


**SPECIAL ISSUE** Bridging the Gap – Advancing Electrochemical Power-to-X Technologies towards Industrial Application

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# A Well-Advanced High-Throughput Test System for Electrocatalytic Screening Applications Under Industrial Relevant Conditions – A Perspective to Accelerate Electrolysis Research and Development

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## ABSTRACT

Electrolysis is a dynamic research area in which both mature and new promising processes, such as alkaline water electrolysis and electrochemical CO<sub>2</sub> reduction, are under enormous development pressure due to their high relevance for the energy sector. High-throughput (HT) technologies are efficient screening platforms that can accelerate research activities and significantly shorten development times. Over the past 25 years, various HT platforms have found their way into electrochemical research. These typically have one or more major disadvantages: they are characterized by abstract experimental conditions, designed for a specific application or process, or generate insufficiently comparable data. In this publication, we present a newly developed HT test system that enables the parallel operation of 16 electrochemical bench-scale flow cells under industry-relevant test conditions. The specially developed modular flow cell can be operated variably in the fully automated system and allows research into the most common applications in electrochemistry for many different processes with a focus on all relevant variants of water electrolysis and electrochemical CO<sub>2</sub> reduction. Both the HT system and the developed flow cell are designed to accelerate the generation of reliably reproducible data with high comparability in order to strengthen scientific exchange. The fully automated process control, online analysis and programmable feedback loops of the HT test system provide great potential for the design of experiment strategies. The implementation of Design of Experiment strategies will maximize the testing efficiency of this innovative research system.

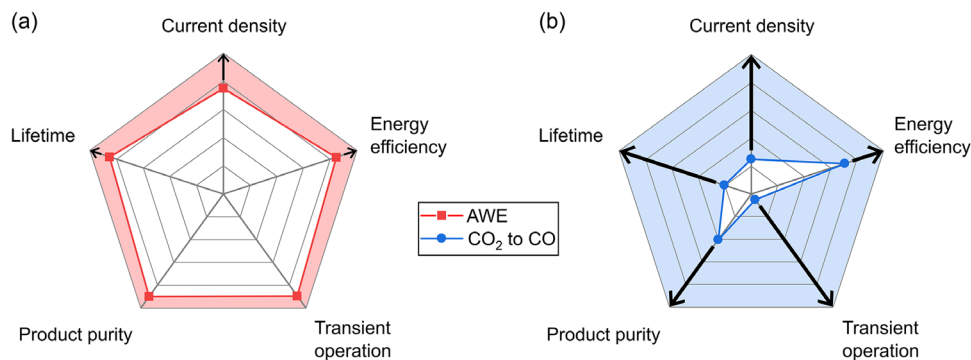
## 1 | Introduction

Electrolysis will play a pivotal role in the global energy transition toward achieving net zero carbon emissions. The production of green hydrogen from water electrolysis, powered by renewable

energy sources, is expected to be a cornerstone of this transition [1, 2]. Further Power2X processes, such as the electrochemical reduction of CO<sub>2</sub>, have received increasing scientific attention over recent years [3]. These innovative electrolysis technologies hold promise for the conversion of the greenhouse gas CO<sub>2</sub> into

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**FIGURE 1** | Optimization potentials (coloured areas) of key performance indicators (KPI) for next-generation alkaline water electrolysis (AWE, red squares) and CO<sub>2</sub> to CO (blue circles) electrolyzers. Net charts indicate the technology-specific current status of the individual KPIs on the scale of the targeted optimization goals for current density, lifetime, energy efficiency, product purity, and transient operation. State-of-the-art values are dimensionless and plotted against KPI values expected in the future [7, 9].

fuels, valuable products, or chemical feedstocks, contributing to the development of a sustainable closed-carbon cycle [4–6].

