conversion	Temperature	Catalyst particle size	Reactivation
Type of active metal content	Product yield	Pressure	Catalyst surface area	Stability
Number of active metals	Product selectivity	Time		Catalyst Carrier
Catalyst carrier	Turnover number	Molar amount of catalyst		
		Molar amount of 2,4-dinitrotoluene		

Prior to the data analysis, a selection process was performed, in which the parameters of the descriptor system were selected (Table 5, coloured cells). Only quantifiable data were examined and thus sustainability parameters were not analysed. On the other hand, there is insufficient data on variables related to the physical properties of catalysts. Therefore, catalyst properties were also excluded from the analysis. This allows the relationship between catalyst performance and reaction conditions to be investigated. To sum, two types of descriptor variable group were studied include 9 different parameters (grey) for correlation analysis.

In addition, catalyst composition group was also examined (purple): active metal content, type and number of active metal component and catalyst carrier. Catalyst composition as a parameter group was also included in the analysis, since the aim of the work was to map the relationship between input (composition) and output parameters such as catalyst performance. This group include quantifiable (active metal content) data, which are treated in the same way as the other quantifiable parameters. Furthermore, there are quality parameters that can be classified and are generally considered to be a third dimension with colour markings.

4.4.1. DATA DISTRIBUTION

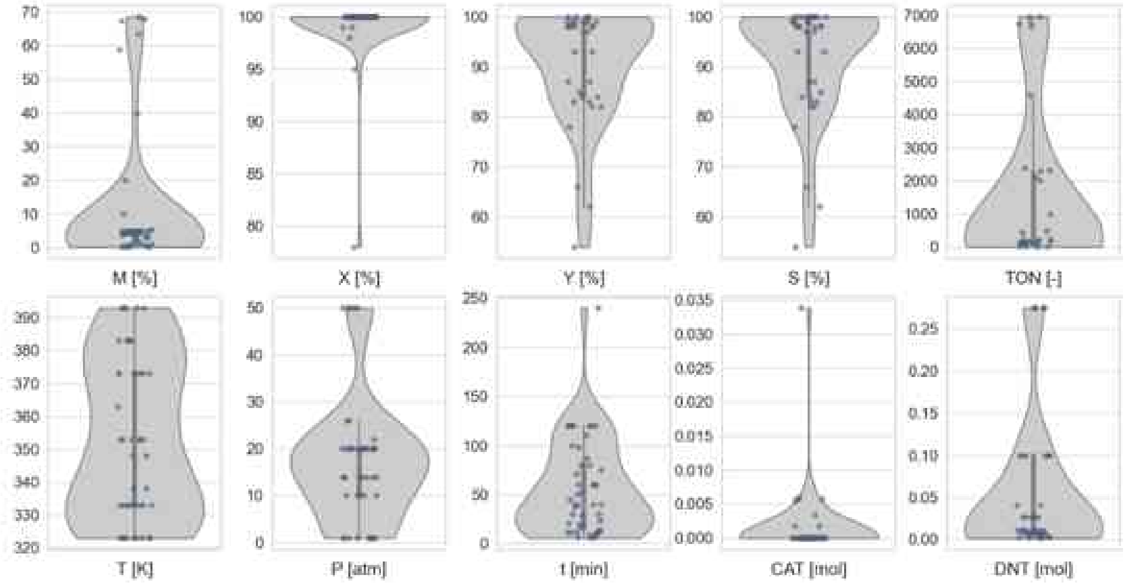


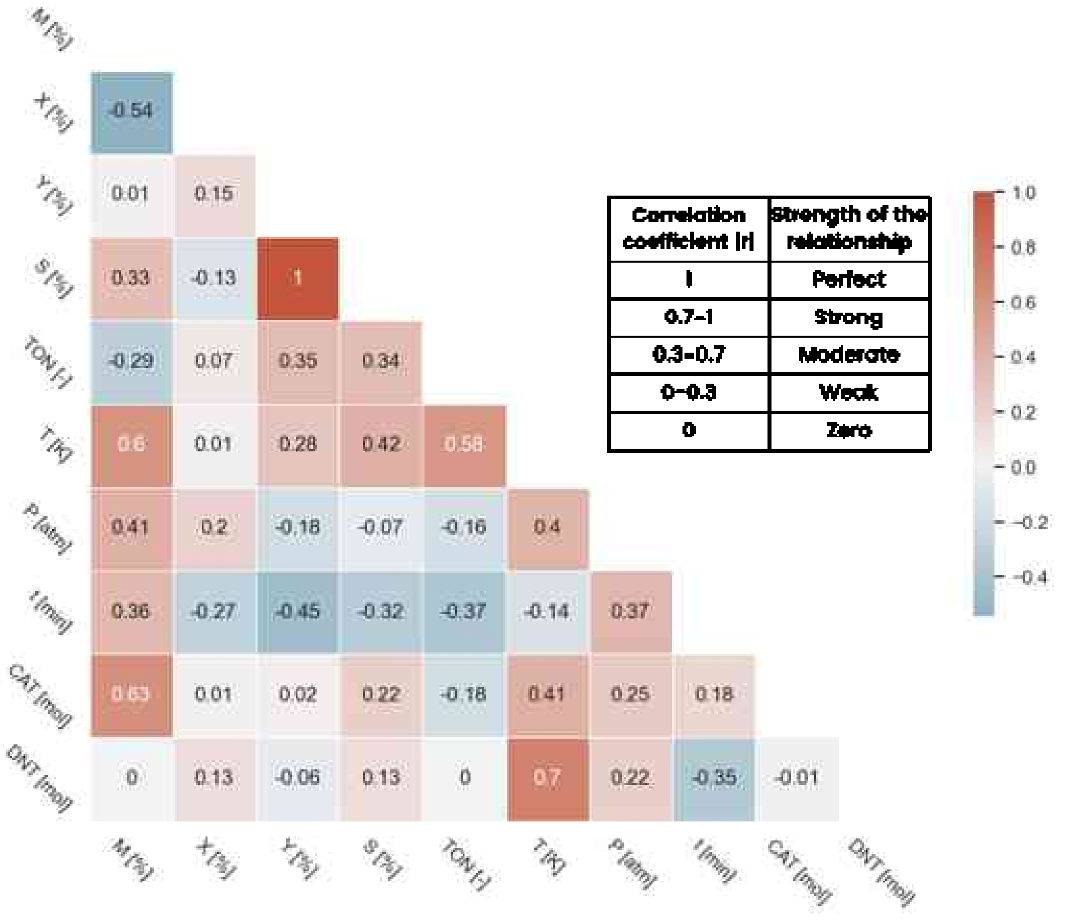
Figure 32 *Violin plot diagram of the selected variables [M =active component w/w%, X=conversion, Y=yield, S=selectivity, TON=turnover number, T=temperature, P=pressure, t=time, CAT=molar amount of active component, DNT=molar amount of dinitrotoluene]*

Before correlation analysis, it is necessary to examine the numerical data set applied. The violin plot of the catalyst library was created using the studied parameters (Figure 32). This special type of plot can show the data median, the data range between the quarters and the whole distribution of different data values. The width of the curve means the frequency of the data point.

The sample was heterogeneous in terms of data values. Based on the diagrams of the different parameters, it can be concluded that the data do not follow the normal distribution. In most cases, the form of the violin is much more similar to the bimodal distribution or the log normal distribution with its asymmetries. In case of maximum conversion (X), time (t), catalyst amount (CAT), there is also a shape that shows that there may be exceptions in the system.

One of the conditions for correlation analysis is the evaluation of the outliers and their omissions. To ensure that outliers do not affect correlation analysis, the data were filtered. By displaying the data, the extreme values were selected and removed from the data set. This filtering occurred by defining the limit values of the justified parameters. The catalyst with a maximum conversion rate of more than 50%, a product rate of more than 50%, a reaction time of less than 240 minutes and a pressure of less than 50 atm was analysed. Unfortunately, data cleaning processes reduce the available data, but also reduce the distortion of correlation analysis results.

4.4.2. CORRELATION ANALYSIS



	M [%]	X [%]	Y [%]	S [%]	TON [-]	T [K]	P [atm]	t [min]	CAT [mol]	DNT [mol]
M [%]										
X [%]	-0.54									
Y [%]	0.01	0.15								
S [%]	0.33	-0.13	1							
TON [-]	-0.29	0.07	0.35	0.34						
T [K]	0.6	0.01	0.28	0.42	0.58					
P [atm]	0.41	0.2	-0.18	-0.07	-0.16	0.4				
t [min]	0.36	-0.27	-0.45	-0.32	-0.37	-0.14	0.37			
CAT [mol]	0.63	0.01	0.02	0.22	-0.18	0.41	0.25	0.18		
DNT [mol]	0	0.13	-0.06	0.13	0	0.7	0.22	-0.35	-0.01	

Correlation coefficient |r|	Strength of the relationship
1	Perfect
0.7–1	Strong
0.3–0.7	Moderate
0–0.3	Weak
0	Zero

Figure 33 *Results of Spearman correlation analysis [The colour of each element of the matrix shows the strength of the connection between the parameters. The table shows the indicators used in relation to the value of the correlation coefficient*[200]*. (M =active metal content, X=conversion, Y=yield, S=selectivity, TON=turnover number, T=temperature, P=pressure, t=time, CAT=molar amount of active component, DNT=molar amount of dinitrotoluene)]*

Figure 33 summarizes the results of Spearman's correlation analysis in a correlation matrix, and the correlation coefficient values are indicated by colour and number in the heat map. The figure was created based on the filtered data set. The entire data set’s correlation matrix and the difference correlation matrix are included in the attachments (Appendix 4-5).

The correlation coefficients calculated between individual parameter pairs vary greatly. The results show that there is a practically perfect correlation and a strong relationship. Product yield (Y) and product selectivity (S) are perfect because these two parameters can be derived from each other. Therefore, from the perspective of the MIRA21 model, it is justified to modify the parameter set and remove one. There is also a strong correlation between the quantity of starting material (DNT), and the temperature (T). As a results, the higher the amount of starting material, the higher the reaction temperature used. The correlation coefficient of the catalyst material quantity-temperature pair is 0.41. Accordingly, in the case of larger starting materials (DNT, CAT), the catalysts are tested at higher temperature.

A moderate relationship between the active metal content (M) and CAT was determined. These data are also derived from each other, so the relationship between them is clear. Therefore, the higher correlation coefficient value of CAT-M-TON-T is not surprising.

Promising conclusion was that the catalyst’s maximum conversion and active metal content had a moderate negative relationship (ρ =-0.54). This would indicate that the conversion decreases as the quantity of active components of the catalyst increases. This question must be overcome to reach the right conclusion. Similar results were obtained in the product yield-reaction time pairs (ρ =0.45). Temperature-pressure, metal content-pressure and temperature-selectivity relationships can also be characterized with moderate correlation coefficient between 0.4 and 0.42.

Accordingly, the correlation analysis can determine that there is a parameter among the selected that must be deleted from the model because it significantly distorts it. Furthermore, the parameter weight should be considered, but this requires further investigation into a larger data set.

4.4.3. EVALUATION OF DNT HYDROGENATION DATABASE

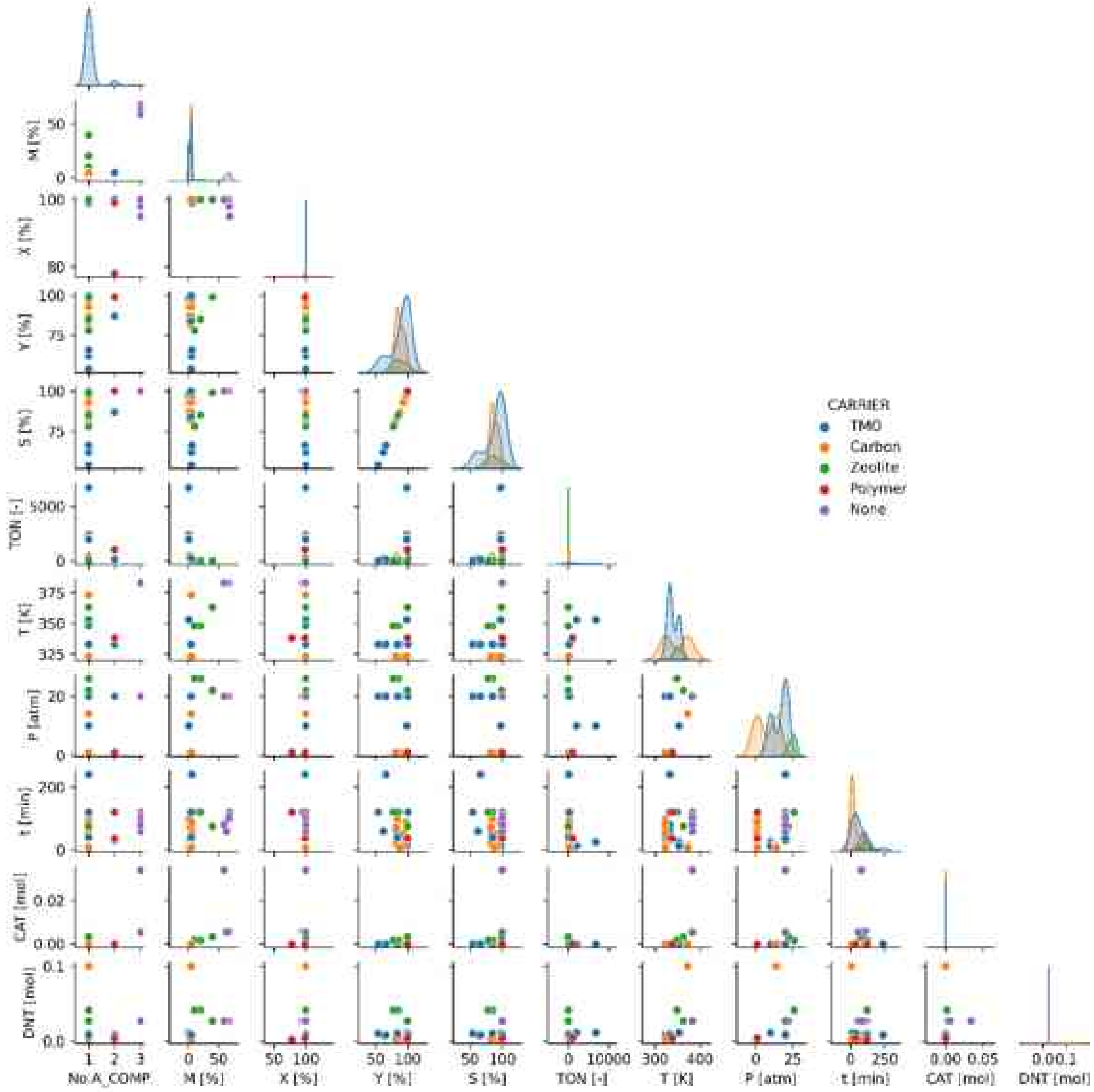


Figure 34 *Pair plot analysis of descriptors by catalyst carrier (TMO-transition metal oxide) [No.A_COMP=number of active component, M =active metal content, X=conversion, Y=yield, S=selectivity, TON=turnover number, T=temperature, P=pressure, t=time, CAT=molar amount of active component, DNT=molar amount of dinitrotoluene]*

A pair plot can confirm or deny the conclusions drawn from the correlation analysis. The selectivity-yield plot clearly demonstrates the linear relationship between the data that is not even rejected by separation according to the catalyst carriers. The high correlation between temperature and the amount of DNT previously mentioned during correlation analysis is no longer so clear. In this case, even after data filtering, there is an outlier in the system, which can have a significant impact on the correlation coefficient.

As regards catalyst support, it is difficult to distinguish patterns because the results are quite different even for specific support. It was also found well-worked transition metal, carbon, polymer, or zeolite-based catalysts. From the point of view of distribution, catalysts supported by TMOs need a lower active metal content and can achieve greater selectivity under certain conditions.

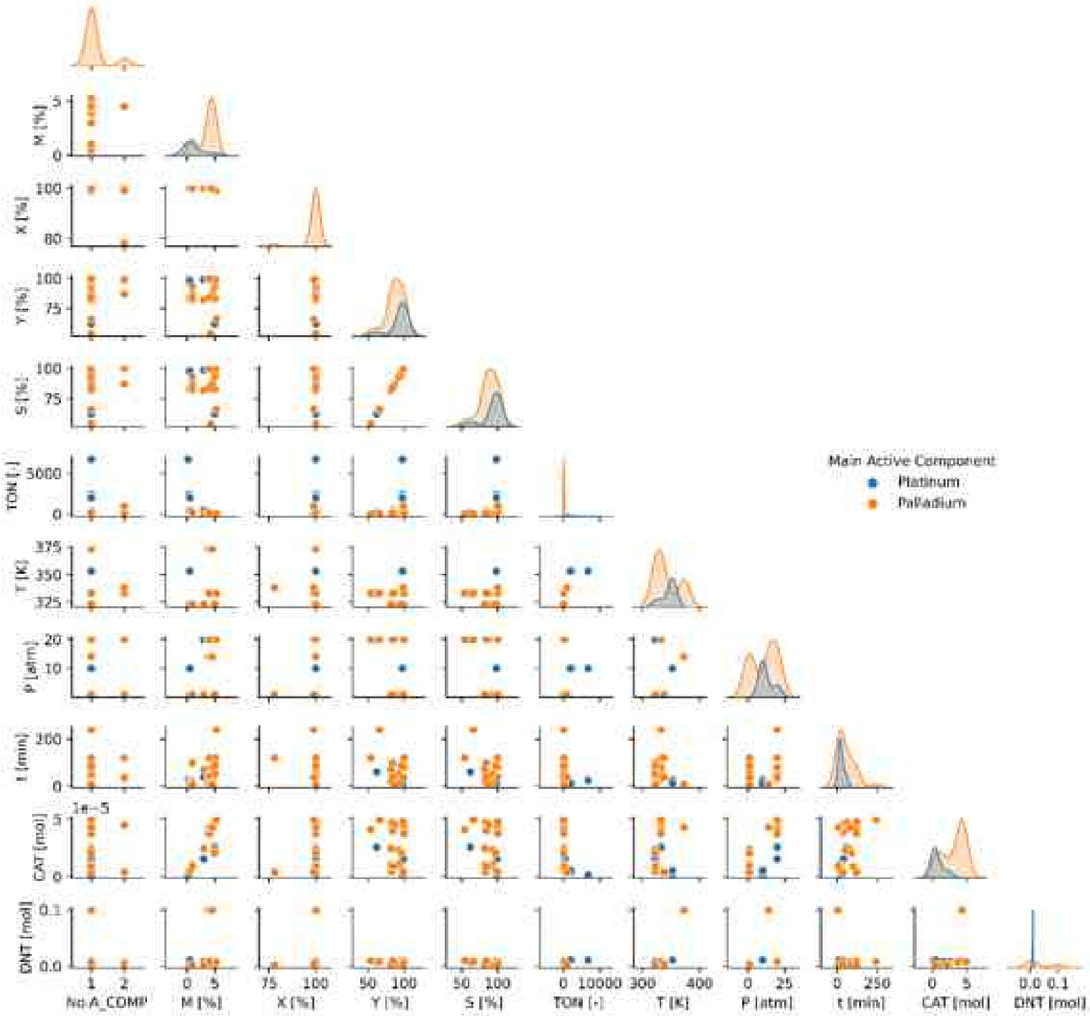


Figure 35 *Pair plot analysis of descriptors by the catalyst's main active component fNo.A_COMP=number of active component, M =active metal content, X=conversion, Y=yield, S=selectivity, TON=turnover number, T=temperature, P=pressure, t=time, CAT=molar amount of active component, DNT=molar amount of dinitrotoluene]*

Because the most common active components of catalysts are palladium and platinum (Figure 35). It is worth investigating the plots of the pair classified by noble metals. Although the joint examination of the two metals and the nickel is technically important, the data analysis of the nickel experiment is not favourable, because the conditions and parameters of the nickel experiment can be completely different. This is an interesting point of analysis that should be considered later. In this figure, it is much easier to classify data points into classes. Platinum-containing catalysts form an independent group at several locations. These catalysts have a lower content of precious metals and can achieve maximum conversion in a shorter time. The performance of these catalysts is excellent. They have a 100% conversion rate and a relatively high TON value.