The development of next-generation electrolyzers represents a complex challenge, as technological solutions rely on the concurrent development of new materials, compounds, cells, and stacks with improved properties. At the same time, the performance of parameters must be optimized in multiple directions. This requires consideration of complex interdependencies between various aspects of electrolyzer technology. Particularly relevant key performance indicators are shown in Figure 1 using the example of two electrolysis processes. Alkaline water electrolysis (AWE) is a well-established and mature process. As politics, science and society attribute an ever-increasing role to hydrogen, researchers are working on further optimizing the AWE process. According to estimates, AWE has the greatest optimization potential with respect to current density, lifetime, and energy efficiency. However, product purity and the operable load range (transient operation) are expected to offer less scope for improvement (Figure 1a) [7]. CO<sub>2</sub> electrolysis, on the other hand, is still at more of a fundamental research stage. Among different possible reduction products, the electrochemical reduction of CO<sub>2</sub> to CO is the most promising due to the simple reaction mechanism [8]. Figure 1b shows the low technological readiness of this process, which requires improvement of the full scope of KPIs (current density, lifetime, energy efficiency, transient operation, and product purity) [9]. There is hardly any literature investigating the transient operation of CO<sub>2</sub> to CO electrolysis.

This publication introduces an innovative high-throughput (HT) test system for electrolysis research with a focus on AWE and CO<sub>2</sub> electrolysis. With the unique experimental possibilities, this system stands out significantly from previous systems. This work outlines a perspective in which HT research is fundamentally expanded and driven to a more advanced level.

First, the current state of HT research is assessed and the recurring disadvantages of previous systems are analyzed. Second, the system approaches are described, which were guiding during the system engineering and will be decisive for the desired progress with this development in HT research. This is followed by a detailed presentation of the HT system and HT electrolysis

flow cell developed, as well as initial experimental data. Finally, the potential of the application of Design of Experiment (DoE) strategies, the diverse process and application possibilities of the system as well as the potential for accelerating electrolysis research are discussed.

## 2 | High-Throughput Experimentation

HT experimentation refers to methods and systems designed to perform automated, accelerated testing and analyses of an increased number of samples or process conditions. It involves either rapid serial or parallel workflow processing, thereby, significantly accelerating research efforts and, hence, minimizing the time-to-market, e.g., novel catalysts, materials, compounds, or processes [10, 11]. HT technology has its roots in pharmaceutical research [12] and was soon adopted in research and development in areas such as materials science [13] or heterogeneous catalysis [14]. For electrochemical processes, additional electrochemistry-related parameters substantially increase the complexity of the parameter space compared to conventional catalysis. To address this complexity, HT technologies are a promising experimental approach for research and development in electrochemical research [15]. However, the extended parameter space and complexity of electrochemical systems hampered the emergence of such technologies in this field [16, 17]. The first electrochemical HT developments were batch systems with an array structure approach, for example, based on microtiter plates or wafers, with which the individual samples are automatically and serially scanned in X-Y direction [18–20]. They enable the testing of either one electrolyte vs. many electrodes [21–25] or one electrode vs. many electrolytes [26, 27]. Other systems perform HT batch screening by running experiments in parallel [28–32]. HT batch systems are also commercially available [33–35]. The references listed report on static systems without convective electrolyte flow [21–35]. Later, HT systems with electrochemical flow cells were developed [36–40].

The electrocatalytic HT systems developed up until now have some major disadvantages. The batch cell systems described in the literature are strongly diffusion-controlled and typically provide only the testing of undivided cells. The maximum current densities are far below the range of interest for industrial

applications. Flow cell systems, on the other hand, achieve higher current densities and have improved concentration gradients due to the electrolyte flow through the cell. However, due to miniaturization, they are characterized by laminar flow and in some cases also only provide undivided cells. Electrocatalysis on a production scale is generally practiced in divided flow cells at high current densities, in which turbulent mixing is common [15]. Compared to those industrial standards, previously developed HT systems in electrochemical research provide abstract experimental conditions. However, testing under industry-relevant conditions is crucial to evaluate the operational capability of processes or materials for industrial applications.

Another disadvantage is that previous electrocatalytic HT systems were developed specifically for certain applications or processes. This specific design typically impedes or even prevents the investigation of other applications or processes with these systems. This severely limits the variability of previous systems. Due to its high relevance, electrocatalysis is a highly dynamic research field. The focus of research is in constant discourse in science and politics and can therefore readily change. High system variability is essential to fully exploit the potential of a HT system in the diverse and dynamic field of electrochemical research.

Insufficient comparability of experimental results with literature data is a well-known issue in electrochemistry and hinders progress in electrochemical process development [41–43]. An additional disadvantage of previous HT systems is that both abstract experimental conditions and application- or process-specific system designs impair the comparability of the data generated.

Abstract experimental conditions, limited system variability and impaired data comparability are major disadvantages of the previously developed HT systems in electrochemical research. These disadvantages primarily arise from their simplified design, which is typically intended to reduce development time and investment costs. Such systems have proven to be an effective solution for early-stage screening in particular pre-defined applications and processes. However, the disadvantages discussed hinder a HT platform from unleashing its full potential under the current requirements of electrochemical research.

Over the past four years, based on the objectives and consultancy of IET-1 (Jülich, Germany) as well as the engineering and construction of hte GmbH (Heidelberg, Germany) an innovative HT system tailored to electrolysis processes with a focus on water electrolysis and electrochemical CO<sub>2</sub> reduction processes was developed [44]. The main objective of this project is to provide a HT environment for bench-scale flow cells to accelerate the development activities of advanced laboratory-scale processes under industry-relevant conditions toward higher technology readiness. Rather than focusing on one process or application, our HT system is designed to operate with the most common applications and processes in electrochemistry, offering a high degree of variability. The aim is to provide a research platform generating reliable and reproducible high-quality data, which can be readily compared with common literature data to strengthen the scientific exchange.

### 3 | System Design Approach

The developed HT system is designed to accelerate testing under industry-relevant conditions and to provide high system variability, data comparability as well as reproducibility. This also required the development of a new electrochemical flow cell. To meet all requirements, the system design is based on four key principles: parallelization, automation, modularization, and standardization. These key principles guided the development process of the entire system as well as the flow cell and are described in more detail in the corresponding sections in the following chapters.

The HT approach is realized in the developed system through parallel screening. The parallel operation of up to 16 flow cells simultaneously will significantly reduce the screening time compared to conventional setups.

All important process and electrochemical parameters are controlled or measured fully automated. The high degree of automation allows systematically programmed test protocols to be executed without human interaction. This improves reproducibility and comparability, further minimizes screening time and creates ideal conditions for the implementation of advanced statistical design of experiment strategies.

The developed HT system and electrochemical flow cell have a modular design. Both can be adapted to changing research requirements, enabling the screening of a wide range of applications and processes for maximum variability.