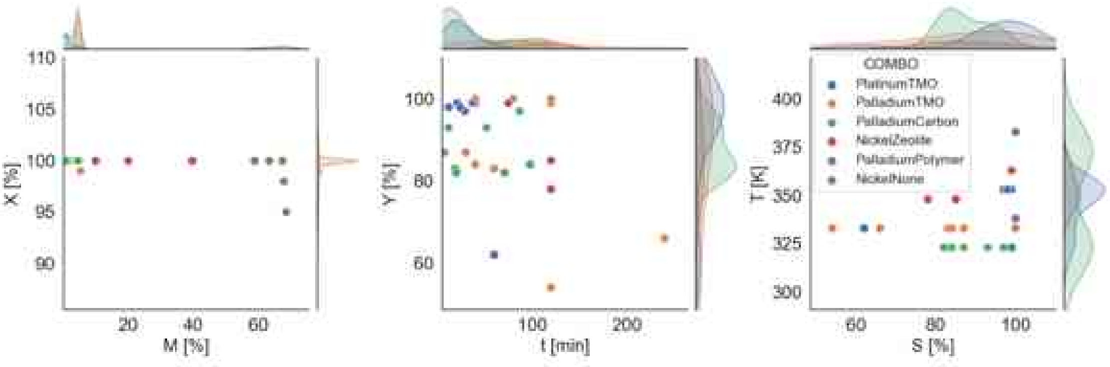


Figure 36 *Joint plots of active metal content – maximum conversion (first), time-yield (second), selectivity – temperature (third) by catalyst combinations (combination of catalyst compositions marked by different colours.)*

Returning to correlation analysis, pair plot analysis helps to better examine pairs that show a certain degree of correlation. 3 parameter pairs were highlighted, the 2D diagram being coloured according to the catalytic system (Figure 36). The three pair plot can conclude that in case of moderate correlation, it is worthwhile to examine the data set more closely, because the correlation may not exist, or a specific parameter causes the correlation. For example, the temperature-selectivity diagram of nickel-zeolite and platinum-transition metal oxide catalysts could show correlation. Given the volume of data, this correlation should be treated with uncertainty.

The more data sets are diverse and the more catalysts they contain, the higher the reliability of results and the better the observed trends can be supported. As a result, the analysis of data continued with a larger database, forming the foundation for catalyst database for the catalytic hydrogenation of nitrobenzene, but with a more extensive set of data, revised descriptor system.

4.5. MIRA23 – THE UPDATED DATABASE

4.5.1. NEW CATALYST LIBRARY

Based on previous experience with the construction of databases and the comparison of catalysts, the updating of databases has begun. In the new database, I have tried to increase the number of catalysts suitable for the hydrogenation of nitrobenzene, using a more in-depth literature research process, which also contains the latest literature results. The new database was modified in accordance with previous experiences both in data collection and in parameter list.

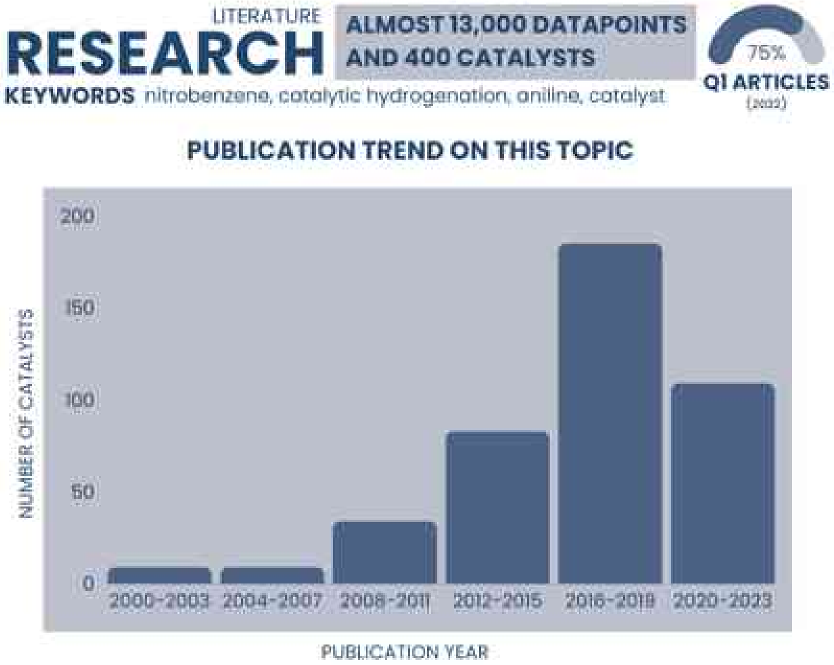


Figure 37 *New database information*

The general information of the database together with the parameter selection are summarized in Figure 37. The literature search resulted in a MIRA library of nearly 13,000 data points and 391 catalysts[136–141,143–148,152–156,158–161,163–172,175,176,178,194,201–315].

In recent years, the number of research on this topic, i.e., the number of people involved in catalyst development, has increased considerably. This is apparent from the publication data. The published data are reliable according to the quality of the journals (75% Q1 articles).

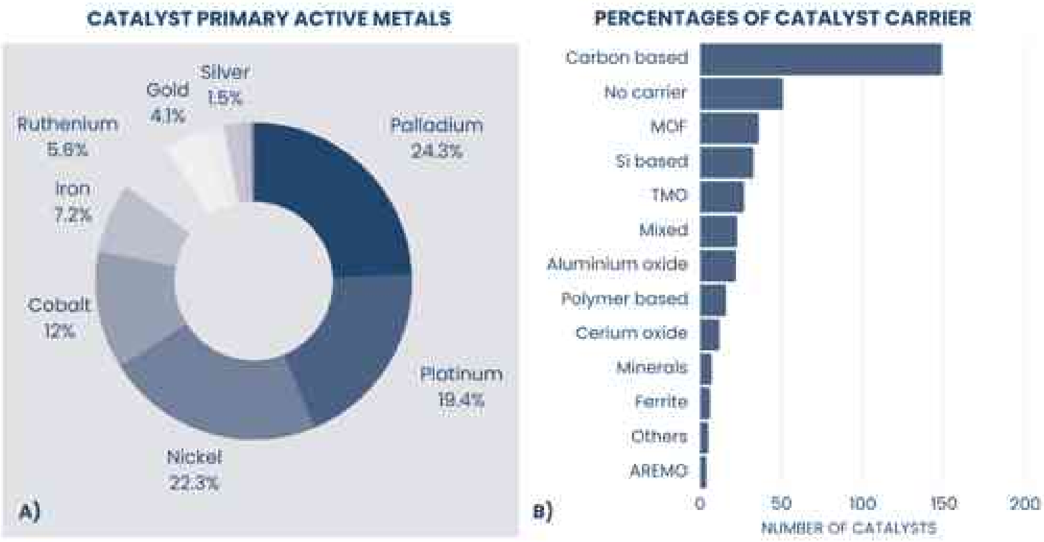


Figure 38 *Composition of studied catalyst [TMO-transition metal oxide, AREMO – alkali rare earth metal oxide, Si based – silicon oxide and SBA-15]*

A database of highly diverse catalytic systems was created and analysed a wide range of catalyst components (Figure 38). Especially for selected reactions, palladium, platinum, and nickel catalysts are still being developed with catalyst supports, mainly carbon-based solutions.

4.5.2. EXTENDED DATA COLLECTION

Based on the previous experience and information, some new parameters have been added to the extended database. Subsequently, according to data processing and data analysis, the MIRA system was modified by comparing the new information with the original descriptor system.

The following new parameters have been added to the database:

1) The TOF value was included in the parameter that describes the performance of the catalyst. The reason for the inclusion of the TOF parameter is also empirical. Within a course, PhD students analysed various reactions in which most of the feedback was related to the TOF parameter. By examining the TOF value in a larger data set, it is possible to avoid it leaving the system due to sampling errors.
2) The catalyst weight is an experimental parameter usually available in literature studies if other information is not specified. The catalyst mass is determined according to the amount used during the catalyst test experiment.

3) The transformation of sustainable parameters has been the most important. The content of the subjectivity and information of the selected parameters is inappropriate in terms of data analysis and its information content was also low. Therefore, it was necessary to modify this parameter class. In addition to existing parameters of the descriptor system, new quantitatively sustainable parameters have been included in data collection. It was characterized stability with quantifiable data and the catalyst price, furthermore three parameters related to catalyst sustainability have been included as new parameters. Bystrzanowska et al. carried out the heterogeneous metal catalysts greenness ranking according to toxicity of pure metals and metal salt, endangered elements, and life cycle assessment point of view[316]. During their research, they created an element ranking using the TOPSIS algorithm, which is also used in sustainability parameters (Table 6).

Table 6 *New stability parameters and definitions*

Notation	Name	Unit	Definition
STAB	Stability	-	From 0 to n, where 0 is no information about stability, 1 means that the stability was investigated, 2,3… the number of cycles until the conversion remains above 90%
Greenness	Toxicity	-	Rank of the active component according to toxicity[316]
	Endangered elements	-	Rank of the active component according to endangered element scenario[316]
	Life cycle assessment	-	Rank of the elements according to life cycle assessment[316]

4) The estimated catalytic cost has been added to the system based on the active component to emphasize the industrial applicability. The catalyst price is given on the basis of the number of active components in a gram of catalyst.

4.5.3. CORRELATION ANALYSIS

Previous correlation studies have shown the strength of relationships between individual parameters. Afterwards, a correlation analysis was carried out on the expanded catalyst database designed for the hydrogenation of nitrobenzene. The analysis was conducted in the same way as the previous one, on the selected appropriate parameters. In advance, a violin plot with strip plot were prepared that would be suitable for examining the distribution of the data set.

Prior to the analysis of correlation data, the data set was statistically analysed. Based on this, parameters of low relative frequency were filtered that do not participate in correlation analysis. Product yield was excluded from the analysis according to the previous tests correlation result. Another difference is that the TOF, the catalyst mass, and the catalyst price are included in the analysis.

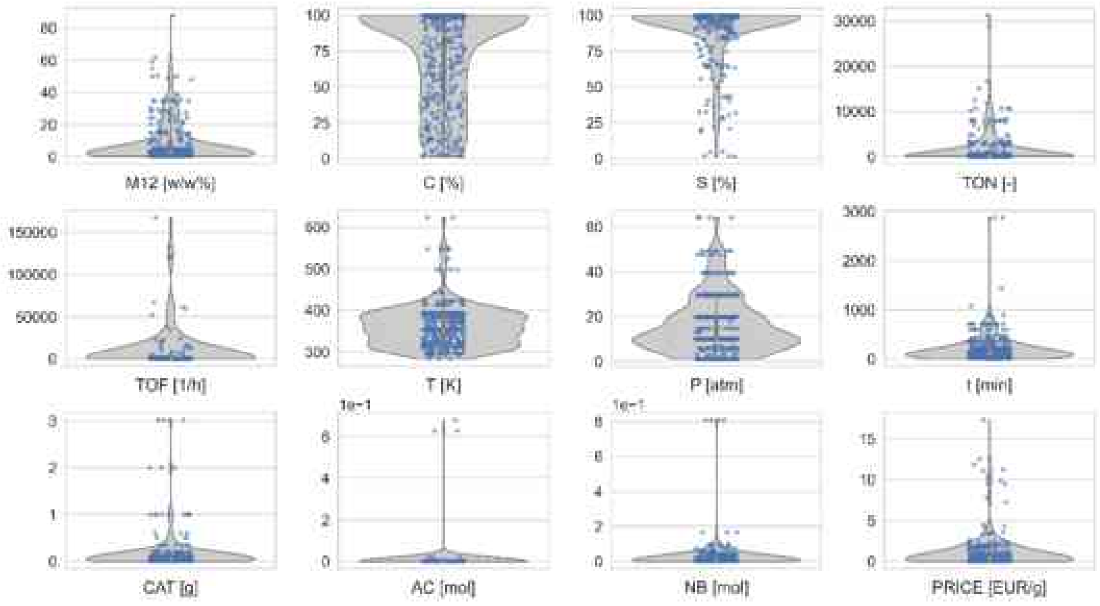


***Figure 39** Combined violin and strip plot of the parameters in nitrobenzene hydrogenation database [M12 = main active component, C=conversion, Y=yield, S=selectivity, TON=turnover number, TOF=turnover frequency, T=temperature, P=pressure, t=time, CAT=weight of catalyst, AC=molar amount of active component, NB=molar amount of nitrobenzene, PRICE=catalyst price according to the active component]*

Figure 39 shows the distribution of the series of applied catalyst data by parameter. Because of the analysis of the data set of individual parameters, most parameters have deviations that must be removed for statistical analysis. The outliers of the data set were filtered with interquartile ranges (IQR), and the general rule is that the outlier is an observation that falls below the 25th percentile – 1.5 *IQR or above the 75th percentile + 1.5 * IQR[317].

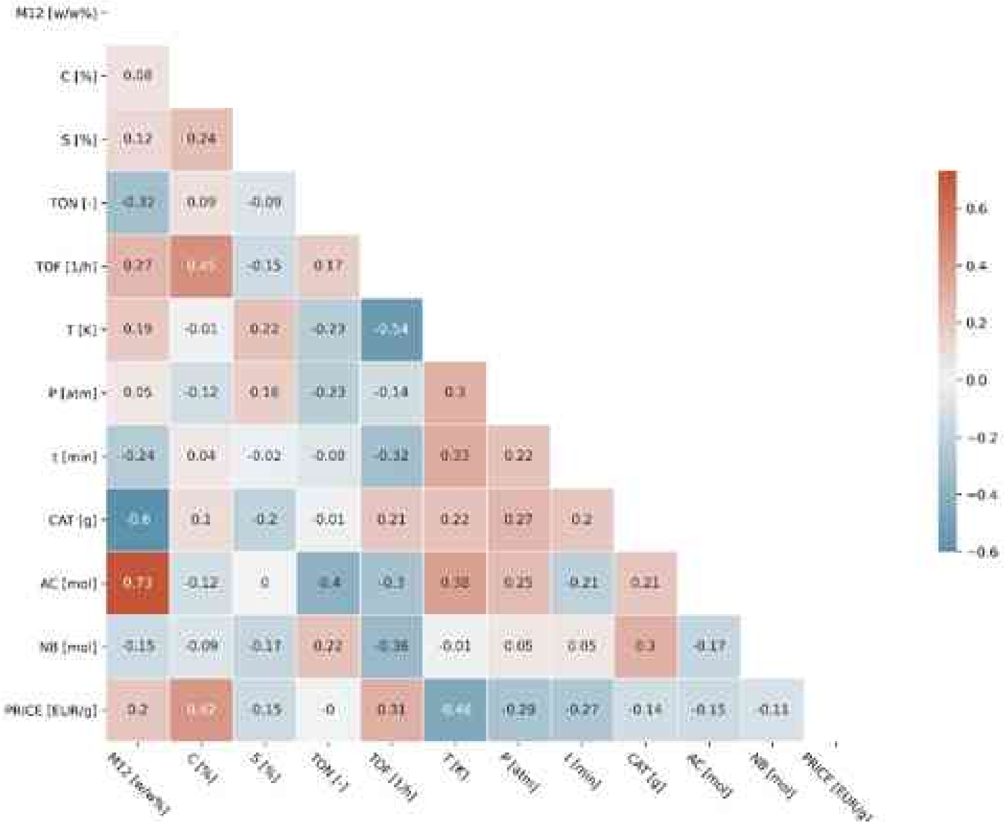


Figure 40 *Correlation analysis after data filtering of nitrobenzene hydrogenation database [M12 = main active component w/w%, C=conversion, Y=yield, S=selectivity, TON=turnover number, TOF=turnover frequency, T=temperature, P=pressure, t=time, CAT=weight of catalyst, AC=molar amount of active component, NB=molar amount of nitrobenzene, PRICE=catalyst price according to the active component]*

Data were not normally distributed, regardless of data filtering, so the correlation analysis of parameters was performed with Spearman's correlation analysis (Figure 40). Only parameters that the data set was complete or almost complete are included in correlation analysis. Looking at the filtered data set, it can be observed that, statistically, the descriptor system has one strong correlation relationships.

This was only because the active component w/w% (M12) was also included in the data analysis and must be related to the molar quantity of active components (AC) and the weight of catalyst (CAT). Furthermore, TOF values had a moderate correlation with most parameters. This was possible because there are few parameters available from TOF values and the distribution was not representative.

Most of the parameters showed a weak correlation. The moderated correlation parameter pairs will be show later. There was a moderate correlation between temperature and price, as well as between active components and time. Moderate correlation coefficients were also obtained for the conversion and price relationships, TON, and the quantity of active components.

4.5.4. MODIFIED DESCRIPTOR SYSTEM

Based on experience gained during the construction of the catalyst library and the correlation analysis, the catalyst ranking MIRA 2023 were characterized with a descriptor system of 13 parameters and calculated the MIRA23 number from it as a fresh method.

The yield parameter has been omitted based on the results of MIRA21 correlation analysis. With respect to data frequency, the performance of heterogeneous catalysts can be well characterized by TOF values but is lower than other (26%) characteristics and the correlation results show moderate connections, this parameter has also been omitted from the comparison. The three parameters of the catalyst greenness were also included in a single parameter (forming an average).

Figure 41 *MIRA23 descriptor system*

Figure 41 shows the modified descriptor system, which identifies catalysts in a single, comparable quantifiable data. Using the 13-parameter descriptor system, catalysts were analysed according to the MIRA23 model, ranked, and classified them into categories.

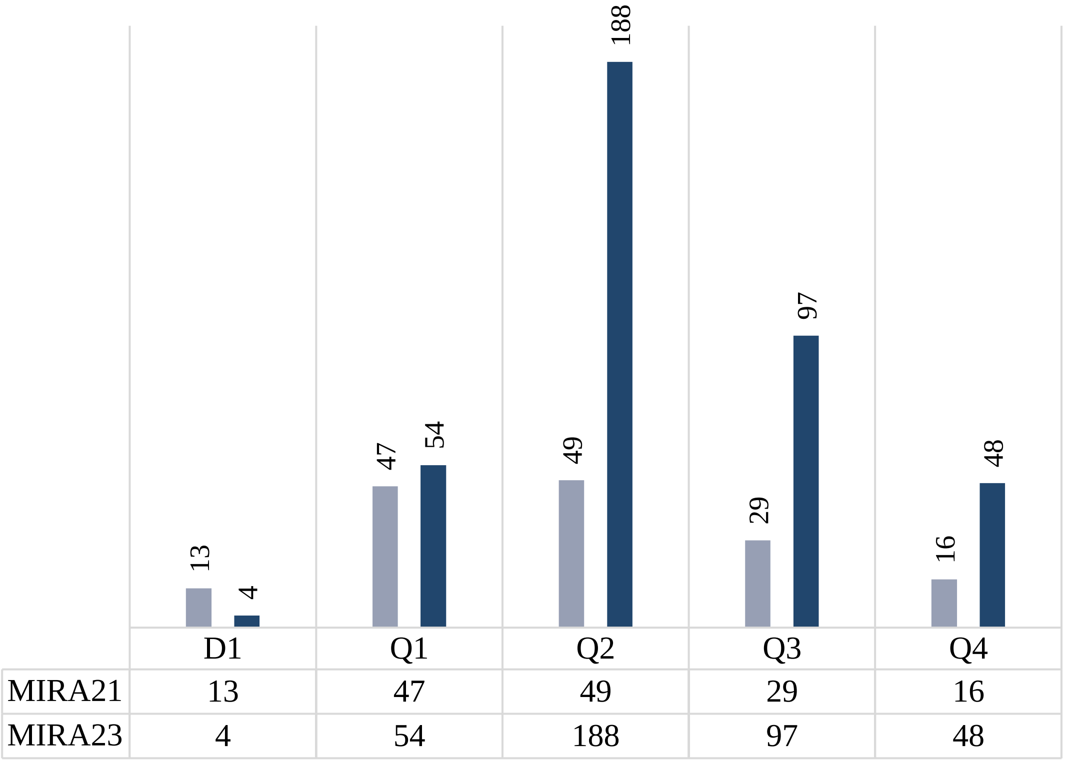
Table 7 *Detailed description of 13 parameter descriptor system*

Descriptor system						
Cat.		**No.**	**Notation**	**Name**	**Unit**	**Definition**
Catalyst performance	I.	1.	X_{REAmax}	Maximum conversion	n/n%	Maximum reactants conversion achieved on a given catalyst
		2.	S_{PR}	Product Selectivity	n/n%	Product selectivity for maximum conversion
		3.	TON_{PR}	Turn Over Number	-	Number of moles of product formed per 1 mol active metal when the maximum conversion reached
Reaction conditions	II.	4.	$Tmax_{conv.}$	Temperature	K	Reaction temperature for maximum conversion
		5.	$P_{max.conv.}$	Pressure	atm	Reaction pressure for maximum conversion
		6.	$t_{max.conv.}$	Time	min	Time required to reach maximum conversion
		7.	$n_{cat.}$	Molar amount of initial catalyst	mol	The molar amount of the active metal involved in the reaction - in case of several metals, the sum of molar numbers
		8.	n_{start}	Molar amount of starting reactant	mol	The initial amount of starting reagent involved in the reaction
Catalyst conditions	III.	9.	CPS	Catalyst Particle Size	nm	Average particle size of the catalyst
		10.	CSA	Catalyst Surface Area	m^2/g	Catalyst (active metal + support) surface area
Sustainability parameters	*IV.*	*11.*	*STAB*	*Stability*	-	*From 0 to n, where 0 is no information about stability, 1 means that the stability was investigated, 2,3... the number of cycles until the conversion remains above 90%*
		12.	*Greenness*	*Toxicity*	-	*Rank of the active component according to toxicity*[316]
				Endangered elements	-	*Rank of the active component according to endangered element scenario*[316]
				Life cycle assessment	-	*Rank of the elements according to life cycle assessment*[316]
		13.	*PR*	*Price*	*euro/g*	*Price is of 1 gram catalyst according to the active component concentration in euro*

Table 7 shows the detailed description of the modified descriptor system. As it can be seen, among the sustainability parameters (light blue colour) of catalysts, toxicity, endangered elements, and life cycle assessment categories, since they can be evaluated on the same scale, it had characterized them with quantifiable data such as the average value and identified them with the name "Greenness".