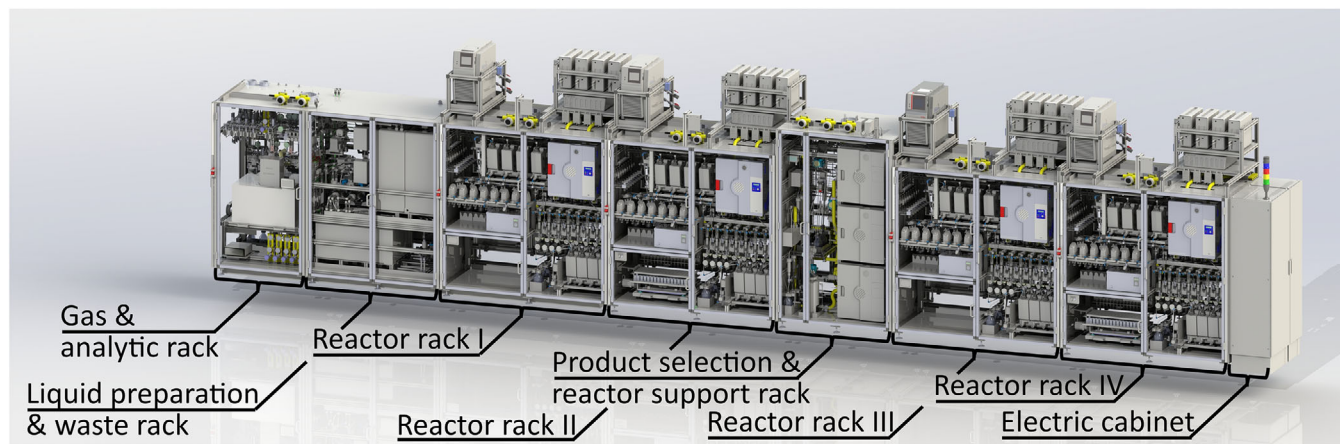
The use of standardized components for the system and the development of a flow cell according to the standards of commercially available laboratory cells will significantly improve the reproducibility and comparability of the generated data. Furthermore, documentation and test protocols will also be standardized.

#### 3.1 | High-Throughput Test System

The developed HT system consists of different system module racks (Figure 2). Due to the modular design, the configuration of the system can be adapted to the individual research requirements.

The gas and analytic rack consists of gas modules for the supply of N<sub>2</sub> and CO<sub>2</sub> process gases as well as the gas chromatograph (GC). Furthermore, it provides corresponding GC gas supply infrastructure for the automated measurement of gaseous product samples supplied by the product selection/reactor support rack.

The liquid preparation and waste rack consists of pumps, two large electrolyte feed tanks for anode and cathode respectively as well as a large waste tank on the bottom of the rack. By an automated electrolyte management system, electrolytes with defined conductivities for both anode and cathode can be automatically prepared, refilled, distributed to individual feed tanks in the reactor racks and disposed of in a waste tank at the end of its usage. This way the operator does not get in contact with the electrolyte throughout the operation of the HT system.



**FIGURE 2** | Three-dimensional (3D) layout of the 16-fold parallelized high-throughput system for electrolysis. The image was provided by hte GmbH (Heidelberg, Germany) for publication.

In the product selection and reactor support rack, the gaseous product streams of each electrochemical cell are selected in a multiport valve and sequentially sent to the GC for online analysis. The other streams are passed to the exhaust. This module also contains a humidification section for the gas feed.

The entire HT system comprises a total number of 16 bench-scale flow cells. The flow cells are operated within the reactor racks I–IV, each containing four electrolyzers. The distribution over four individual reactor racks allows the parallel operation of only 4, 8, or 12 cells and the setting of different process parameters in each reactor rack. A highly simplified flow sheet for a single cell is depicted in Figure 3. Each cell has an individual anolyte and catholyte feed tank. The feed tanks are automatically filled with the prepared electrolytes from the large storage tanks of the liquid preparation and waste rack. Two operating modes are available for the electrolyte flow. In single-pass mode, the electrolyte flows from the feed tank through the electrolyzer and phase separator directly to the waste tank. In recycle mode, the electrolyte returns to the feed tank after phase separation to recycle the electrolyte solutions. The electrolyte cycles of all 32 half-cells are fully separated. Hereby, liquid products can be enriched in recycle mode, which facilitates the detection of liquid products close to the detection limit. Furthermore, the emergence of stray currents within the system is prevented [45]. The diaphragm pumps enable cell-specific flow rates in the range of 1–100 mL/min. Each electrolyte stream can be preheated in a heat exchanger up to 80°C before it is pumped through the flow cell. For optimum temperature control, each reactor rack has a cell oven that heats the cell housing, connectors, and supply pipes to the pre-defined process temperature. The process pressure in the HT system can be set in the range from 0 to 2 barg. The pressure is controlled individually for each half-cell. For the use of gas diffusion electrodes, a gas feed can additionally be fed to the flow cell. This is particularly important when performing CO<sub>2</sub> electrolysis in the system where a CO<sub>2</sub> feed is required. Both the anolyte and catholyte product streams are equipped with individual phase separators downstream of the flow cell. An autosampler enables automated liquid sampling for offline liquid analyses after phase separation. The gaseous product phases are conveyed to the product selection rack for online GC analysis. The online GC enables one measurement every 8 min, resulting