4.5.5. MIRA23 RANKING

From the normalized and weighted data, the MIRA23 number were calculated, which characterizes the system together with the number of known parameters. In this case, the model is applied completely identical to MIRA21, and the data are not filtered.



	D1	Q1	Q2	Q3	Q4
MIRA21	13	47	49	29	16
MIRA23	4	54	188	97	48

Figure 42 *Classification of catalyst with MIRA21 and MIRA23 model*

Analysing 391 catalysts, 4 were classified into D1 and 54 into Q1. These catalysts have 11 or 12 parameters available. The complete list can be found in the appendix (Appendix 6). Figure 42 shows the comparison of the first database with the newer and larger database. The figure shows that as the data amount increases, classification approaches the normal distribution much better, thereby increasing the representativeness of the sample.

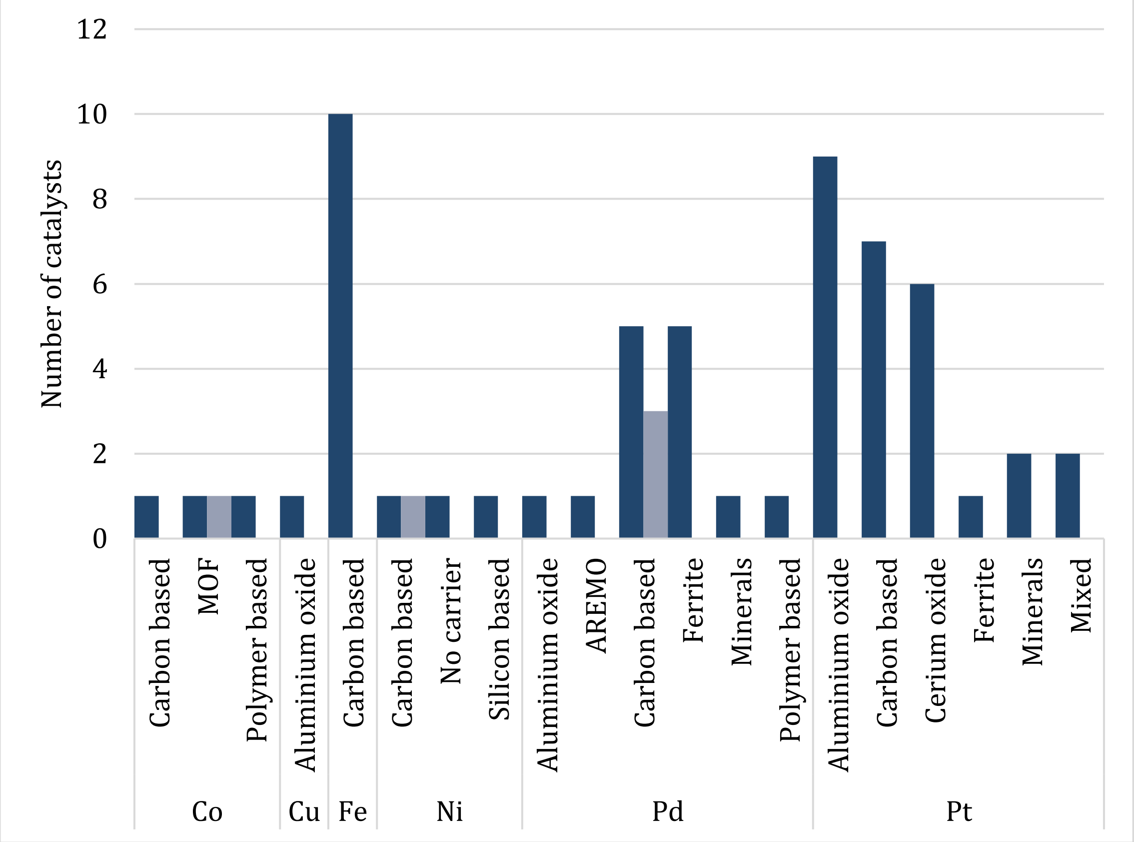


Figure 43 *Q1 Catalytic systems of MIRA21 ranking, dark purple shows the main active metal, light purple shows the second active metal [MOF=metal organic framework, AREMO=alkali rare earth metal oxide.]*

Of almost four hundred catalysts, 58 received such high marks that they entered the first quarter. The following diagram shows the composition system of the Q1 catalyst (Figure 43). On the x axis, the results are grouped by active component and catalyst carrier, allowing easy access to combinations that have obtained good Q1 certification.

The results demonstrate that most of the catalysts that get best results are carbon based iron and aluminium oxide-based platinum catalysts, in addition to platinum or palladium based on carbon and platinum based on cerium oxide. Since each row represents a separate catalyst, the quantitative feature in this case indicates that the combination of a catalyst carrier and catalyst shows really good results with several parameter settings.

About 40% of catalysts contain carbon-based support, 40% of which is also N-doped porous carbon, and there are also a significant number of carbon nanotube catalysts. In addition, active carbon, carbon black and rarer carbon foam and carbonated cellulose have also achieved excellent results in the hydrogenation reaction of nitrobenzene.

Q1 catalysts have an average conversion rate of 91% and a selectivity rate of 92%, with an average TON value of 3012. Laboratory experiments were usually conducted between 300 and 400 K, with the pressure applied already on a larger scale (1-50), but typically on 14 atm. The average reaction time was 144 minutes, but some catalysts achieved almost 100% conversion time in 8-10 minutes[299]. In most cases, the stability tests of catalysts have been successful and more than 90% conversion has been achieved by the tests over 15 cycles[158]. The catalyst price is about €0.63/gram depending on the active component. In the case of palladium and platinum, active components are used in all forms between 0.07 and 5 w/w%.

4.5.6. CATALYST COMPOSITION OPTIMIZATION

Figure 44 *Multi-criteria decision making options*[318] *[AREMO – Alkaline rare earth metal oxide, No Ac – No active component, MOF – Metal organic frameworks]*

Based on the MIRA23 model and classification, promising components can be determined for catalytic design and research. If confronted with an industrial or academic decision-making task aimed at choosing the right catalyst, several data-related challenges would be encountered. There is a multiparametric system on which the catalyst must be selected based on the criteria. The parameter units are not uniform, the target direction is opposite. It does not necessarily know the statistical characteristics of the descriptors.

To resolve the difficulties and to solve the catalyst selection task, there are several multi-criteria decisions making (MCDM) options (Figure 44) related to the procedure of MIRA23[318]. In the satisfaction degree method, the value of each parameter is set to a level that it cannot accept above or below, depending on the direction of the parameter (minimum or maximum). By Maximin and maximax techniques, normalized but unweighted data sets are considered for each catalyst. In the case of maximin method, the best alternative is selected from the worst options, while in case of maximax method, the best of the best options is selected. The lexicographic methodology requires a clear order of importance, which may be difficult even for the decision maker. There are also several weighting methods that can help decision makers in the right direction, such as the SMART or Analytic Hierarchy Process method[319].

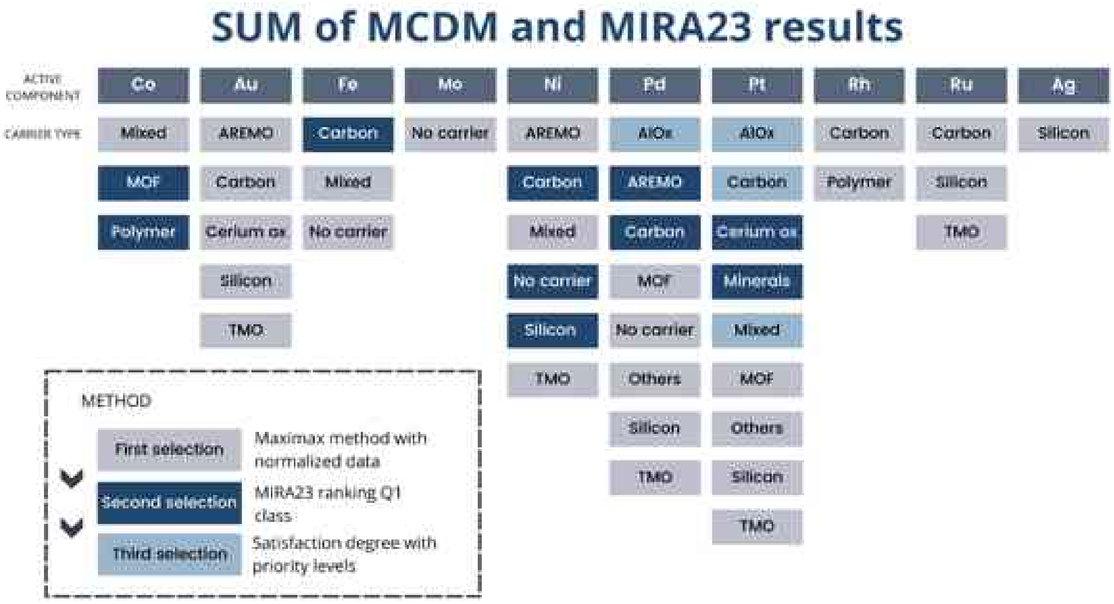


***Figure 45** Comparison of MCDM and MIRA23 results*

MDCM is an extremely complex scientific field, including many other methods. My goal was to compare the results of MIRA23 with the relevant methods and to draw further conclusions from it. The results are summarized in Figure 45. Using the maximax method, the normal data series were selected, placed them in the order of descending, and then selected the best catalyst. The results showed that there were 157 catalysts of this type, which the first selection was considered. The above combinations are all part of the maximax results. The method of satisfaction degree was also carried out, where different levels for each parameter were set, resulting in 13 catalysts. the minimum conversion (88%), selectivity (98%), TON (337), the maximum price (0.49 EUR/g) and time (240 min) were determined, adjusted for the average.

In the diagram, cells marked in dark blue give the most suitable catalyst combination by the maximax and MIRA23 method, while light blue cells provide the most suitable catalyst combinations based on three methods. Based on these, the best is proven to be the aluminium oxide supported palladium or platinum catalysts, as well as carbon based platinum and mixed supports with platinum.

4.5.7. ROLE OF WEIGHTING

The parameters describing the catalyst were first weighed based on professional experience. It was then examined whether the correlation between the individual parameters is worth changing the weighting. As a result of correlation analysis, the descriptor system was modified, but maintained the weight. The MIRA23 weighting system was also tested, in which the weights were changed and examined their effects on ranking. As a result, the priority ranking for performance (PER), sustainability (SUS) and reaction conditions (COND) has been completed. The individual parameters were weighted from 1 to 10, and according to different preferences, increasing scores were given to more parameters.

Before the test, outliers were removed and reduced the possibility of result distortion. The data were standardized between 0 and 1, so that they were independent of the measurement unit and on the same scale. The weights were set to be so that their sum is 1(Eq. (8)):

$$S_i = \sum_j w_j * x_{ij} = 1 \tag{8}$$

where w_j the weight of the j component, and x_{ij} the values of the ‘j’ parameter in case of ‘i’ catalyst. The sum of the normalized values multiplied by weight gives the catalyst point.

To see the weight effect of a parameter class, two levels were set as weight and change them according to the parameter class. In the preliminary version, weight was not applied, in the second case performance was preferred, in the third case already reaction conditions and in the fourth case sustainability parameters. The main result is shown in Figure 46. For this figure, the best 58 catalysts were used from different weight cases that formed the Q1 class in the previous one to make the comparison authoritative.

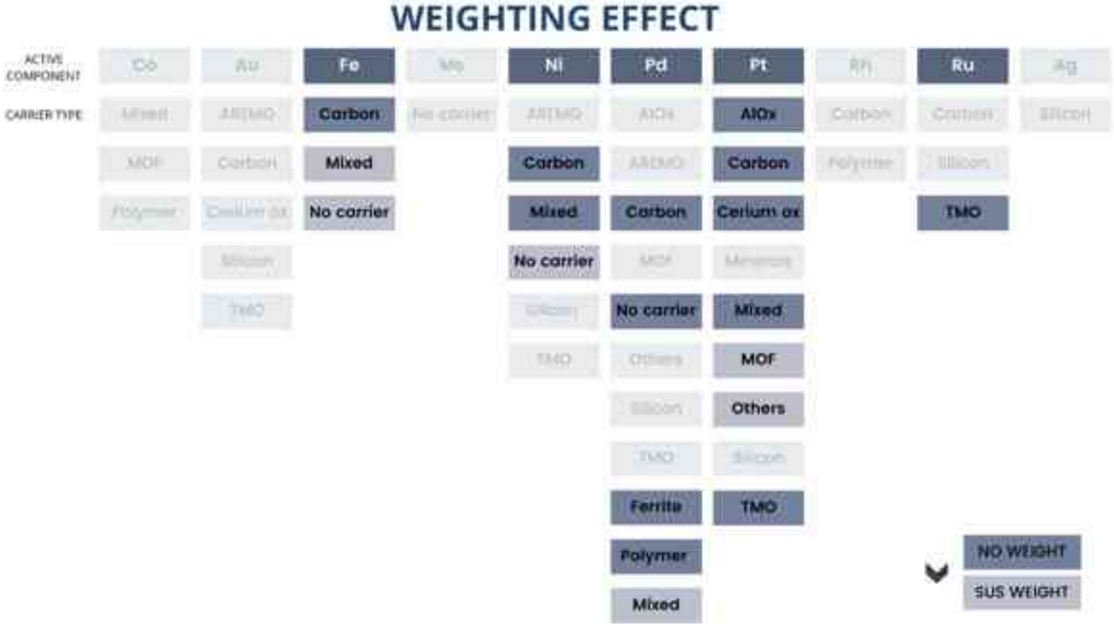


Figure 46 Effect of weighting (SUS weight – sustainability parameters are preferred)

Compared to the previous MCDM comparison, catalyst compositions were marked that were not included in the Q1 class of the new ranking in light grey. It was marked in light blue the catalyst combinations typical of the Q1 unweighted class. Those that have appeared in the case of sustainability are in light green. The other two weight cases cover the other two. Compared to the original MIRA23 weighted system, the biggest difference is that catalysts containing cobalt have been eliminated, but ruthenium catalysts have been included among the best.

Even from this diagram, the weighting of sustainability parameters has a significant impact on ranking and classification. To verify this, the catalysts were compared on the basis of their CAT ID number, divided into specific catalyst units. In the first three weighting systems, the agreement is 88–97 % in the Q1 class, whereas in the case of sustainability it is about 20 %. On this basis, the descriptor system can be said to be the most sensitive to sustainability among the four parameter classes.

4.5.8. PATTERN RESEARCH

In the field of data mining, ongoing work has focused on patterning and classification processes. In this process, joint plots were created, and extracted important information from the data. Consequently, in the search for patterns, the relationship between parameters were investigated and the influences of catalyst output parameters on input parameters.

For two-variable tests, a scatter plot was first prepared that contained all two-variable variations and their results in a matrix. The appendix contains the distribution matrix presented according to the active component (Appendix 7.)

The joint plot diagram is graphical representation that are used to visualize the relationship between two numerical variables. In each diagram, two variables appear on the x- and y axes, and the third analysis aspect is coloured. This usually means categorization of active components or categorization of catalyst carriers. All parameter combinations were prepared, and subsequently, an assessment was made to identify trends for each combination. The objective was to determine if an apparent grouping could be created based on the catalyst composition. Of the more than 300 figures, only a select few are emphasized. These figures serve the purpose of either reviewing the results of correlation analysis or illustrating specific patterns.

Figure 47 shows the relationship between the conversion and the concentration of the active component of the catalyst. The diagram is classified by active component quality. When drawing filtered data sets, there is no clear correlation in the data, but the colour of several groups in both cases is very well separated.

Figure A demonstrates 4 distinct groups as concentration functions. The largest data set group used a low concentration of active components, and the results were extremely dispersed in terms of conversion values. This includes most catalysts containing palladium or platinum. The following group consists of cobalt catalysts, where the concentration of the active components used is higher, but conversion has completely different values. The third group is composed of nickel catalysts with an active component content greater than 10w/w%, but these have a particularly low conversion rate. The fourth group is composed of catalysts with high concentrations of nickel or iron and achieves high conversion. For groups, the number of elements is decreasing, and the standard deviation is decreasing. This leads to the conclusion that, in the case of palladium and platinum catalysts, the concentration of active metals is not a decisive factor in conversion. On the other hand, even in different parameter sets, high conversion can be achieved with iron-containing catalysts. The data distribution is presented by the distribution functions of the diagrams on both sides, in which case the most common element distribution is displayed.

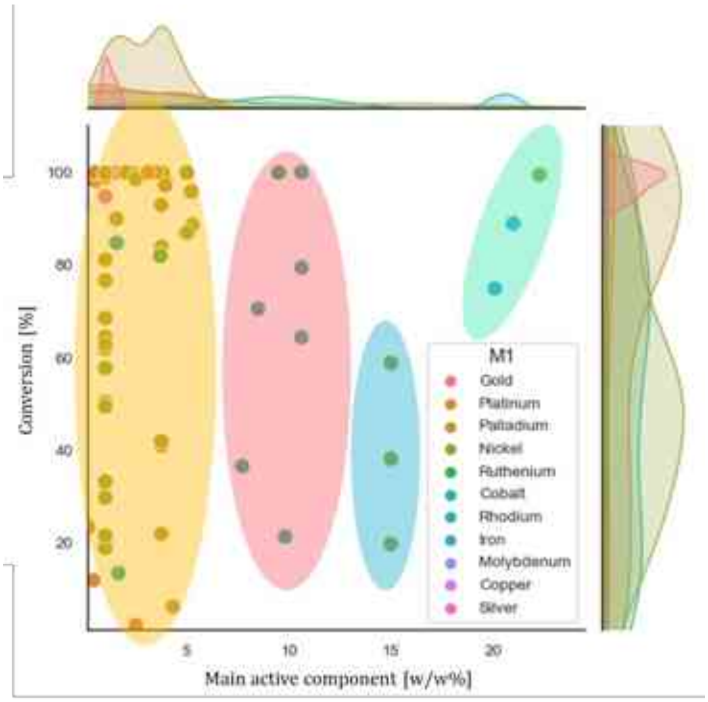


Figure 47 *Joint plot of Main active component concentration (w/w%) and Conversion classified by catalyst active component (M1)*

The same four groups are visible in the catalyst support. The second three groups are almost completely homogeneous, indicating that the catalyst's active components and support pairs have produced these results. The second group consists of cobalt catalysts based on polymers, the third group consists of nickel catalysts without support, and the fourth group consists of iron and nickel catalysts based on carbon. Of course, it also seems that, with low active metal concentrations, most palladium and platinum catalysts have achieved nearly 100% conversion.

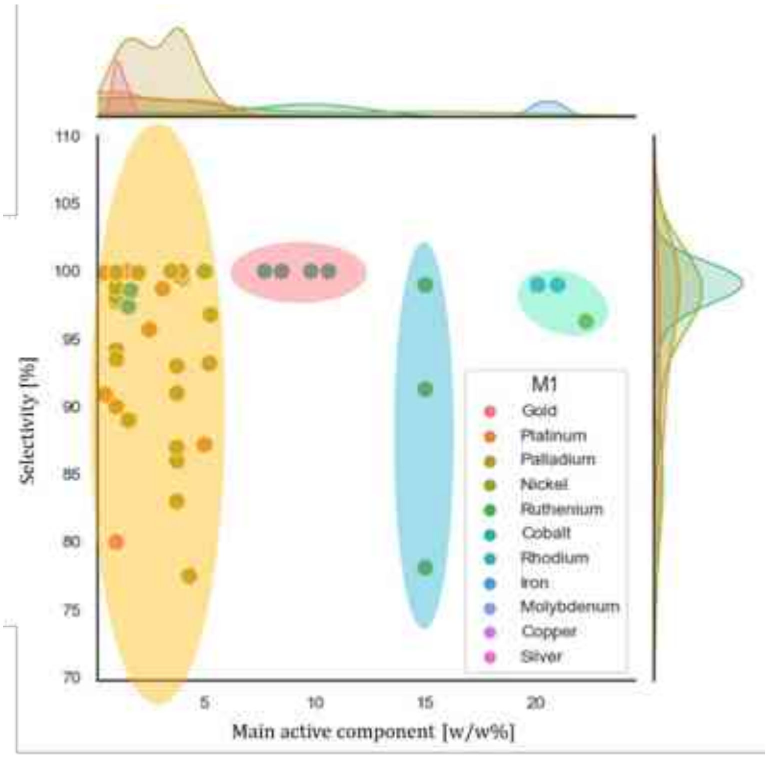


Figure 48 *Active component concentration vs. selectivity results*

The distribution of this group as a selectivity function was also examined, and found that the results were completely similar, with the only difference that the selectivity function here is much smaller ranging from 80 to 100 % (Figure 48).