in a measurement frequency of approximately 2 h/cell for a 16-fold system. The use of online GC analyses and ex-situ liquid analyses is indispensable for the planned system operation with CO<sub>2</sub> electrolysis processes [46].

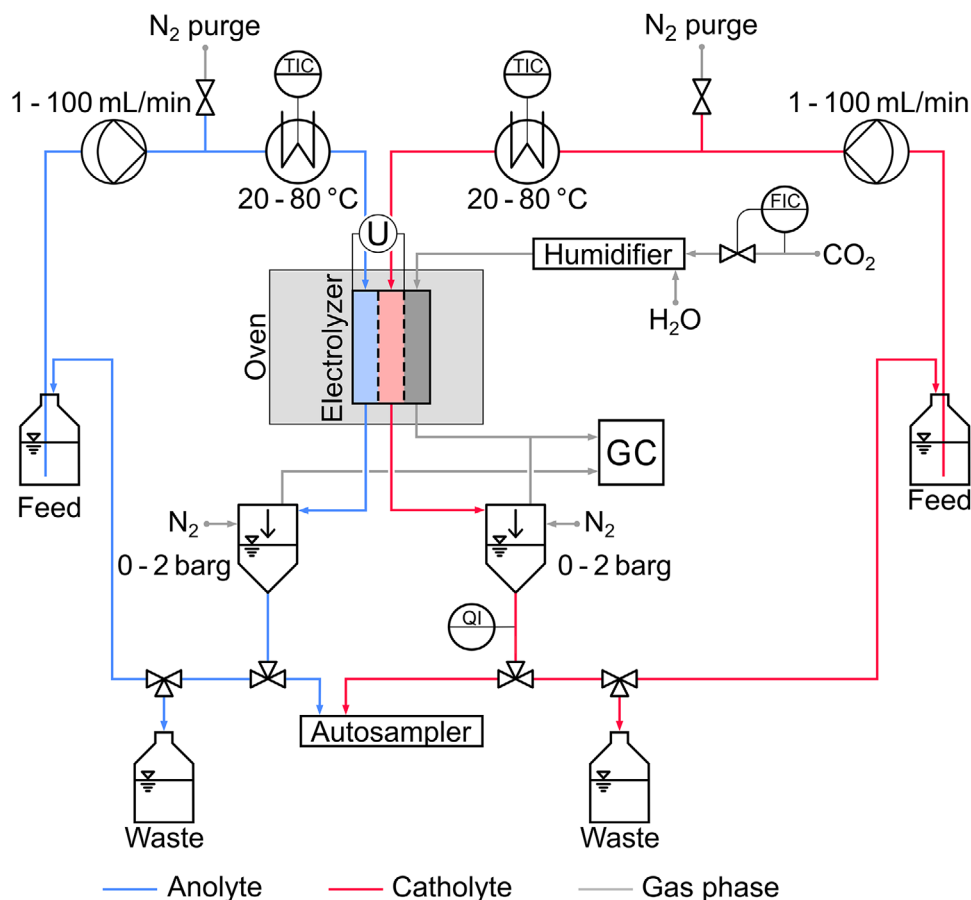
Each reactor rack is equipped with a multi-channel potentiostat from Metrohm AG. Four channels of each multi-channel potentiostat are equipped with a bipotentiostat module, a booster and electrochemical impedance spectroscopy modules. Further modules can be retrofitted into the multi-channel potentiostats depending on the application requirements.