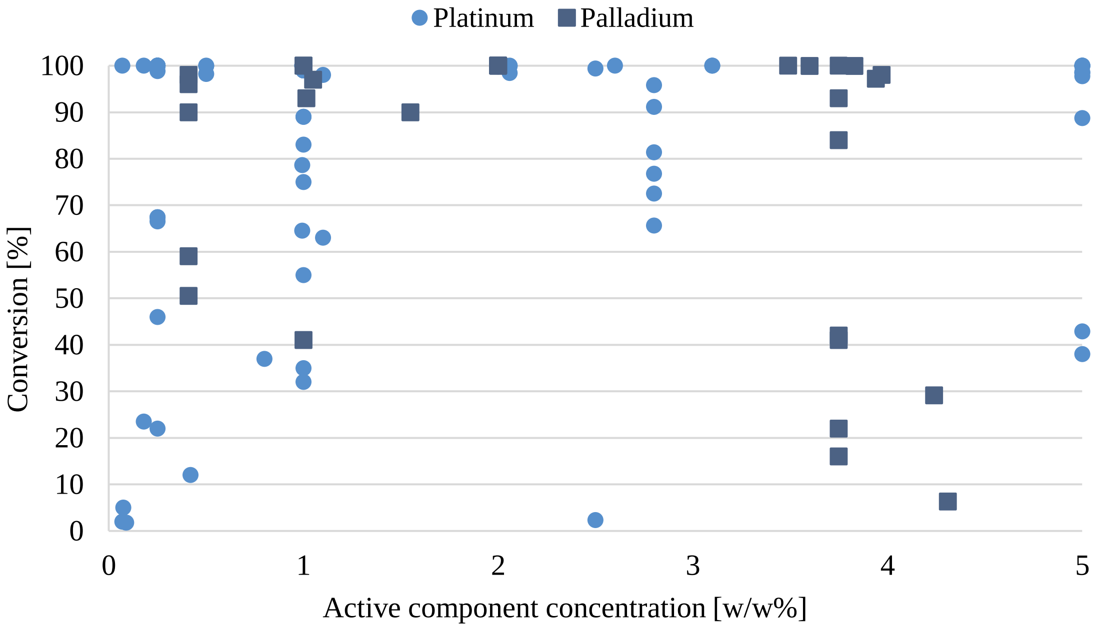


Figure 49 *Palladium and platinum catalyst conversion results according to active component concentration*

As previously highlighted, due to the inability to draw any further conclusions in the case of the most used catalysts, palladium and platinum, the following figure shows the conversion values obtained as a function of the concentration of the active components of the catalyst (Figure 49). The palladium catalyst produced typically has a palladium concentration between 0-1 w/w% and between 3.5 and 4.5 w/w%. In platinum catalysts, the typical concentrations are between 0-1 and 2-3 w/w%, but there are also catalysts with a platinum content of 5 w/w%. The conversion results show that there is an extremely large standard deviation for different concentrations. The impact of additional parameters on conversion is important.

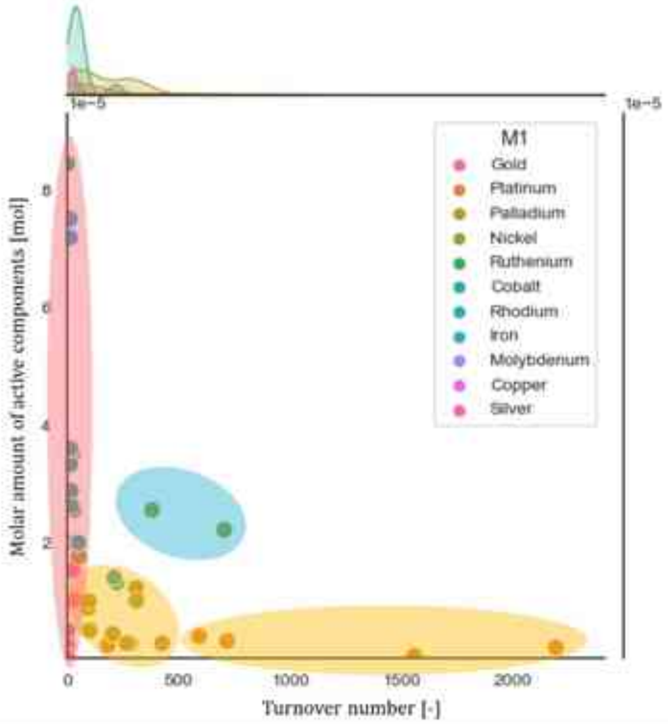


Figure 50 *TON-Active component joint plot results sorted by active component type*

The paired correlation coefficient of the parameter TON and the quantity of active component material was -0.4. Negative correlation means that the larger one value is, the smaller the other value. The size of the coefficient shows a moderate correlation. When you look at the filtered data set, this result is partially recognizable (Figure 50). This can be explained by the fact that the value of TON itself is inversely proportional to the amount of catalyst. However, correlation is influenced by additional values such as initial nitrobenzene amounts and selectivity.

It can also be seen that there are different results according to active components, although the limits are less clear. It should be noted that among platinum catalysts there are catalysts that have achieved outstanding TON values with low active component content.

It can be also forming clusters in the TON-AC component diagram. Cobalt-type compounds form a group (green) with a very low number of TONs, but with variable active component quantity. The next clearly distinguishable group (yellow) are catalysts with a low platinum content that can be characterized by high TON values. And between them (red and blue) there are two other groups. The group marked in red clearly contains catalysts that contain nickel, while the other group is much more diverse, but mainly contains palladium.

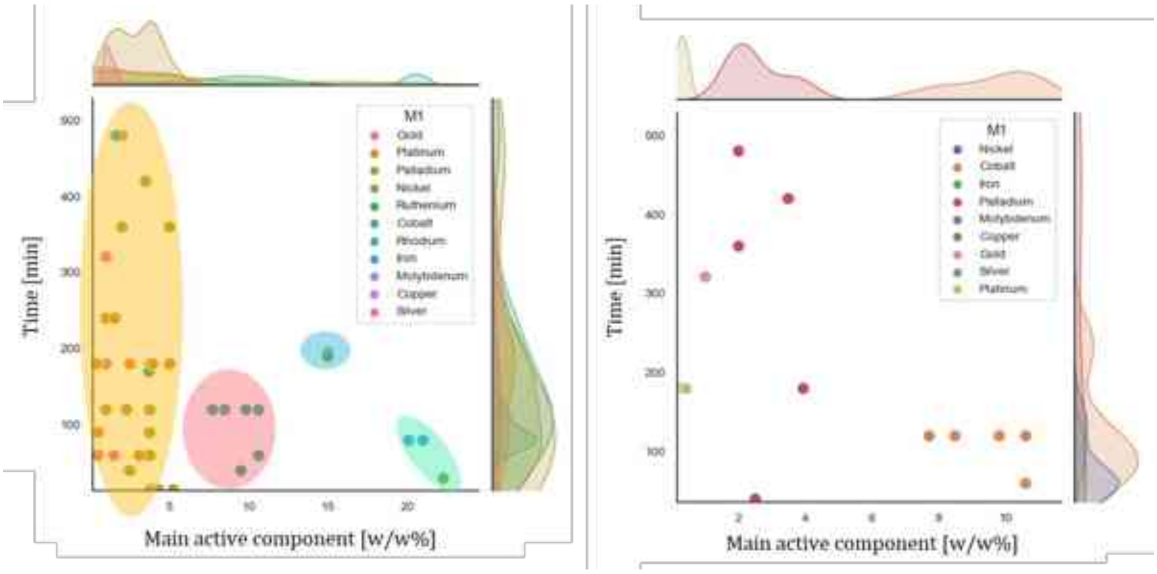


Figure 51 *Active component concentration versus reaction time results*

Figure 51 shows the concentration of the active component and the reaction time. The difference between the figures is that a single filter was used in the first figure, a data filter was used twice in the second figure. The data were filtered using the same statistical method.

After the first filter, four groups are also visible, which were first examined in the confined diagram according to the active component. The two different groups in the second figure are groups of about 8-10% cobalt catalysts with a reaction time of less than 150 minutes. In addition, Palladium red data points are visible, which are very dispersed in terms of reaction time, with an active component concentration of 2 to 4 %.

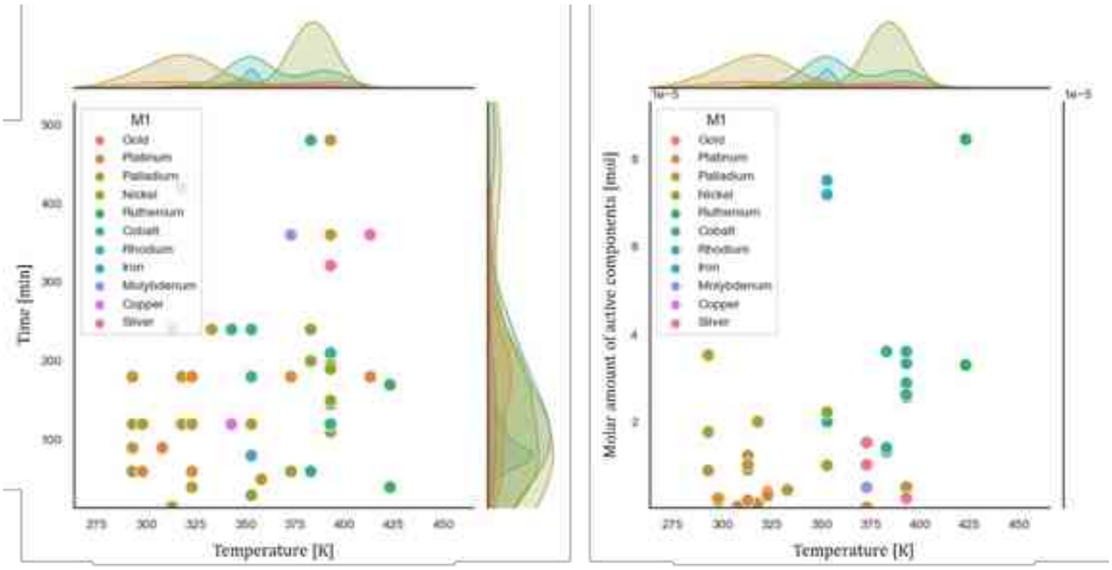


Figure 52 *Temperature joint plots vs. time, active component*

The correlation coefficient between the applied reaction temperature and the reaction time parameter was +0.33 (Figure 52). The relationship between the two parameters is not clear and unfounded. At higher temperatures, the range of response time is expanded. Another questionable correlation is that between active components and temperatures (ρ=+0.38). On the right, different reaction temperatures are typical depending on the quality of the catalyst involved. In this case, trends depend on composition, so it is difficult to observe.

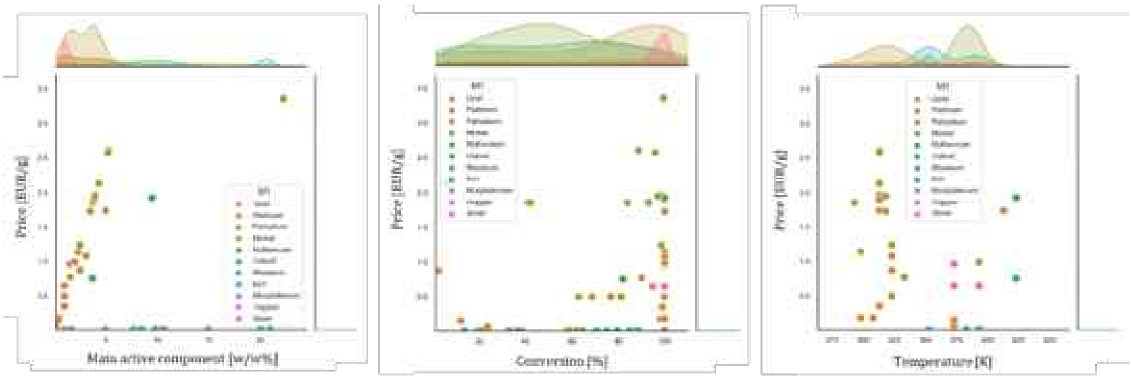


Figure 53 *Active component concentration, Conversion [C], Temperature [T] vs. price*

The following table shows the relationship between the price and the three highlighted parameters (Figure 53). In the first example, a linear relationship between the content of catalyst active components and the price based on the quality of the catalyst active components can be seen. Based on spring data of the year, catalysts containing gold, palladium and platinum are most cost dependent. The middle figure shows the correlation between conversion and catalyst price. Catalysts that achieve 100% conversion, mainly palladium and platinum catalysts, have great potential for cost optimization. Some well-functioning catalyst converters have been tested, and they are associated with a very high price. The third figure also aims to examine the results of correlation analysis. There is no direct correlation between the catalyst price and the reaction temperature used. Examining together with the medium figure can find catalyst compositions that achieve high conversion at a certain temperature and at a low price. Lower-cost palladium and platinum catalysts achieved a conversion rate of about 100% between 300 and 350 K, while gold, cobalt and ruthenium catalysts were similar at a higher temperature of 375-425 K.

In the analysis, it has happened several times that the distribution function of the joint plots has not been shown or appears as a needle. This can be due to the characteristics of the data distribution.

4.6. OUTLOOK

After the database data analysis developed during our work, an attempt was made to use the investigated nitrobenzene database for machine learning purposes. Our goal was to estimate the value of the output parameter by using a subset of the collected parameters as input parameters.

The process was shown in Figure 54. Following data collection and analysis, data cleaning and filtering was performed, then the input and output parameters were determined for machine learning. We selected the parameters based on the previous data analysis results of MIRA (catalyst performance, reaction condition and catalyst composition parameters).

Training and test datasets were generated, and various linear and nonlinear algorithms were used during model building to find the most suitable predictive model.

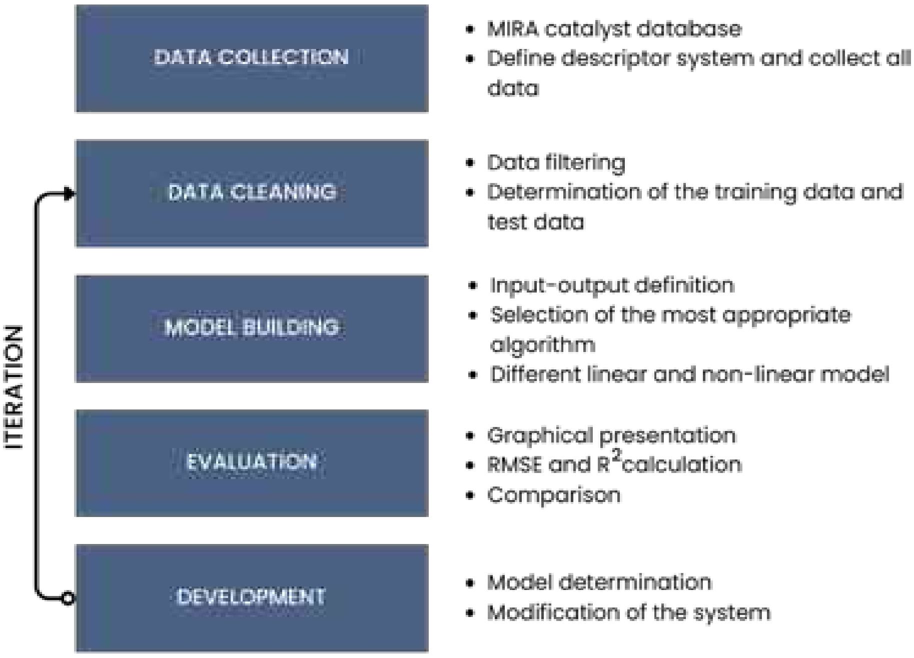


Figure 54 Application process of machine learning for the MIRA database

During the evaluation, the results were graphically presented and RMSE (root-mean-square error) and R^2 values were calculated. The results from a professional perspective were assessed and further development opportunities were discussed for the supervised machine learning method. In the next phase, our initial database was returned to, and the development process was initiated once again.

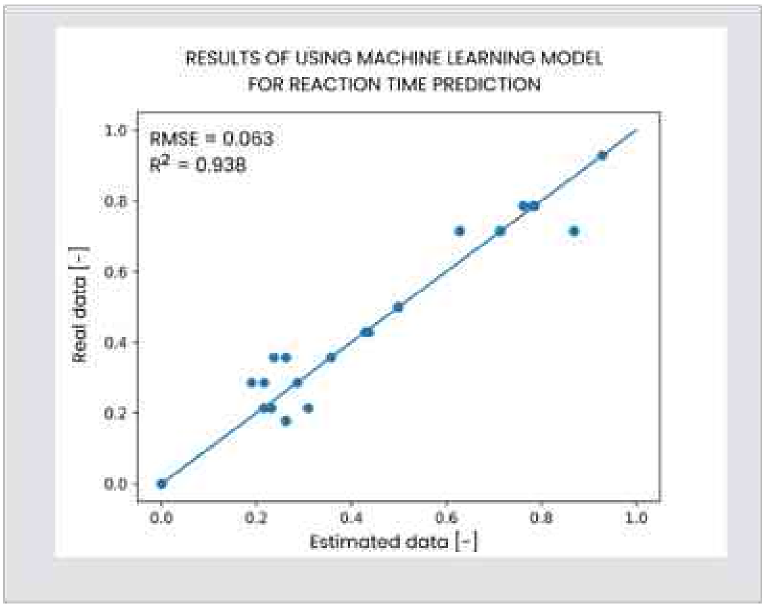


Figure 55 Using of machine learning for prediction of reaction time

Continuing and constant refinement is being carried out beyond the doctoral research. Increasingly promising results are being achieved through the continuous fine-tuning of machine learning.

One of the promising results from our second test is being illustrated in the Figure 55. My goal is to utilize this example to showcase the application potential of the database in machine learning. In the scatter plot, normalized values are indicated. The straight line in the diagram represents the linear relationship and perfect correlation between the real data and estimated data according to our input parameters. The better the point fits into the line, the better the prediction, i.e. the input parameter can predict what the output parameter is and how long the reaction time is.

By applying machine learning in catalysis research, we have gained access to new and substantial amounts of data, the evaluation of which remains a priority task in the future. Evaluating new information not only expands our existing knowledge but also raises additional questions regarding the efficiency of our model and data collection process. The depth of this understanding enables us to enhance our machine learning model, fine-tune parameters, and improve predictive performance, thereby facilitating catalyst design and research processes.

Utilizing the vast amount of data provided by machine learning helps uncover connections and patterns within the data, expanding our knowledge and more effectively applying information to solve real industrial problems.

5. SUMMARY

JOURNEYING FROM TRADITIONAL LITERATURE RESEARCH TO MACHINE LEARNING

The phrase best summarizes the last four years of my travel during my doctoral work. My doctoral research commenced with the development of aromatic nitro compounds catalytic hydrogenation processes and the exploration of corresponding hydrogenation catalysts. Among the traditional literature research, numerous inquiries arose, leading to the initiation of data collection.

The process of data collection evolved into a comprehensive catalyst database. Data comparison transformed into the MIRA (Miskolc Ranking) model, while data analysis transitioned an Exploratory Data Analysis. Subsequently, data utilization progressed into Machine Learning.

The catalyst database currently contains over 15,000 data points, inclusive 450 distinct catalyst combinations and experimental hydrogenation tests on aromatic nitrogen compounds. This extensive catalyst library comprises scientific sources and their quality, publication dates, the composition of the catalyst, and the parameters that characterize the catalyst.

These parameters, along with the database itself, are the basis of the MIRA model. Miskolc Ranking methodology embodies a multi-step approach for identifying novel patterns, potentially useful and interpreted in data collected for the selected catalytic reaction. This functional and practical mathematical framework aids in precise catalyst characterization and comparison. Employing a 15-parameter system, catalysts were characterized via model, the data was standardized and weighed, and then created a single quantifiable data using mathematical formulas. The catalysts were ranked and classified according to the MIRA number obtained so far to facilitate comparison. This model extends to catalysts used in hydrogenation reactions of nitrobenzene and dinitrotoluene.

A comprehensive review of the model was carried out through Exploratory Data Analysis. The data set was examined, carried out data cleaning and data filtering. Based on correlation analysis, modifications were made to the model, and an expanded database was established for validation. Using the MIRA model, best catalyst combinations were determined. Exploratory data analysis also produced additional chemical insights, revealing patterns within parameters pairs and catalyst compositions. Completed data sets have been transferred to machine learning applications and form training and testing sets for various parameter predictions.

In essence, extracting valuable chemical information from data leads to profound acquisition of knowledge. Personally, this journey has provided knowledge of multivariate data analysis and promoted understanding of artificial intelligence applications, thus enriching my knowledge of catalyst hydrogenation of aromatic nitro compounds.

Furthermore, my doctoral research has led to fruitful results. The establishment of MIRA highlighted the shortcomings of contemporary publishing practices. As a result, members of the Institute of Chemistry have aligned themselves with the standard MIRA descriptor systems in their publications.

The MIRA model also extends to a successful method of literature research, validated through education implementation. University students use the system t identify process weaknesses, which leads to valuable insight into the application of the model. Work is currently underway to use this model for the data base of methanol production from carbon dioxide.

Cooperation between the Institute of Chemistry and computer scientists aims to exploit artificial intelligence for chemical applications. I look forward to the continuation of this collaborative effort.

Overall, my work summarizes the latest research findings and introduces novel catalyst design approach, based on data-driven research. It also provides tools for the development of industrial catalysts, bridges the gap between the results of scientific research and the practical application through the MIRA model. The consideration of sustainability parameters clearly shifts catalyst qualification towards application. This work serves as an example of data analysis and the integration of artificial intelligence to optimize industrial processes.

6. ÖSSZEFOGLALÓ

UTAZÁS A „HAGYOMÁNYOS" IRODALOMKUTATÁSTÓL A GÉPI TANULÁSIG

Az itt álló kifejezés a legjobb összefoglalása a kutató munkám során megtett utamnak. A doktori kutatásom az aromás nitrovegyületek katalitikus hidrogénezési folyamatának fejlesztésével kezdődött, és a hozzá kapcsolódó hidrogénező katalizátorok feltérképezésével folytatódott. A „hagyományos" irodalomkutatás során számos kérdés merült fel, amely az adatgyűjtés elkezdéséhez vezetett.

Az adatok halmaza egy átfogó katalizátor-adatbázissá alakult át. Az adatok összehasonlítása a MIRA (Miskolc Ranking) modell létrejöttéhez, míg az adatelemzés az úgynevezett feltáró adatelemzéshez vezetett (Exploratory Data Analysis). Később ez a folyamat a gépi tanulásban látszott kiteljesedni.

A katalizátor-adatbázis jelenleg több, mint 15.000 adatot tartalmaz, amely 450 különböző katalizátor kombinációt és kísérleti hidrogénezési tesztet ír le aromás nitrovegyületekre. Kiterjed az adatok tudományos forrásaira, azok minőségére, a publikálás körülményeire, a katalizátorok összetételére, előállítására, katalizátort jellemző paraméterekre.

Az adatbázis paraméterei, valamint a benne foglalt adathalmaz alkotják a MIRA modell alapját. A MIRA egy többlépcsős módszer, amely olyan új információ azonosítására szolgál az adott katalitikus reakcióra vonatkozó adatok révén, amely potenciálisan felhasználhatók a katalizátorfejlesztésben és tervezésben. Ez a funkcionális és gyakorlatias matematikai keret segít az egzakt katalizátorjellemzésben és azok összehasonlításában. A modell egy 15 paraméteres deszkriptor rendszerből épül fel, amelynek segítségével megvalósítható a katalizátorok jellemzése. Az adatok normalizálása és a paraméter szerinti súlyozás után egy matematikai formula szerint kiszámítható a MIRA szám. A katalizátorok ez alapján rangsorolhatók és osztályozhatók, ez által könnyebbé téve az összehasonlítást. A modell jelen alkalmazásban a nitrobenzol és a dinitrotoluol katalitikus hidrogénezésére használt katalizátorokra terjed ki.

Az adatok átfogó felülvizsgálata feltáró adatelemzéssel történt. Ennek során az adathalmaz átvizsgálásra került, adattisztításon és adatszűrésen esett át. Korrelációs analízis segítségével megvizsgálásra kerültek a paraméterek közötti kapcsolatok és erősségeik. Az addigi tapasztalatok és az elemzés eredményeképpen módosított adatgyűjtéssel egy kibővített adatbázis létrehozására került sor, szintén a nitrobenzol hidrogénezésére vonatkozóan. A kibővített adatbázis katalizátorainak adatelemzését követően megtörtént a MIRA21 modell felülvizsgálata és kismértékű módosítással létrejött a MIRA23 modell. A MIRA23 szerinti minősítést követően meghatározásra kerültek a legjobb katalizátor kombinációk. A páronkénti adatelemzés során mintázatok fedezhetők fel, amelyek új információként szolgálnak a különböző katalizátor kombinációkról. Az adattisztításon és szűrésen átesett adathalmaz felhasználásának kiterjesztése a továbbiakban a gépi tanulásban való alkalmazásra terjedt ki. Ennek során az adathalmazt, mint tanuló és teszt adathalmazt használja az algoritmus, hogy prediktáljon általunk kiválasztott tulajdonságokat a bemeneti paraméterek alapján. A kutatási munka jelenleg is folyamatban van.

A doktori értekezés az adatokból történő értékes kémiai információk kinyerésére irányult, melynek révén új ismeret szerezhető. Számomra ez a négy év a többváltozós adatelemzés és a mesterséges intelligencia tudományának, inkább egy szeletének, megismerését eredményezte, valamint alkalmazási lehetőségeinek végtelen tárházát mutatta be. Ezzel egyidejűleg bizonyítva azt is, hogy már a gépi tanulás kiindulási adathalmazának megválasztásánál is rengeteg kérdés merül fel, amely további komplex előkészítési folyamatokat igényel. Emellett tudásom szélesedett azáltal, hogy az aromás nitrovegyületek hidrogénezésére alkalmas katalizátorok fejlesztési irányvonalait és kutatási eredményeit is megismertem.

Ezenfelül a doktori munkám további pozitív hozadékkal járt. A MIRA létrehozása rávilágított arra, hogy a kortárs publikációs gyakorlat meglehetősen rendszertelen és hiányos. A MIRA alkalmazásával a Kémiai Intézet publikációi sokkal átláthatóbb és precízebb módon kerülnek megírásra. A modellt továbbá volt lehetőség egyetemi hallgatók segítségével is tesztelni, amely nemcsak a MIRA továbbfejlesztését támogatta, hanem a hallgatók számára is betekintést nyújtott a modell alkalmazási lehetőségeibe. Jelenleg is folyik kutatási munka, melynek során a metanol szén-dioxidból történő gyártására adaptáljuk a rendszert. A doktori értekezés tovább erősítette a Kémiai Intézet és a mesterséges intelligencia tudományában jártas egyetemi szakemberek együttműködését kémiai kutatási témákban, amelyben szeretnék a továbbiakban is részt venni.

Összefoglalva tehát elmondható, hogy a doktori munka összefoglalja az aromás nitrovegyületek hidrogénező katalizátorainak legfrissebb és legígéretesebb kutatási eredményeit és új, adatalapú kutatásra épülő katalizátor tervezési stratégiát mutat be. Emellett egy olyan eszköz, amely az ipari katalizátorok fejlesztésében is előnyt jelenthet, hiszen áthidalja azokat a réseket, amelyek a tudományos kutatás és a gyakorlati alkalmazás között jelentkeznek. A fenntarthatósági paraméterek figyelembevételével például egyértelműen az ipari megvalósíthatóságra kerül a hangsúly. Ezenfelül az adatelemzés és a mesterséges intelligencia integrációja egy ipari katalizátorfejlesztésbe jó példaként szolgál a vegyipari innovációs lehetőségekre.

7. NEW SCIENTIFIC RESULTS – THESES

1. A MATHEMATICAL FRAMEWORK WAS DEVELOPED FOR CHARACTERIZING CATALYSTS BY A SINGLE QUANTITATIVE DATA.

***Figure T 1** Visualization of MIRA21 number*

As a result of the research MIRA (MIskolc RAnking) model was established, which can be used to characterize catalysts described in scientific research using a single quantitative data. 13 parameters that can be used to characterize catalyst were determined. 13-parameter descriptor system was created, each characterized by quantifiable data. The parameters are divided into four groups, namely the variables that characterize the performance of the catalyst, the parameters that describe the reaction conditions, the properties of catalyst and the sustainability parameters. A mathematical procedure was developed that allows easy identification and comparison of catalysts. In mathematical procedures, parameters are normalized, then weighted by group, and the number of MIRAs is generated using mathematical formulas. Catalysts ranked and classified become comparative by the number of MIRAs.

***Figure T 2** Descriptor system of the model*

2. THE DESCRIPTOR SYSTEM AND THE UNIQUE WEIGHT FACTORS HAVE BEEN REVIEWED ON THE BASIS OF WHICH A GENERAL WEIGHT FACTOR HAS BEEN PROPOSED, MAKING BOTH THE RANKING AND CLASSIFICATION CLEAR.

During PhD work, the descriptor system was revised by Exploratory Data Analysis. Based on the evaluation, version MIRA23 was created by slightly modifying the MIRA21 model. The weighting of all parameters was validated.

The system is suitable for the characterization of catalysts for hydrogenation of nitrobenzene, and it can be applied on any catalytic system based on the test[180,199].

3. MIRA21(23) DATA- AND CATALYST RATING SYSTEM, TOGETHER WITH MACHINE LEARNING, CAN BE USED TO CREATE NEW CATALYST DESIGN STRATEGIES.

The steps taken in doctoral thesis provide a new catalyst design strategy. The first step is the application of the MIRA model, then the Exploratory Data Analysis and the application Machine Learning methods.

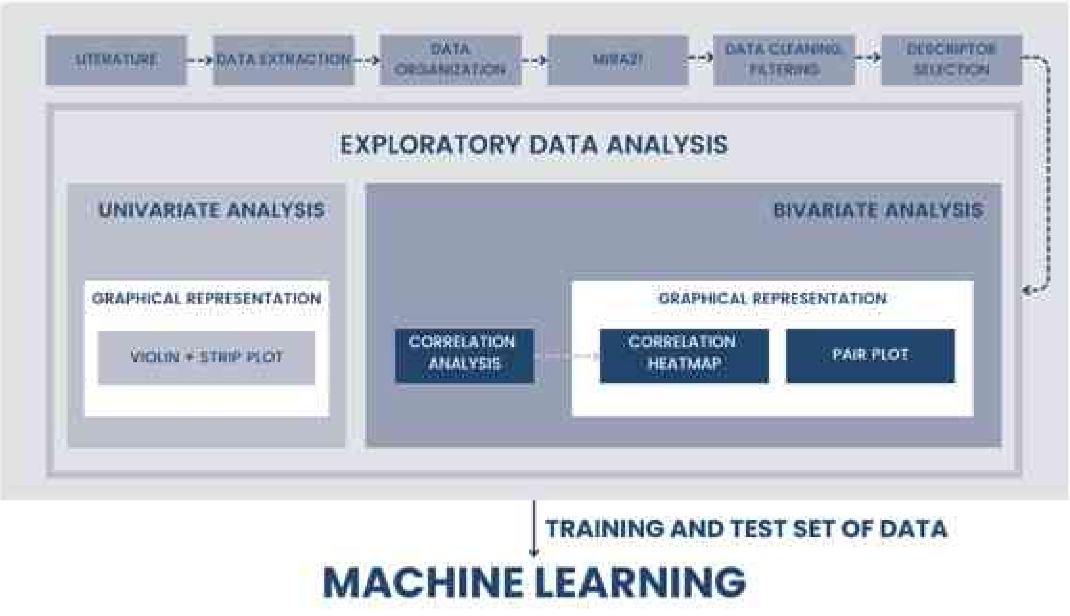


Figure T 3 *Catalyst design strategy*

4. FOR SPECIFIC ACTIVE METALS, IT IS POSSIBLE TO DETERMINE THE CONCENTRATION RANGE TO ACHIEVE A CONVERSION LEVEL HIGHER THAN A CERTAIN LEVEL.

Based on the examination of the parameter system of the developed multivariate method, chemical information was produced with the help of Exploratory Data Analysis. In the analysis of joint plots, groups of different compositions and performance characteristics can be distinguished depending on the composition of the catalyst. Based on Figure T4, in case of iron active component, a conversion of more than 70% can be achieved at a concentration of 20-22%.

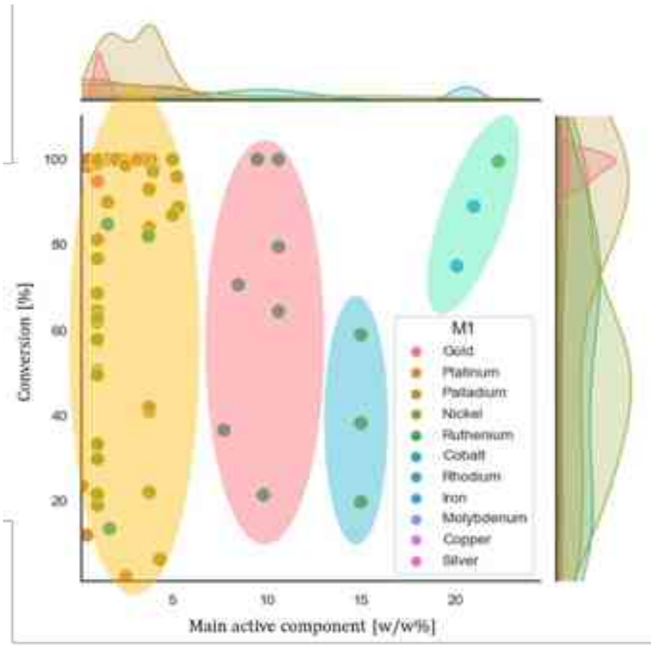


Figure T 4 *Joint plot of Active component concentration (M12 w/w%) and Conversion classified by catalyst active component(M1)*

5. IN CATALYST RESEARCH, SUSTAINABILITY PARAMETERS WERE FIRST DEFINED AS DESCRIPTOR INDICATORS OF CATALYSTS USING MATHEMATICAL PROCEDURES.

Table T 1 *Sustainability parameters*

Sustainability parameters	*IV.*	*11.*	*STAB*	*Stability*	-	*From 0 to n, where 0 is no information about stability, 1 means that the stability was investigated, 2,3… the number of cycles until the conversion remains above 90%*
		12.	*Greenness*	*Toxicity*	-	*Rank of the active component according to toxicity*[316]
				Endangered elements	-	*Rank of the active component according to endangered element scenario*[316]
				Life cycle assessment	-	*Rank of the elements according to life cycle assessment*[316]
		13.	*PR*	*Price*	*euro/g*	*Price is of 1 gram catalyst according to the active component concentration in euro*

8. LIST OF PUBLICATIONS

PUBLICATIONS RELATED TO THE SUBJECT OF THE DISSERTATION

1. Alexandra Jakab-Nácsa, Emőke Sikora, Ádám Prekob, László Vanyorek, Milán Szőri, Renáta Boros Zsanett, Károly Nehéz, Martin Szabó, László Farkas, Béla Viskolcz, Comparison of Catalysts with MIRA21 Model in Heterogeneous Catalytic Hydrogenation of Aromatic Nitro Compounds, *MDPI Catalysts,* 2022, 12 (5), 467, https://doi.org/10.3390/catal12050467**, IF 3.9**

2. Alexandra Jakab-Nácsa, Viktória Hajdú, László Vanyorek, László Farkas, Béla Viskolcz, Overview of Catalysts with MIRA21 Model in Heterogeneous Catalytic Hydrogenation of 2,4-Dinitrotoluene, *MDPI Catalysts*, 2023, 13(2), 387, https://doi.org/10.3390/catal13020387, **IF 3.9**

3. Alexandra Jakab-Nácsa, Attila Garami, Béla Fiser, László Farkas, Béla Viskolcz, Towards Machine Learning in Heterogeneous Catalysis—A Case Study of 2,4-Dinitrotoluene Hydrogenation, *MDPI International Journal of Molecular Sciences*, 2023, 24 (14), 11461, https://doi.org/10.3390/ijms2 41411461, **IF 5.6**

FURTHER PUBLICATIONS

1. Viktória Hajdu, Alexandra Jakab-Nácsa, Gábor Muránszky, István Kocserha, Béla Fiser, Tibor Ferenci, Miklós Nagy, Béla Viskolcz, László Vanyorek, Precious-Metal-Decorated Chromium(IV) Oxide Nanowires as Efficient Catalysts for 2,4-toluenediamine Synthesis, *International Journal of Molecular Science*, 2021, 22(11), 5945, https://doi.org/10.3390/ijms22115945 , **IF 6.2**
2. Alexandra Jakab-Nácsa, Dávid Stomp, László Farkas, George Kaptay, Large NaCl-Effect on the Decomposition Rate of Chlorate Ions in HCl-Containing Brine Solutions and Its Consequences for Chlor-Alkali Industry, Periodica Polytechnica – Chemical Engineering, 2021, 65 (2),238-242, https://doi.org/10.3311/PPch.14634 , **IF 2022 1.3**

PRESENTATION RELATED TO THE DISSERTATION

1. Borsodi Vegyipari Napok
Rangsorolhatók-e a kémiai reakciók katalizátorai?
17 November 2021, Miskolc, Hungary

2. XXVIII. Nemzetközi Vegyészkonferencia
Katalizátorok összehasonlíthatósága a MIRA21 modell alapján
27-29 October 2022 , Nagyvárad, Romania

3. International Conference on Chemical Engineering
Classification of Catalysts with MIRA21 Model in Heterogeneous Catalytic Hydrogenation of Aromatic Nitro Compounds
March 20-21, 2023, Rome, Italy

POSTERS RELATED TO THE DISSERTATION

1. 26th International Congress of Chemical and Process Engineering
   Utilization of laboratory testing and a mathematical model for the purpose of selecting a heterogeneous hydrogenation catalyst
   21-25 August, 2022, Prague, Czech Republic
2. 4. MKE Nemzeti Vegyészkonferencia
   Aromás nitrovegyületek katalitikus hidrogénezésére alkalmas katalizátorok fejlesztése az iparban
   10-12 July, 2023, Eger, Hungary

FURTHER POSTERS

1. 12th European Congress of Chemical Engineering
   Wastewater Treatment Optimization of Nitration of Aromatics
   15-19 September, 2019, Florence, Italy
2. 26th International Congress of Chemical and Process Engineering
   Cooperation of cities and local companies for climate change adaptation
   21-25 August, 2022, Prague, Czech Republic
3. 4. MKE Nemzeti Vegyészkonferencia
   Ipari szürkevíz visszaforgatási lehetőségek vizsgálata a LIFE projekt keretén belül
   10-10 July, 2023, Eger, Hungary

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10. APPENDIX

1. EXCEL DATABASE SYSTEM[106]

CAT ID	CONVERSION	YIELD	SELECTIVITY	TON	TEMPERATURE
HNB_FUY2018_1	100 %	100%	100%	7056	313 K
–	–	–	–	–	–
–	–			–	–

2. FIRST TEST SYSTEM RESULTS – NITROBENZENE HYDROGENATION CATALYSTS

RANK	CAT. ID	CAT. Name in journal	KNOWN parameters	MIRA21 number	Class.
13	HNB_HYD2016_3	Ni/C-Al2O3	15	11.53	Q1
14	HNB_BEI2012_3	Pt/TiO2/RGO	14	11.51	Q1
15	HNB_CHE2009_1*	1 wt% Pd/HT	14	11.51	Q1
16	HNB_FUY2018_2	PtCo nanoparticle	14	11.49	Q1
17	HNB_FUY2018_1	PtCo nanoflower	14	11.47	Q1
18	HNB_BEI2005_2	Pt CNT	15	11.42	Q1
19	HNB_GUA2017_2	Pd/CNT	15	11.41	Q1
20	HNB_BEI2012_2	Pt/RGO	14	11.33	Q1
21	HNB_BEI2005_1	Pt CNT	15	11.33	Q1
22	HNB_CHE2009_2*	1 wt% Pd/MgO	14	11.32	Q1
23	HNB_CHE2009_3*	1 wt% Pd/ϒ-Al2O3	14	11.32	Q1
24	HNB_BEI2013_3	Pd/MWCNT-SA-3.6	15	11.31	Q1
25	HNB_GUA2017_1	Pd/NCNT	15	11.30	Q1
26	HNB_BEI2007_3	Pt/CNTs LRT	15	11.30	Q1
27	HNB_BEI2012_1	Pt/TiO2	14	11.29	Q1
28	HNB_BEI2013_1	Pd/MWCNT-SA-6.0	15	11.28	Q1
29	HNB_GUA2020_2	Pt/CeO2-R	15	11.26	Q1
30	HNB_BEI2014_1	Pd/Fe_2O_3	15	11.11	Q1
31	HNB_BEI2008_1	Pd/FSA	14	11.09	Q1
32	HNB_BEI2013_5	Pd/MWCNT-IM	15	11.08	Q1
33	HNB_BEI2010_1	5wt% Pt/MWNT	14	11.04	Q1
34	HNB_WUH2016_1	C-Fe3O4-Pd	14	10.98	Q1
35	HNB_GUA2020_1	Pt/CeO2-C	15	10.96	Q1
36	HNB_BLO2015_7	Ru-14	14	10.95	Q1
37	HNB_GUA2020_1	Pt CeO2-R-300	15	10.91	Q1
38	HNB_TAI2017_2	Co@NMC-700	15	10.90	Q1
39	HNB_TIA2019_1	Co-NSPC-N	15	10.90	Q1
40	HNB_INC2018_1	Pd/NH2-UiO-66	15	10.87	Q1
41	HNB_BEI2010_2	27,4wt% Pt/MWNT	14	10.85	Q1
42	HNB_BEI2010_3	50wt% Pt/MWNT	14	10.84	Q1
43	HNB_POR2016_4	50 wt% NiO/Al2O3+SiO2	15	10.84	Q1
44	HNB_BEI2007_1	Pt/CNTs HRT	15	10.83	Q1
45	HNB_POR2016_2	0,3 wt% Pd/Al2O3/1,85	15	10.83	Q1
46	HNB_POR2016_1	1 wt% Pd/Al2O3	15	10.78	Q1
47	HNB_GUA2020_3	Pt/CeO2-P	15	10.76	Q1
48	HNB_BLO2015_6	Ru-12	14	10.72	Q1
49	HNB_BLO2015_5	Ru-7	14	10.72	Q1
50	HNB_GUA2020_4	Pt CeO2-C-600	15	10.71	Q1
51	HNB_TAI2017_4	Co@NMC-900	15	10.70	Q1
52	HNB_GUA2017_3	Pd/CNT	15	10.67	Q1
53	HNB_BLO2015_3	Ru-5	14	10.63	Q1
54	HNB_MIS2019_2	Pt/N-BCNT	14	10.63	Q1
55	HNB_BEI2010_4	10wt% Pt/C	14	10.62	Q1