The fully automated HT system is controlled by the hteControl4 software from hte GmbH. A new interface to the NOVA software package was developed for the system and implemented in hteControl4™. Together with the new interface, it is possible to create automated feedback loops and perform parameter adjustments for all process and electrochemical parameters in parallel operation mode.

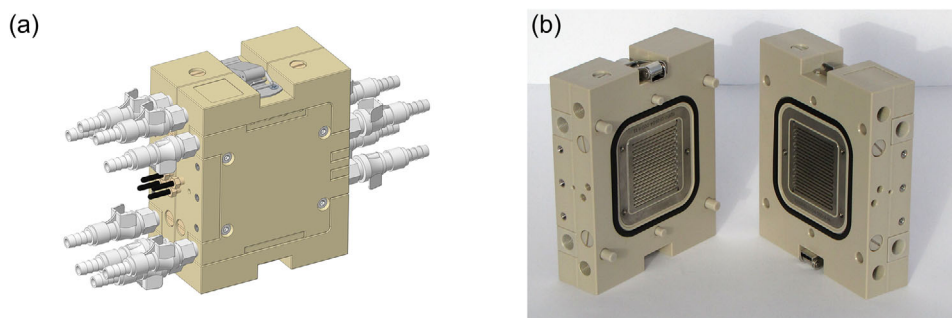
The system must be designed to operate with chemically various electrolytes, as it will be used to investigate different processes (AWE and CO<sub>2</sub> electrolysis) [46–48]. To prevent metal ion contamination due to chemical corrosion of steel components in the electrolyte circuit, all system components in contact with the electrolyte (pipes, valves, sensors, etc.) are therefore made of inert polymers.

### 3.2 | Modular Electrochemical Flow Cell

A modular electrochemical flow cell tailored to the requirements of the introduced HT system was developed in collaboration between hte GmbH (Heidelberg, Germany), Fraunhofer Institute for Microengineering and Microsystems (IMM, Mainz, Germany), and Institute of Energy Technologies (IET-1, Jülich, Germany) [44]. A 3D CAD image of this cell is shown in Figure 4a. The design is oriented toward standards of commercially available electrochemical flow cells in order to produce well-comparable data. The developed cell has a modular design. Individual components are therefore interchangeable or optionally applicable, allowing a variety of cell configurations to be realized for a high



**FIGURE 3** | Simplified flow sheet of the high-throughput (HT) system for one electrolyzer cycle. Abbreviations: temperature indicator controller (TIC), flow indicator controller (FIC), pressure indicator controller (PIC) and quality indicator (QI). QI indicates the conductivity sensor. The depicted device symbols are according to ISO 10628-1.