RANK	CATALYST ID	CAT. Name in Journal	KNOWN parameters	MIRA21 number	Class.
56	HNB_BLO2015_4	Ru-11	14	10.56	Q2
57	HNB_XIA2019_1	Ni-Zn/AC-350	15	10.51	Q2
58	HNB_MIS2019_3	Rh/N-BCNT	14	10.49	Q2
59	HNB_HAN2010_1	Ni-5/SiO2-EN	15	10.41	Q2
60	HNB_POR2016_4	50 wt% NiO/Al2O3+SiO2	15	10.36	Q2
61	HNB_BEI2013_6	Pd/AC	14	10.32	Q2
62	HNB_BEI2007_2	Pt/AC HRT	15	10.31	Q2
63	HNB_BLO2015_2	Ru-16	14	10.31	Q2
64	HNB_CAR2018_1	AuPd/TiO2 (MIM)	14	10.21	Q2
65	HNB_POR2008_1	NiFC1	14	10.18	Q2
66	HNB_POR2008_2	NiFC2	14	10.17	Q2
67	HNB_HYD2016_1	Ni/C	15	10.16	Q2
68	HNB_TIA2019_2	Co-NSPC-C	15	10.10	Q2
69	HNB_POR2008_3	NiFC3	14	10.07	Q2
70	HNB_GUA2020_6	Pt CeO_2-P-600	15	10.05	Q2
71	HNB_BEI2013_4	Pd NPs-4.3	14	10.04	Q2
72	HNB_WUH2019_1	Co@CN-800	13	9.99	Q2
73	HNB_TOU2020_1	PdB	14	9.92	Q2
74	HNB_CAR2018_7	AuPd/TiO_2 (SIM)	14	9.89	Q2
75	HNB_BEI2010_5	5wt% Pt/C	14	9.88	Q2
76	HNB_BLO2015_1	Ru-18	14	9.77	Q2
77	HNB_TAI2017_9	Co@NC@SiO_2-800	14	9.75	Q2
78	HNB_LAN2020_2	γ-Fe_2O_3/NPC-700	14	9.70	Q2
79	HNB_TAI2017_12	Co@NMC-800 (1:2)	13	9.60	Q2
80	HNB_LAN2020_1	γ-Fe_2O_3/NPC-600	14	9.58	Q2
81	HNB_CAR2018_3	Pd/TiO_2 (MIM)	14	9.56	Q2
82	HNB_TIA2019_3	Co-NSPC-S	15	9.55	Q2
83	HNB_CHA2016_3	Ni1.99P-s-1 h	13	9.54	Q2
84	HNB_BEI2017_1	Co_3S_4	13	9.51	Q2
85	HNB_TOK2004_1	Pt/C 200 °C-2h	14	9.47	Q2
86	HNB_TAI2017_5	Co/NMC-800	13	9.47	Q2
87	HNB_TAI2017_7	Co@NMC-800-H2SO4	13	9.43	Q2
88	HNB_TAI2017_8	Co@NC-800	14	9.42	Q2
89	HNB_CAR2018_6	AuPd/TiO_2 (CIM)	14	9.39	Q2
90	HNB_HAR2019_1	FeOx@CN-hpes-400	13	9.39	Q2
91	HNB_LAN2020_4	γ-Fe_2O_3/NPC-900	14	9.35	Q2
92	HNB_TOK2004_3	Pt/C 500 °C-2h	14	9.35	Q2
93	HNB_TIA2019_4	Co-NSPC-Cl	15	9.32	Q2
94	HNB_TOK2004_2	Pt/C 300 °C-2h	14	9.31	Q2
95	HNB_CAR2018_4	AuPd/MgO (MIM)	14	9.30	Q2
96	HNB_HYD2008_4*	Ru/SBA-15	12	9.25	Q2
97	HNB_LAN2020_5	γ-Fe_2O_3/NPC-1000	14	9.22	Q2
98	HNB_HYD2008_5*	Ru/SBA-15	12	9.15	Q2
99	HNB_POR2008_4	RNi	13	9.12	Q2
100	HNB_HAN2010_4	Ni-15/SiO_2-EN	13	9.11	Q2
101	HNB_TOK2004_4	Pt/C 600 °C-2h	14	9.08	Q2
102	HNB_HYD2008_3*	Ru/SBA-15	12	9.03	Q2
103	HNB_CHA2016_2	Ni1.91P-s-0.5 h	13	8.97	Q2

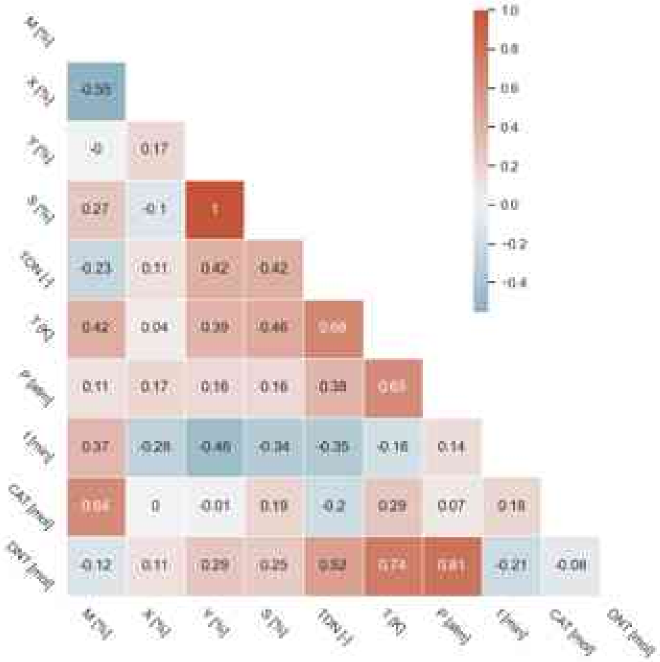
RANK	CATALYST ID	CAT. Name in Journal	KNOWN parameters	MIRA21 number	Class.
104	HNB_CHA2016_4	Ni2.05P-s-3 h	13	8.88	Q3
105	HNB_GUA2020_3	Pt CeO_2-C-300	13	8.88	Q3
106	HNB_NAN2014_1	Pt/AlO(OH)	11	8.81	Q3
107	HNB_HAN2010_2	Ni-5/SiO_2-NI	15	8.80	Q3
108	HNB_BEI2007_4	Pt/AC LRT	15	8.76	Q3
109	HNB_HAN2010_3	Ni-5/SiO_2-AC	15	8.76	Q3
110	HNB_HAN2010_3	Ni-5/SiO_2-AC	15	8.75	Q3
111	HNB_CHA2016_1	Ni1.96P-s-10 min	13	8.69	Q3
112	HNB_TAI2017_1	Co@NMC-600	14	8.63	Q3
113	HNB_HYD2008_2*	Ru/SBA-15	12	8.60	Q3
114	HNB_BEI2005_3	Pt AC	14	8.50	Q3
115	HNB_BEI2005_1	Cu/SiO_2	11	8.49	Q3
116	HNB_TAI2017_10	Ni@NMC-800	13	8.43	Q3
117	HNB_BEI2013_7	Pd/Al_2O_3	13	8.42	Q3
118	HNB_CAR2018_5	AuPd/C (MIM)	14	8.38	Q3
119	HNB_TOK2004_5	Pt/C 750 °C-3h	14	8.38	Q3
120	HNB_TAI2017_6	CoOx@NMC-800	13	8.26	Q3
121	HNB_XIA2021_2	0.075%Pt/SBA-15	14	8.17	Q3
122	HNB_GUA2020_5	Pt CeO_2-P-300	13	8.16	Q3
123	HNB_HAN2010_5	Raney Ni	13	8.09	Q3
124	HNB_CAR2018_2	Au/TiO_2 (MIM)	14	8.04	Q3
125	HNB_HYD2008_1*	Ru/SBA-15	12	7.99	Q3
126	HNB_TAI2017_11	Fe@NMC-800	13	7.92	Q3
127	HNB_NAN2014_3	Pt/MWCNTs	10	7.92	Q3
128	HNB_XIA2021_3	0.07%Pt/ZrO_2	14	7.90	Q3
129	HNB_XIA2021_4	0.09%Pt/γ-Al_2O_3	14	7.89	Q3
130	HNB_NAN2014_2	Pt/Al_2O_3	10	7.86	Q3
131	HNB_BEI2013_8	Pd/SiO_2	13	7.84	Q3
132	HNB_NAN2014_4	Pt/AC	10	7.80	Q3
133	HNB_GLA2002_3	Pd/CSXU	10	7.77	Q3
134	HNB_GLA2002_2	Pd/CA1	10	7.57	Q3
135	HNB_SHA2015_2	Pt/C	12	7.45	Q3
136	HNB_BEI2013_9	Pd/MgO	13	7.41	Q3
137	HNB_GLA2002_1	Pd/CN1	10	7.26	Q4
138	HNB_FUY2018_3	Pt/C	11	6.97	Q4
139	HNB_SHA2006_1	Meso Ni–B	9	6.95	Q4
140	HNB_SHA2000_1	Pd-B/SiO_2(fresh)	9	6.91	Q4
141	HNB_SHA2000_2	Pd-B/SiO_2 (473 K)	9	6.91	Q4
142	HNB_DAL2015_1*	Pd/AM	11	6.90	Q4
143	HNB_NAN2014_5	Pt/TiO_2	10	6.90	Q4
144	HNB_SHA2000_3	Pd-B/SiO_2 (673 K)	9	6.88	Q4
145	HNB_QIN2016_2	Ni-Fe-1/SiO_2	10	6.77	Q4
146	HNB_QIN2016_3	Ni-Fe-2/SiO_2	10	6.76	Q4
147	HNB_SHA2000_6	Pd/SiO_2 (fresh)	9	6.66	Q4
148	HNB_SHA2000_4	Pd-B/SiO_2 (873 K)	9	6.54	Q4
149	HNB_SHA2000_5	Pd-B/SiO_2 (973 K)	9	6.48	Q4
150	HNB_SHA2006_2	Regular Ni–B	9	6.41	Q4
151	HNB_DAL2015_2*	Pd/CNF/monolith	11	6.39	Q4
152	HNB_NAN2014_6	Pt/MCM-41	10	6.34	Q4
153	HNB_SHA2000_7	Pd-B	8	5.78	Q4
154	HNB_QIN2016_1	Fe/SiO_2	9	5.71	Q4

3. SECOND TEST – DNT CATALYST RESULTS

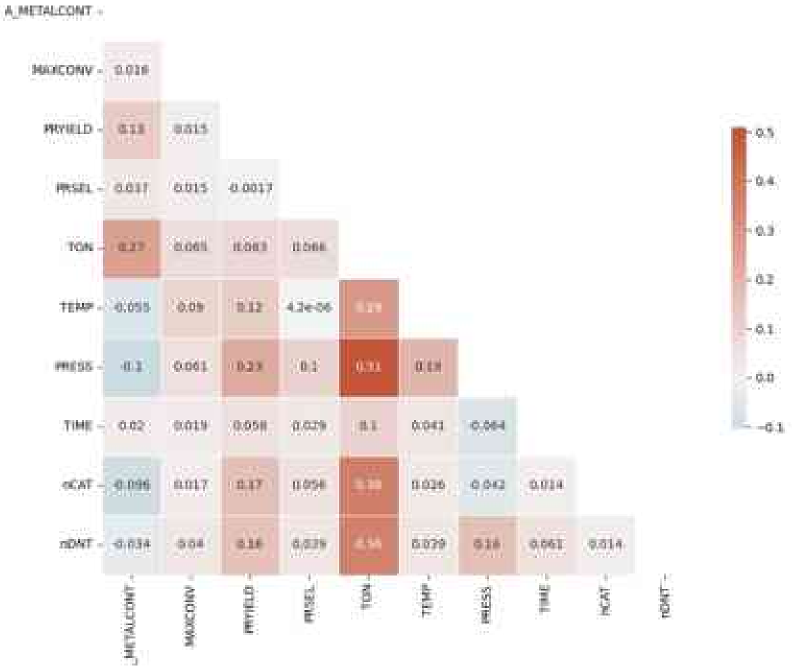
No.	CATALYST ID	Catalyst name	Catalyst support	Main active component	Known Par.	MIRA21 number	Class
1	HDNT/MIS/2021/2/2	Pt/CrO$_2$	Chromium(IV)-dioxide	Platinum	15	11.50	D1
2	HDNT/MIS/2021/2/1	Pd/CrO$_2$	Chromium(IV)-dioxide	Palladium	15	11.49	D1
3	HDNT/MIS/2021/3/1	Pd/NiFe$_2$O$_4$	Nickel ferrite	Palladium	15	11.45	D1
4	HDNT/TIA/2020/1/3	15Pt/ZrO$_2$-300	Zirconium-dioxide	Platinum	13	11.44	D1
5	HDNT/TIA/2020/1/4	15Pt/ZrO$_2$-400	Zirconium-dioxide	Platinum	13	11.43	D1
6	HDNT/TIA/2020/1/2	15Pt/ZrO$_2$-200	Zirconium-dioxide	Platinum	13	11.42	D1
7	HDNT/MIS/2021/1/2	Pd/maghemite	Maghemite	Palladium	15	11.35	D1
8	HDNT/TIA/2020/1/5	45Pt/ZrO$_2$-300	Zirconium-dioxide	Platinum	13	11.06	D1
9	HDNT/TIA/2020/1/6	60Pt/ZrO$_2$-300	Zirconium-dioxide	Platinum	13	11.01	Q1
10	HDNT/TIA/2020/1/7	85Pt/ZrO$_2$-300	Zirconium-dioxide	Platinum	13	11.00	Q1
11	HDNT/MIS/2021/3/2	Pd/CoFe$_2$O$_4$	Cobalt ferrite	Palladium	15	10.84	Q1
12	HDNT/SHA/2012/1/1	Ni/HY catalyst	HY molecular sieve	Nickel	15	10.77	Q1
13	HDNT/MIS/2021/1/1	Pt/maghemite	Maghemite	Platinum	15	10.67	Q1
14	HDNT/MIS/2021/3/3	Pd/CuFe$_2$O$_4$	Copper ferrite	Palladium	15	10.48	Q1
15	HDNT/MIS/2022/1/3	Pd/NiFe$_2$O$_4$-NH$_2$	Nickel-ferrite	Palladium	13	10.31	Q1
16	HDNT/MIS/2022/1/1	Pd/CoFe$_2$O$_4$-NH$_2$	Cobalt-ferrite	Palladium	13	10.28	Q2
17	HDNT/MIS/2021/1/3	Pd-Pt/maghemite	Maghemite	Palladium	14	10.14	Q2
18	HDNT/PUN/1999/1/5	20% Ni/HY	HY zeolite	Nickel	14	9.50	Q2
19	HDNT/DAL/1997/1/2	PVP-Pd-1/4 Pt	PVP	Palladium	13	9.47	Q2
20	HDNT/PUN/1999/1/6	10% Ni/HY	HY zeolite	Nickel	14	9.44	Q2
21	HDNT/MES/2001/1/1	MGPd05	Chemviron SC XII active carbon	Palladium	13	9.24	Q2
22	HDNT/MES/2001/1/3	MGPd1b	Chemviron SC XII active carbon	Palladium	13	9.23	Q2
23	HDNT/MES/2001/1/8	MGPd5a	Chemviron SC XII active carbon	Palladium	13	9.20	Q2
24	HDNT/HAN/2001/1/2	B	Chemically activated carbon	Iridium	13	9.19	Q2
25	HDNT/MES/2001/1/4	MGPd1c	Chemviron SC XII active carbon	Palladium	13	9.19	Q2
26	HDNT/HAN/2001/1/1	A	Chemically activated carbon	Iridium	13	9.19	Q2
27	HDNT/MES/2001/1/7	MGPd5	Chemviron SC XII active carbon	Palladium	13	9.17	Q2
28	HDNT/MES/2001/1/5	MGPd1d	Chemviron SC XII active carbon	Palladium	13	9.14	Q2
29	HDNT/MES/2001/1/2	MGPd1a	Chemviron SC XII active carbon	Palladium	13	9.13	Q2
30	HDNT/MES/2001/1/6	MGPd3	Chemviron SC XII active carbon	Palladium	13	9.06	Q3

31	HDNT/HAN/2001/1/3	C	Steam activated carbon	Palladium	13	9.03	Q3
32	HDNT/MIS/2022/1/2	$Pd/CdFe_2O_4$-NH_2	Cadmium-ferrite	Palladium	13	8.95	Q3
33	HDNT/ZUR/1987/1/1	0.5 % Pt/Al_2O_3	Al2O3	Platinum	13	8.88	Q3
34	HDNT/HAN/2001/1/4	D	Steam activated carbon	Palladium	13	8.79	Q3
35	HDNT/HAN/2001/1/5	E	Oleophilic carbon black	Palladium	13	8.78	Q3
36	HDNT/PUN/1999/1/2	20% Ni/SiO_2	SiO2	Nickel	14	8.63	Q3
37	HDNT/SHA/2012/1/4	Ni-La6-B		Nickel	12	8.36	Q3
38	HDNT/SHA/2012/1/3	Ni-La4-B		Nickel	12	8.34	Q3
39	HDNT/SHA/2012/1/2	Ni-La2-B		Nickel	12	8.33	Q3
40	HDNT/SHA/2012/1/1	Ni-La0-B		Nickel	12	8.32	Q3
41	HDNT/SAP/2004/1/3	Pt/C in ethanol	Active carbon	Platinum	13	8.02	Q3
42	HDNT/SHA/2012/1/5	Ni-La8-B		Nickel	12	7.92	Q3
43	HDNT/SAP/2004/1/2	Pt/C in ethanol	Active carbon	Platinum	13	7.90	Q3
44	HDNT/TIA/2020/1/1	15Pt/ZrO_2-100	Zirconium-dioxide	Platinum	13	7.85	Q4
45	HDNT/PUN/1999/1/4	20% Ni/HZSM-5	HZSM-5	Nickel	14	7.75	Q4
46	HDNT/TAE/1993/1/1	SA	Activated carbon	Palladium	11	7.58	Q4
47	HDNT/TAE/1993/1/3	DA	Activated carbon	Palladium	11	7.52	Q4
48	HDNT/TAE/1993/1/2	SAON	Activated carbon	Palladium	11	7.50	Q4
49	HDNT/TAE/1993/1/4	DAON	Activated carbon	Palladium	11	7.49	Q4
50	HDNT/PUN/1999/1/1	20% Ni/Al_2O_3	Al2O3	Nickel	14	7.43	Q4
51	HDNT/BAR/2000/1/1	Pd(AAEMA)2/EMA/EGDMA	Polymer-supported complex	Palladium	13	7.27	Q4
52	HDNT/DAL/1997/1/1	PVP-$PdCl_2$	PVP	Palladium	10	6.99	Q4
53	HDNT/PUN/1999/1/3	20% Ni/TiO_2	TiO2	Nickel	14	6.95	Q4
54	HDNT/TAE/1993/1/7	VB	Carbon Black	Palladium	10	6.80	Q4
55	HDNT/TAE/1993/1/8	VON	Carbon Black	Palladium	10	6.80	Q4
56	HDNT/TAE/1993/1/6	DAOH	Activated carbon	Palladium	10	6.80	Q4
57	HDNT/TAE/1993/1/5	DAOS	Activated carbon	Palladium	10	6.80	Q4
58	HDNT/SAP/2004/1/1	Pt/C in $scCO_2$	Active carbon	Platinum	13	6.68	Q4

4. RESULTS OF SPEARMAN CORRELATION ANALYSIS WITH FULL DATA SET



5. RESULTS OF SPEARMAN CORRELATION ANALYSIS – DIFFERENCES OF THE UNFILTERED AND FILTERED DATA SET RESULTS



6. FULL RANKING OF THE NITROBENZENE HYDROGENATION CATALYSTS

Rank	Cat ID	Cat appellation	Known p.	MIRA21	Class
1	HNB/CAM/2021/109/2	Pt(OUT)/HNT	12	10.5	D1
2	HNB/CAM/2021/109/1	Pt(IN)/HNT	12	10.3	D1
3	HNB/ZHA/2020/92/4	Pt/CeO2-R-600	12	10.0	D1
4	HNB/LI/2015/35/1	Pt/CMK-3	11	9.9	D1
5	HNB/NIE/2020/93/5	Pt/m-Al2O3-400-20mg	12	9.8	Q1
6	HNB/ZHA/2021/113/1	0.07%Pt/@-ZrO2/SBA-15	12	9.8	Q1
7	HNB/NIE/2020/93/7	Pt/m-Al2O3-400-30mg	12	9.8	Q1
8	HNB/NIE/2020/93/2	Pt/m-Al2O3-400-20mg	12	9.8	Q1
9	HNB/NIE/2020/93/6	Pt/m-Al2O3-400-20mg	12	9.8	Q1
10	HNB/NIE/2020/93/8	Pt/m-Al2O3-400-40mg	12	9.7	Q1
11	HNB/WAN/2013/19/2	Pd/MWCNT-SA-4.3	11	9.6	Q1
12	HNB/PRE/2022/116/1	Pd/CF	12	9.6	Q1
13	HNB/HAJ/2022/118/2	Pd/ZnFe2O4	12	9.5	Q1
14	HNB/HUA/2019/91/1	Ni/Zn/AC-350	12	9.5	Q1
15	HNB/HAJ/2022/115/1	Pd/MnFe2O4 (573K)	12	9.5	Q1
16	HNB/ZHA/2020/92/3	Pt/CeO2-R-600	12	9.5	Q1
17	HNB/PRE/2020/95/1	5% w/w Pd-CC	12	9.5	Q1
18	HNB/LV/2020/105/6	Υ-Fe2O3/NPC-800	11	9.4	Q1
19	HNB/LV/2020/105/3	Υ-Fe2O3/NPC-800	11	9.4	Q1
20	HNB/LV/2020/105/9	Υ-Fe2O3/NPC-800	10	9.4	Q1
21	HNB/LV/2020/105/10	Υ-Fe2O3/NPC-800	11	9.4	Q1
22	HNB/TUR/2015/24/1	Pd/C	11	9.4	Q1
23	HNB/LV/2020/105/11	Υ-Fe2O3/NPC-800	10	9.3	Q1
24	HNB/LV/2020/105/2	Υ-Fe2O3/NPC-700	11	9.3	Q1
25	HNB/LV/2020/105/7	Υ-Fe2O3/NPC-800	11	9.3	Q1
26	HNB/LV/2020/105/1	Υ-Fe2O3/NPC-600	11	9.3	Q1
27	HNB/NIE/2020/93/4	Pt/m-Al2O3-400-20mg	12	9.2	Q1
28	HNB/LV/2020/105/12	Υ-Fe2O3/NPC-800	11	9.2	Q1
29	HNB/NIE/2020/93/3	Pt/m-Al2O3-400-20mg	12	9.2	Q1
30	HNB/SHI/2016/45/4	Pt/H-NCNTs	11	9.2	Q1
31	HNB/ZHA/2020/92/2	Pt/CeO2-R-300	11	9.2	Q1
32	HNB/ZHA/2007/128/3	Pt/CNTs LRT	11	9.2	Q1
33	HNB/NIE/2020/93/1	Pt/m-Al2O3-400-20mg	12	9.1	Q1
34	HNB/NIE/2020/93/9	Pt/m-Al2O3-400-40mg	11	9.1	Q1
35	HNB/ZHA/2020/92/6	Pt/CeO2-C-600	11	9.1	Q1
36	HNB/PRE/2021/114/1	Pd-Pt/CB	11	9.0	Q1
37	HNB/ZHA/2022/117/1	Pd/PIL-Tf2N	11	9.0	Q1
38	HNB/PRE/2021/114/2	Pd-Pt/Fe2O3-CB	11	9.0	Q1
39	HNB/PRE/2021/114/3	Pd-Pt/NiO-CB	11	9.0	Q1
40	HNB/ZHA/2012/12/3	Pt/TiO_2/RGO	11	9.0	Q1
41	HNB/ZHA/2020/92/1	Pt/CeO2-R-300	11	9.0	Q1
42	HNB/HAJ/2022/118/3	Pd/NiZnFe2O4	11	9.0	Q1
43	HNB/WAN/2010/1/1	Ni-5/$SiO_{2\text{-EN}}$	11	9.0	Q1
44	HNB/LV/2020/105/8	Υ-Fe2O3/NPC-800	11	9.0	Q1
45	HNB/SHI/2016/45/3	Pt/L-NCNTs	11	9.0	Q1
46	HNB/HAJ/2022/115/2	Pd/MnFe2O4 (623K)	11	8.9	Q1
47	HNB/HAJ/2022/118/1	Pd/NiFe2O4	11	8.9	Q1
48	HNB/SAN/2009/122/1	1 wt% Pd/HT	11	8.9	Q1
49	HNB/HAJ/2022/115/3	Pd/MnFe2O4 (673K)	11	8.9	Q1
50	HNB/SAN/2009/122/1	1 wt% Pd/Υ-Al2O3	11	8.9	Q1
51	HNB/SAN/2009/122/1	1 wt% Pd/MgO	11	8.9	Q1
52	HNB/HE/2021/111/1	CuAlOx 1,10-phen	11	8.9	Q1
53	HNB/SHI/2016/45/2	Pt/oCNTs	11	8.9	Q1
54	HNB/ZHA/2017/55/3	Co@NMC-800	11	8.8	Q1

55	HNB/WAN/2020/97/2	Co NPs/NC	11	8.8	Q1
56	HNB/GAO/2019/88/1	Co-NSPC-N	11	8.8	Q1
57	HNB/ZHA/2020/92/8	Pt/CeO2-P-600	11	8.8	Q1
58	HNB/WAN/2016/44/1	Co-Ni NW	11	8.8	Q1
59	HNB/WAN/2019/74/3	Fe-N-C-700	11	8.8	Q2
60	HNB/ZHA/2012/12/2	Pt/RGO	11	8.8	Q2
61	HNB/ZHA/2012/12/1	Pt/TiO2	11	8.8	Q2
62	HNB/LIU/2016/39/1	Ni-B/SiO2sol	11	8.8	Q2
63	HNB/ZHA/2007/128/2	Pt/AC HRT	11	8.7	Q2
64	HNB/ZHA/2007/128/1	Pt/CNTs HRT	11	8.7	Q2
65	HNB/SHI/2016/45/1	Pt/CNTs	11	8.7	Q2
66	HNB/DAI/2019/77/4	Co/NC-0.30	11	8.7	Q2
67	HNB/DU/2013/18/1	Pt@MIL-101 9000:1	10	8.7	Q2
68	HNB/SUN/2010/2/3	5wt% Pt/MWNT	10	8.6	Q2
69	HNB/SUN/2010/2/8	27.4wt%Pt/MWNT	10	8.6	Q2
70	HNB/WAN/2020/97/1	Co SAs/NC	11	8.6	Q2
71	HNB/COU/2016/40/3	0,3 wt% Pd/Al2O3/5	11	8.6	Q2
72	HNB/COU/2016/40/2	0,3 wt% Pd/Al2O3/1,85	11	8.6	Q2
73	HNB/SUN/2010/2/5	5wt% Pt/MWNT	10	8.6	Q2
74	HNB/SUN/2010/2/2	5wt% Pt/MWNT	10	8.6	Q2
75	HNB/COU/2016/40/1	1 wt% Pd/Al2O3	11	8.6	Q2
76	HNB/VAN/2019/79/2	Pt/N-BCNT	10	8.5	Q2
77	HNB/LIN/2019/84/2	0.25% Pt/α - MoC	10	8.5	Q2
78	HNB/LIN/2019/84/3	0.25% Pt/α - MoC	10	8.5	Q2
79	HNB/SUN/2010/2/4	5wt% Pt/MWNT	10	8.5	Q2
80	HNB/LAN/2008/123/1	Pd/FSA	10	8.5	Q2
81	HNB/SUN/2010/2/1	5wt% Pt/MWNT	10	8.4	Q2
82	HNB/LIN/2019/84/6	0.25% Pt/α - MoC	10	8.4	Q2
83	HNB/SUN/2010/2/9	10wt%Pt/C	10	8.4	Q2
84	HNB/MIA/2018/69/1	PtCo nanoflower	10	8.4	Q2
85	HNB/MOH/2012/9/1	Ni/SBA-15	10	8.4	Q2
86	HNB/DU/2013/18/2	Pt@MIL-101 36000:1	9	8.4	Q2
87	HNB/TIA/2021/108/1	Pt2/C3N4	10	8.4	Q2
88	HNB/MOH/2012/9/2	Ni/MgO	10	8.4	Q2
89	HNB/VAN/2019/79/1	Pd/N-BCNT	10	8.4	Q2
90	HNB/LIU/2020/101/6	NiMo-10.7	10	8.4	Q2
91	HNB/LV/2020/105/4	Ɣ-Fe2O3/NPC-900	10	8.4	Q2
92	HNB/LIU/2020/101/4	NiMo-10.7	10	8.4	Q2
93	HNB/SUN/2010/2/6	27.4wt%Pt/MWNT	10	8.3	Q2
94	HNB/LV/2020/105/5	Ɣ-Fe2O3/NPC-1000	10	8.3	Q2
95	HNB/WAN/2013/19/6	Pd/AC	10	8.3	Q2
96	HNB/WAN/2013/19/3	Pd/MWCNT-SA-3.6	9	8.3	Q2
97	HNB/WAN/2013/19/1	Pd/MWCNT-SA-6.0	9	8.3	Q2
98	HNB/MAH/2008/127/1	NiFC1	10	8.3	Q2
99	HNB/MAH/2008/127/2	NiFC2	10	8.3	Q2
100	HNB/DON/2017/57/1	Pd/CNT	9	8.3	Q2
101	HNB/DON/2017/57/2	Pd/NCNT	9	8.3	Q2
102	HNB/DU/2013/18/3	Pt@MIL-101 72000:1	9	8.2	Q2
103	HNB/VAN/2019/79/3	Rh/N-BCNT	10	8.2	Q2
104	HNB/MAH/2008/127/3	NiFC3	10	8.2	Q2
105	HNB/SUN/2010/2/10	5wt%Pt/C	10	8.2	Q2
106	HNB/WAN/2019/74/5	Fe-N-C-700	10	8.2	Q2
107	HNB/QU/2018/83/5	Pd-6Ni-N-C60	10	8.2	Q2
108	HNB/WAN/2020/97/3	Co NPs/AC	11	8.2	Q2
109	HNB/WAN/2013/19/5	Pd/MWCNT-IJM	9	8.2	Q2
110	HNB/WAN/2021/106/2	Pd/LDH1	10	8.2	Q2
111	HNB/DU/2013/18/4	Pt@MIL-101 72000:1	9	8.2	Q2

112	HNB/ZHA/2007/128/4	Pt/AC LRT	11	8.2	Q2
113	HNB/WAN/2010/1/2	Ni-5/$SiO_{2\text{-NI}}$	11	8.2	Q2
114	HNB/GAO/2019/88/5	Co-NSPC-N/N2	10	8.2	Q2
115	HNB/QU/2019/85/4	Ni-N-C60	10	8.2	Q2
116	HNB/QU/2017/49/3	Ni/C60-Ac-B4-30	10	8.2	Q2
117	HNB/LIN/2019/84/5	0.25% Pt/α - MoC	10	8.2	Q2
118	HNB/WAN/2019/74/4	Fe-N-C-800	10	8.2	Q2
119	HNB/EAS/2015/28/5	Ru-7/Fe3O4-ϒ Fe2O3	9	8.1	Q2
120	HNB/ZHA/2017/55/2	Co@NMC-700	10	8.1	Q2
121	HNB/DAI/2019/77/5	Co/NC-0.35	10	8.1	Q2
122	HNB/GAO/2019/88/6	Co-NSPC-N/KSCN	10	8.1	Q2
123	HNB/HAR/2012/13/1	Pd/PEG4000	10	8.1	Q2
124	HNB/RAJ/2012/10/1	Ni/rutile-500	10	8.1	Q2
125	HNB/LIU/2021/112/4	NiCu/C@SiO2-800	10	8.1	Q2
126	HNB/COU/2016/40/4	50 wt% NiO/Al2O3+SiO2	11	8.1	Q2
127	HNB/RAJ/2012/10/2	Ni/anatase-500	10	8.1	Q2
128	HNB/GAO/2019/88/2	Co-NSPC-C	10	8.1	Q2
129	HNB/XIO/2020/104/5	2Co-1Zn@NC-800	9	8.1	Q2
130	HNB/EAS/2015/28/6	Ru-12/ Fe3O4-ϒ Fe2O3	9	8.0	Q2
131	HNB/SUN/2010/2/7	50wt%Pt/MWNT	10	8.0	Q2
132	HNB/GAO/2019/88/7	Co-NSPC-N/110	10	8.0	Q2
133	HNB/QU/2018/71/1	AuPd/TiO2 (Mim)	10	8.0	Q2
134	HNB/ZHA/2017/55/4	Co@NMC-900	10	8.0	Q2
135	HNB/WAN/2019/74/6	Fe-N-C-800	10	8.0	Q2
136	HNB/DAI/2019/77/7	Co/NC-0.45	10	8.0	Q2
137	HNB/LI/2015/35/2	Pt/C	9	8.0	Q2
138	HNB/LIN/2019/84/4	0.25% Pt/α - MoC	10	7.9	Q2
139	HNB/ZHA/2020/102/1	Ag@SiO2 -CH2O	9	7.9	Q2
140	HNB/HU/2019/80/1	Co@CN-800	9	7.9	Q2
141	HNB/WAN/2019/74/2	Fe-N-C-600	10	7.9	Q2
142	HNB/RAJ/2012/10/3	Ni/TiO_2-500	10	7.9	Q2
143	HNB/WAN/2010/1/3	Ni-5/$SiO_{2\text{-AC}}$	11	7.8	Q2
144	HNB/GAO/2019/88/3	Co-NSPC-S	10	7.8	Q2
145	HNB/LIU/2021/112/5	NiCu/C@SiO2-850	9	7.8	Q2
146	HNB/QU/2018/71/7	AuPd/TiO2(Sim)	10	7.8	Q2
147	HNB/MIA/2018/69/2	PtCo nanoparticle	9	7.8	Q2
148	HNB/DAI/2019/77/3	Co/NC-0.25	10	7.8	Q2
149	HNB/LIN/2019/84/1	0.5% Pt/C	9	7.7	Q2
150	HNB/SUN/2018/68/1	Co@mesoNC	10	7.7	Q2
151	HNB/NIE/2020/93/10	Pt/m-Al2O3-600-40mg	9	7.7	Q2
152	HNB/SUN/2018/68/4	Co@mesoNC	10	7.7	Q2
153	HNB/ZHA/2021/113/2	0.075%Pt/SBA-15	10	7.7	Q2
154	HNB/NIE/2020/93/11	Pt/m-Al2O3-800-40mg	9	7.7	Q2
155	HNB/LIN/2012/11/3	Ni/TiO2@C-11%	10	7.7	Q2
156	HNB/WAN/2021/106/3	Pd/LDH0.8	9	7.7	Q2
157	HNB/LI/2019/90/1	FeOx@CN-hpes	9	7.7	Q2
158	HNB/GAO/2019/88/4	Co-NSPC-Cl	10	7.7	Q2
159	HNB/DON/2017/57/3	Pd/N@CNT-1.5	9	7.6	Q2
160	HNB/LIU/2020/101/7	NiMo-10.7-G	10	7.6	Q2
161	HNB/CHE/2018/64/1	Pd/MIL-101	9	7.6	Q2
162	HNB/CHE/2018/64/7	Pd/SiO3	9	7.6	Q2
163	HNB/CHE/2018/64/14	Pd/UiO-66	9	7.6	Q2
164	HNB/SUN/2018/68/3	Co@mesoNC	10	7.6	Q2
165	HNB/WAN/2021/106/6	Pd/SiO2	9	7.6	Q2
166	HNB/ZHA/2020/92/5	Pt/CeO2-C-300	9	7.6	Q2
167	HNB/LIU/2020/101/5	NiMo-8	9	7.6	Q2
168	HNB/ZHA/2022/117/3	Pd/PIL-PF6	9	7.6	Q2

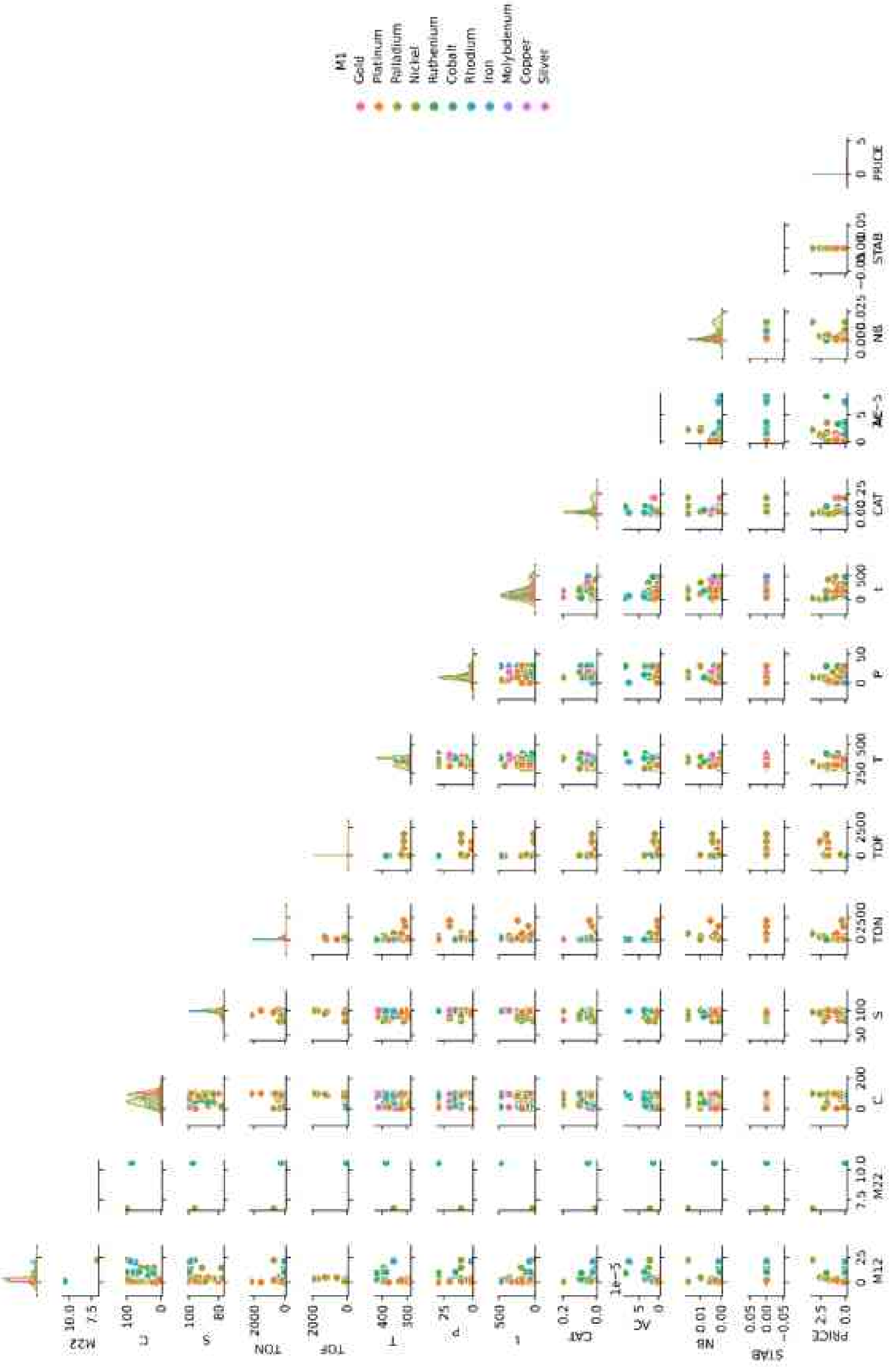
169	HNB/CHE/2018/64/10	Pd/ZIF-8	9	7.6	Q2
170	HNB/CHE/2018/64/12	Pd/HKUST-1	9	7.6	Q2
171	HNB/QU/2018/83/6	Pd-6Ni-COO-C60	9	7.5	Q2
172	HNB/GAR/2019/89/1	PdB	9	7.5	Q2
173	HNB/WAN/2021/106/5	Pd/C	9	7.5	Q2
174	HNB/CHE/2018/64/6	Pd/SiO2	9	7.5	Q2
175	HNB/CHE/2018/64/8	Pd/Υ-Al2O3	9	7.5	Q2
176	HNB/ZHA/2021/113/3	0.07%Pt/ZrO2	10	7.5	Q2
177	HNB/EAS/2015/28/7	Ru-14/FeO-Fe3O4	8	7.5	Q2
178	HNB/QU/2018/83/2	Ni-Pd	9	7.5	Q2
179	HNB/WAN/2021/106/4	Pd/LDH-700	9	7.5	Q2
180	HNB/ZHA/2022/117/2	Pd/PIL-Cl	9	7.5	Q2
181	HNB/ZHA/2021/113/4	0.09%Pt/γ-Al2O3	10	7.5	Q2
182	HNB/LIU/2020/101/3	NiMo-16	9	7.5	Q2
183	HNB/SUN/2018/68/6	Co@mesoNC	10	7.5	Q2
184	HNB/MAK/2012/4/1	Au/TiO_2	9	7.5	Q2
185	HNB/WAN/2021/106/1	Pd/LDH1.2	9	7.5	Q2
186	HNB/PEI/2020/103/4	Pt/Co-No-RT	9	7.5	Q2
187	HNB/QU/2017/49/4	Ni/C60-Ac-B4-30*	9	7.5	Q2
188	HNB/QU/2019/85/3	Ni-COO-C60	9	7.5	Q2
189	HNB/LIU/2021/112/3	NiCu/C@SiO2-750	9	7.5	Q2
190	HNB/PEI/2020/103/6	Pt/Co-CTAB-RT	9	7.5	Q2
191	HNB/MAH/2008/127/4	Rni	9	7.5	Q2
192	HNB/LIN/2012/11/1	Ni/TiO2	9	7.5	Q2
193	HNB/EAS/2015/28/4	Ru-11/Fe3O4-Υ Fe2O3	8	7.5	Q2
194	HNB/GEL/2002/119/3	Pd/CSXU	8	7.5	Q2
195	HNB/LIU/2021/112/6	NiCu/C@SiO2-900	9	7.4	Q2
196	HNB/EAS/2015/28/3	Ru-5/Fe3O4-Υ Fe2O3	8	7.4	Q2
197	HNB/QU/2017/49/2	Ni/C60-Ac-B4_10	9	7.4	Q2
198	HNB/GEL/2002/119/2	Pd/CA1	8	7.4	Q2
199	HNB/CHE/2018/64/13	Pd/HKUST-2	9	7.4	Q2
200	HNB/GEL/2002/119/1	Pd/CN1	8	7.4	Q2
201	HNB/QU/2018/71/3	Pd/TiO2(Mim)	9	7.4	Q2
202	HNB/PEI/2020/103/1	Pt/Co-No-60	9	7.4	Q2
203	HNB/SUN/2018/68/2	Co@mesoNC	10	7.4	Q2
204	HNB/QU/2018/83/4	Pd-6Ni-C60	9	7.4	Q2
205	HNB/CHA/2008/124/4	Ru/SBA-15	8	7.4	Q2
206	HNB/FAN/2014/22/1	Pt/AlO(OH)	8	7.4	Q2
207	HNB/DAI/2019/77/1	Co/NC-0.15	10	7.4	Q2
208	HNB/PEI/2020/103/2	Pt/Co-No-90	9	7.4	Q2
209	HNB/MAK/2012/4/2	Au/CeO_2	9	7.4	Q2
210	HNB/XIO/2020/104/3	2Co-1Zn@NC-900	8	7.4	Q2
211	HNB/HAR/2012/13/5	Pd/PEG4000	9	7.4	Q2
212	HNB/CHE/2018/64/2	Pd/MIL-101	9	7.4	Q2
213	HNB/PEI/2020/103/3	Pt/Co-No-90	9	7.4	Q2
214	HNB/LEN/2016/37/4	Ru/C60 10/1	9	7.4	Q2
215	HNB/LEN/2016/37/5	Ru/C60 20/1	9	7.4	Q2
216	HNB/CHE/2018/64/11	Pd/ZIF-8	9	7.4	Q2
217	HNB/CHA/2008/124/5	Ru/SBA-15	8	7.4	Q2
218	HNB/LEN/2016/37/6	Ru/C60 30/1	9	7.4	Q2
219	HNB/PEI/2020/103/5	Pt/Co-PVP-RT	9	7.3	Q2
220	HNB/QU/2018/71/6	AuPd/TiO2(Cim)	9	7.3	Q2
221	HNB/WAN/2019/78/2	Pd/PVA78000	9	7.3	Q2
222	HNB/HAR/2012/13/2	Pd/PEG4000	9	7.3	Q2
223	HNB/WAN/2014/23/1	Pd/Fe3O4 cat4r	8	7.3	Q2
224	HNB/LEN/2016/37/3	Ru/C60 5/1	9	7.3	Q2
225	HNB/CHA/2008/124/3	Ru/SBA-15	8	7.3	Q2

226	HNB/LEN/2016/37/7	Ru/C60 50/1	9	7.3	Q2
227	HNB/LIU/2019/81/13	Ni-Al2O3-C	9	7.3	Q2
228	HNB/ZHA/2020/92/7	Pt/CeO2-P-300	9	7.3	Q2
229	HNB/SUN/2018/68/5	Co@mesoNC	10	7.3	Q2
230	HNB/QU/2018/71/4	AuPd/MgO(Mim)	9	7.3	Q2
231	HNB/SHI/2016/45/5	5 wt% Pt/C	9	7.3	Q2
232	HNB/XU/2017/56/1	Co3S4	9	7.3	Q2
233	HNB/QU/2019/85/2	Ni-C60	9	7.3	Q2
234	HNB/QU/2017/49/1	Ni/C60-Ac-B4_1	9	7.3	Q2
235	HNB/CHE/2018/64/9	Pd/Ɣ-Al2O3	9	7.3	Q2
236	HNB/LIU/2020/101/1	Ni	10	7.2	Q2
237	HNB/WAN/2019/74/1	Fe-N-C-500	10	7.2	Q2
238	HNB/DAI/2019/77/9	Co/NC-0.30	9	7.2	Q2
239	HNB/LIU/2019/81/12	Ni-TiO2-C	9	7.2	Q2
240	HNB/ZHA/2017/55/9	Co@NC@SiO2-800	9	7.2	Q2
241	HNB/ZHA/2017/55/8	Co@NC-800	9	7.1	Q2
242	HNB/ZHA/2020/102/5	Ag@SiO2 -C7H6O	8	7.1	Q2
243	HNB/CHA/2008/124/2	Ru/SBA-15	8	7.1	Q2
244	HNB/LIN/2012/11/4	Ni/TiO2@C-29.2%	9	7.1	Q2
245	HNB/CHE/2018/64/3	Pd/MIL-101	9	7.1	Q2
246	HNB/LIU/2016/48/4	NiP-s-1h	8	7.1	Q2
247	HNB/HUA/2017/50/6	P123-Pd(OAc)2	8	7.1	Q3
248	HNB/EAS/2015/28/2	Ru-16/Fe3O4-Ɣ Fe2O3	8	7.1	Q3
249	HNB/LIU/2019/81/1	Ni@C	9	7.1	Q3
250	HNB/HAR/2012/13/6	Pd/PEG4000	9	7.1	Q3
251	HNB/LV/2020/105/13	Ɣ-Fe2O3 nanopowder	8	7.1	Q3
252	HNB/LIN/2012/11/2	Ni/TiO2@C-2%	9	7.1	Q3
253	HNB/LEN/2016/37/2	Ru/C60 2/1	9	7.1	Q3
254	HNB/WAN/2013/19/7	Pd/Al2O3	9	7.1	Q3
255	HNB/DAI/2019/77/8	Co/NC-0.30	9	7.0	Q3
256	HNB/HAR/2012/13/3	Pd/PEG4000	9	7.0	Q3
257	HNB/CHE/2018/64/15	Pd/UiO-66	9	7.0	Q3
258	HNB/EAS/2015/28/1	Ru-18	8	6.9	Q3
259	HNB/WAN/2016/44/1	Co-Ni NP	8	6.9	Q3
260	HNB/QU/2018/71/5	AuPd/C(Mim)	9	6.9	Q3
261	HNB/LIU/2019/81/14	Ni-ZrO2-C	9	6.9	Q3
262	HNB/LIU/2020/101/2	NiMo-32	9	6.9	Q3
263	HNB/HAR/2012/13/7	Pd/PEG4001	9	6.9	Q3
264	HNB/WAN/2013/19/4	Pd NPs- 4.3	8	6.8	Q3
265	HNB/QU/2018/71/2	Au/TiO2(Mim)	9	6.8	Q3
266	HNB/CHA/2008/124/1	Ru/SBA-15	8	6.8	Q3
267	HNB/ZHA/2017/55/12	Co@NMC-800 (1:2)	8	6.8	Q3
268	HNB/JIA/2021/107/1	Pd-Ni/Ɣ-Al2O3	7	6.7	Q3
269	HNB/WAN/2013/19/8	Pd/SiO2	9	6.7	Q3
270	HNB/LIU/2019/81/2	Co@C	9	6.7	Q3
271	HNB/ZHA/2017/55/5	Co/NMC-800	8	6.7	Q3
272	HNB/HAR/2012/13/4	Pd/PEG4000	9	6.7	Q3
273	HNB/LIU/2019/81/11	Ni-CeO2-C	9	6.7	Q3
274	HNB/ZHA/2017/55/7	Co@NMC-800-H2SO4	8	6.7	Q3
275	HNB/PRE/2021/110/2	Pd/GCF-AC1	7	6.7	Q3
276	HNB/TIA/2021/108/3	Pt NPs/C3N4	8	6.6	Q3
277	HNB/MOR/2017/53/1	FeS2	8	6.6	Q3
278	HNB/ZHA/2017/55/1	Co@NMC-600	9	6.6	Q3
279	HNB/TOR/2017/59/1	Au/TiO2NT	8	6.6	Q3
280	HNB/CHE/2018/64/5	Pd/MIL-101	7	6.6	Q3
281	HNB/TOM/2014/27/5	Ru/CNT	7	6.5	Q3
282	HNB/LIU/2019/81/6	Ni@C+Al2O3	8	6.5	Q3

283	HNB/ZHA/2020/102/2	Ag@SiO2 -C2H4O	7	6.5	Q3
284	HNB/CHE/2018/64/4	Pd/MIL-101	7	6.5	Q3
285	HNB/LIU/2019/81/9	Ni@C+Hydrotalcite	8	6.5	Q3
286	HNB/ZHA/2020/102/3	Ag@SiO2 -C3H6O	7	6.5	Q3
287	HNB/WAN/2013/19/9	Pd/MgO	8	6.5	Q3
288	HNB/LIU/2019/81/15	Au/CeO2	7	6.4	Q3
289	HNB/PRE/2021/110/1	Pd/GCF	7	6.4	Q3
290	HNB/LIU/2016/48/3	NiP-s-0.5h	7	6.4	Q3
291	HNB/LIU/2019/81/8	Ni@C+SiO2	8	6.4	Q3
292	HNB/LIU/2016/48/5	NiP-s-3h	7	6.4	Q3
293	HNB/ZUO/2017/66/3	PP-1:1-800	7	6.3	Q3
294	HNB/LIU/2019/81/7	Ni@C+TiO2	8	6.3	Q3
295	HNB/ZHA/2020/102/6	Ag@SiO2 -C7H6O0.1	8	6.3	Q3
296	HNB/LIU/2019/81/5	Ni/CeO2-in situ reduced	9	6.3	Q3
297	HNB/LEN/2016/37/1	Ru/C60 1/1	8	6.3	Q3
298	HNB/LIU/2016/48/2	NiP-s-10min	7	6.3	Q3
299	HNB/XIO/2020/104/2	1Co-1Zn@NC-900	7	6.3	Q3
300	HNB/ZHA/2017/55/11	Fe@NMC-800	8	6.2	Q3
301	HNB/SUN/2016/54/2	Ni-Fe-1/SiO2	7	6.2	Q3
302	HNB/SUN/2016/54/3	Ni-Fe-2/SiO2	7	6.2	Q3
303	HNB/XIO/2020/104/1	1Co-2Zn@NC-900	7	6.2	Q3
304	HNB/ZHA/2017/55/10	Ni@NMC-800	8	6.2	Q3
305	HNB/XIO/2020/104/4	1Co-0Zn@NC-900	7	6.1	Q3
306	HNB/LIU/2016/48/6	NiP-s-5h	7	6.1	Q3
307	HNB/ZHA/2017/55/6	CoOx@NMC-800	8	6.1	Q3
308	HNB/PAC/2011/3/4	C60-b	8	6.1	Q3
309	HNB/HUA/2017/50/5	P123-PdCl2	7	6.1	Q3
310	HNB/TIA/2021/108/2	Pt1/C3N4	7	6.1	Q3
311	HNB/LIU/2019/81/4	Ni/CeO2	9	6.0	Q3
312	HNB/LIU/2016/48/7	NiP-s-10h	7	6.0	Q3
313	HNB/LIU/2016/48/8	NiP-s-15h	7	5.9	Q3
314	HNB/LI/2017/61/1	MoS2 microflowers	6	5.9	Q3
315	HNB/QU/2018/83/3	Pd-N-C60	6	5.8	Q3
316	HNB/WAN/2019/78/3	Pd/PVA145000	7	5.8	Q3
317	HNB/YU/2000/125/1	Pd-B/SiO2(fresh)	6	5.7	Q3
318	HNB/WAN/2019/78/4	Pd/PVA205000	7	5.7	Q3
319	HNB/YU/2000/125/2	Pd-B/SiO2 (473 K)	6	5.7	Q3
320	HNB/QU/2019/85/5	Ni-N	6	5.7	Q3
321	HNB/LIU/2019/81/3	Ni@C+CeO2	8	5.7	Q3
322	HNB/ZHA/2020/102/4	Ag@SiO2 -C3H6O0.1	7	5.7	Q3
323	HNB/YU/2000/125/3	Pd-B/SiO2 (673 K)	6	5.7	Q3
324	HNB/QU/2019/85/1	Ni-B4	6	5.7	Q3
325	HNB/QIN/2013/15/12	Ni-Mo-P	6	5.7	Q3
326	HNB/WAN/2019/78/1	Pd/PVA47000	7	5.7	Q3
327	HNB/YU/2000/125/6	Pd/SiO2 (fresh)	6	5.7	Q3
328	HNB/DAI/2019/77/6	Co/NC-0.40	6	5.6	Q3
329	HNB/QIN/2013/15/7	Ni-Co-P	6	5.6	Q3
330	HNB/PRE/2021/110/3	Pd/GCF-AC2	6	5.6	Q3
331	HNB/PRE/2019/87/1	Pd/N-BCNT	6	5.6	Q3
332	HNB/MIA/2018/69/3	Commercial Pt/C	6	5.6	Q3
333	HNB/HUA/2017/50/4	P123-RuCl3*3H2O	6	5.6	Q3
334	HNB/QIN/2013/15/9	Ni-Zn-P	6	5.6	Q3
335	HNB/HU/2019/80/2	Fe@CN-800	6	5.6	Q3
336	HNB/YU/2000/125/4	Pd-B/SiO2 (873 K)	6	5.5	Q3
337	HNB/HU/2019/80/3	Ni@CN-800	6	5.5	Q3
338	HNB/LIU/2019/81/10	Co@C+CeO2	8	5.5	Q3
339	HNB/YU/2000/125/5	Pd-B/SiO2 (973 K)	6	5.5	Q3

340	HNB/XIO/2020/75/1	NOCNTs-d	6	5.5	Q3
341	HNB/QIN/2013/15/6	Ni-Fe-P	6	5.4	Q3
342	HNB/PAC/2011/3/3	C60-b	8	5.4	Q3
343	HNB/PAC/2011/3/2	C60	7	5.4	Q3
344	HNB/LIU/2021/112/2	Ni/C@SiO2-800	6	5.3	Q4
345	HNB/XIO/2020/75/5	NOCNTs-(Fe)	6	5.3	Q4
346	HNB/QIN/2013/15/8	Ni-Cu-P	6	5.3	Q4
347	HNB/QIN/2013/15/2	Ni-Al-P	6	5.2	Q4
348	HNB/ZUO/2017/66/2	PP-1:1-600	6	5.2	Q4
349	HNB/PAC/2011/3/5	C60-(Na)	7	5.2	Q4
350	HNB/PAC/2011/3/1	C60	7	5.2	Q4
351	HNB/LIU/2021/112/7	FeCu/C@SiO2-800	6	5.2	Q4
352	HNB/WAN/2019/74/9	Fe/N-C-700	6	5.1	Q4
353	HNB/HUA/2017/50/3	P123-RhCl3	6	5.1	Q4
354	HNB/DAI/2019/77/2	Co/NC-0.20	6	5.1	Q4
355	HNB/HU/2019/80/4	Cu@CN-800	6	5.1	Q4
356	HNB/LIU/2021/112/1	NiCu@SiO2-H2-800	6	5.1	Q4
357	HNB/LIU/2021/112/8	CoCu/C@SiO2-800	6	5.0	Q4
358	HNB/LIU/2016/48/1	Ni2P-f	6	5.0	Q4
359	HNB/TOM/2014/27/4	Pt/C	5	5.0	Q4
360	HNB/ZUO/2017/66/6	PP-1:3-800	6	4.9	Q4
361	HNB/KAT/2012/8/2	Pt/TiO_2	5	4.9	Q4
362	HNB/KAT/2012/8/3	Pt/C	5	4.9	Q4
363	HNB/QU/2018/83/1	Pd	6	4.9	Q4
364	HNB/ZUO/2017/66/5	PP-1:3-600	6	4.9	Q4
365	HNB/LI/2006/126/1	Meso Ni-B	6	4.8	Q4
366	HNB/HUA/2017/50/1	P123-CoCl2	6	4.8	Q4
367	HNB/XIO/2020/75/2	NOCNTs-c	5	4.8	Q4
368	HNB/HUA/2017/50/2	P123-NiCl2	6	4.8	Q4
369	HNB/LI/2006/126/2	Regular Ni-B	6	4.8	Q4
370	HNB/XIO/2020/75/3	NOCNTs-b	5	4.8	Q4
371	HNB/QIN/2013/15/5	Ni-Mn-P	5	4.7	Q4
372	HNB/QIN/2013/15/1	Ni-P	5	4.7	Q4
373	HNB/WAN/2019/74/8	Fe-C-700	6	4.7	Q4
374	HNB/QIN/2013/15/3	Ni-Ca-P	5	4.7	Q4
375	HNB/QIN/2013/15/4	Ni-Mg-P	5	4.7	Q4
376	HNB/XIO/2020/75/4	NOCNTs-a	5	4.7	Q4
377	HNB/SUN/2016/54/1	Fe/SiO2	5	4.6	Q4
378	HNB/TOM/2014/27/3	Pd/C	5	4.6	Q4
379	HNB/ZUO/2017/66/4	PP-1:3-400	6	4.6	Q4
380	HNB/ZUO/2017/66/1	PP-1:1-400	6	4.6	Q4
381	HNB/QIN/2013/15/10	Ni-La-P	5	4.5	Q4
382	HNB/QIN/2013/15/11	Ni-Ce-P	5	4.5	Q4
383	HNB/TOR/2013/16/1	Au/TiO2-T	5	4.5	Q4
384	HNB/TOR/2013/16/4	Au/SiO2-T	5	4.5	Q4
385	HNB/TOR/2013/16/5	Au/SiO2-H2	5	4.4	Q4
386	HNB/TOR/2013/16/2	Au/TiO2-H2	5	4.4	Q4
387	HNB/TOR/2013/16/3	Au/TiO2-U	5	4.4	Q4
388	HNB/TOM/2014/27/1	Ru/C	5	4.3	Q4
389	HNB/TOR/2013/16/6	Au/SiO2-U	5	4.2	Q4
390	HNB/WAN/2019/74/7	N-C-700	5	3.9	Q4
391	HNB/TOM/2014/27/2	Rh/C	5	3.6	Q4

7. SCATTER PLOT OF THE SELECTED PARAMETERS – NITROBENZENE HYDROGENATION CATALYSTS



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Hálával tartozom a Kémiai Intézet katalitikus hidrogénezéssel foglalkozó csoportjának, akik hozzájárultak a doktori munkám tárgyát képező MIRA21 modell létrehozásához. Köszönöm, Vanyorek Lászlónak, Sikora Emőkének, Prekob Ádámnak és Hajdú Viktóriának a segítségét és együttműködését. Köszönöm Dr. Fiser Bélának és Dr. Szőri Milán egyetemi munkatársaknak, akik támogattak és értékes észrevételeket tettek.

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fejlesztésében való közreműködésért. Hálásan köszönöm továbbá Dr. Bánhidi Olivér, címzetes egyetemi tanár támogatását és online tanóráit, amelyek bevezettek az adatelemzés világába.

Az oktatók és nem oktató munkatársak a Kémiai Intézetben kulcsfontosságúak voltak az utam során. Mindig segítőkészen álltak rendelkezésemre, legyen szó akár technikai kérdésekről, akár szakmai támogatásról. Hálával tartozom értük!

Nem tudom eléggé kifejezni, milyen hálás vagyok a férjemnek. Mindvégig mellettem állt ezen az úton, türelmesen meghallgatta az aggodalmaimat és örömömet, és mindig biztatott a kitartásra. Ő mindig ott volt az oldalamon, hogy erőt adjon, amikor szükségem volt rá, és örömömre osztozzon az eredményeimben. A férjem szeretete és támogatása a legnagyobb kincs az életemben.

Az én két kisgyermekemnek is hálával tartozom, amiért megértéssel fogadták, amikor nem tudtam velük lenni, és támogattak engem azzal, hogy maguk is erőt adtak. Az ő ártatlan örömeik és szeretetük folyamatos inspirációt nyújtanak, és mindig emlékeztetnek arra, hogy miért is vállaltam mindezt. A családomnak mély hálával tartozom mindazért, amit értem tettek. Az ő segítségük nélkül jóval bonyolultabb lett volna az utam.

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