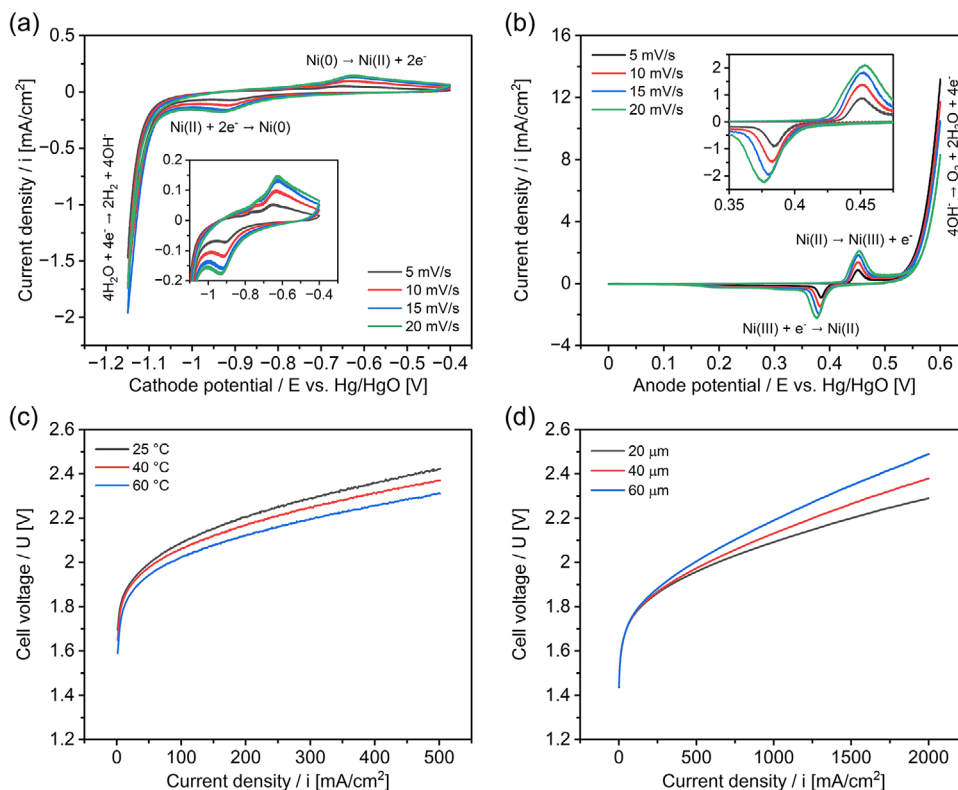


**FIGURE 4** | The newly developed flow cell for the high-throughput (HT) system. A 3D CAD illustration of the closed HT cell (a) and a photo of opened half cells built with zero-gap inlays (b). The flow cell was developed in collaboration between hte GmbH (Heidelberg, Germany), Fraunhofer Institute for Microengineering and Microsystems (IMM, Mainz, Germany) and Institute of Energy Technologies (IET-1, Jülich, Germany). The image of Figure 4a was provided by the Fraunhofer Institute for Microengineering and Microsystems (IMM, Mainz, Germany) for publication.

process and application variability. The design facilitates a simple assembly in order to minimize the risk of operator errors aimed at maximizing the reproducibility of the results.

Each of the two half-cell housings has four fluid connections on the front and two on the rear. The front provides an inlet and outlet connection for the electrolyte and an additional inlet and outlet for a gas phase. The gas phase connections are required for gas diffusion electrodes, e.g., to perform CO<sub>2</sub> electrolysis. The rear provides connections for the supply of a heat transfer

medium which can be fed through the current collector (CC), for additional temperature control. The cell is equipped with two interchangeable CC types: the zero-gap CC and the gap CC. The zero-gap type is required for processes with membrane electrode assemblies (MEA) or thin mesh electrodes. The flow cell equipped with zero-gap CCs is shown in Figure 4b. The gap CC can be used for GDEs, foil electrodes, or voluminous electrodes such as metal foams. The design provides unique scaling capabilities with a well-advanced flow cell in a HT environment. Each half-cell can be equipped with reference



**FIGURE 5** | Different electrocatalytic test procedures in different configurations of the high-throughput (HT) cell. Cathodic (a) and anodic (b) cyclic voltammetry (CV) (0.2 mV step size) with alkaline water electrolysis at different scan rates using 3-electrode setup in gap configuration with Ni foils, a Hg/HgO reference electrode, and Zirfon UTP 220 in 6 M KOH at 25°C. Polarization curve (c) (50 mA/s scan rate, 5 mA step size) with alkaline water electrolysis at different process temperatures in zero-gap configuration using Ni meshes and Zirfon UTP 220 in 6 M KOH. Polarization curve (d) (50 mA/s scan rate, 5 mA step size) in zero-gap configuration using anion exchange membrane water electrolysis with PiperION membranes of different thicknesses and Ni-based catalyst coated gas diffusion layers provided by Dioxide Materials at 60°C.

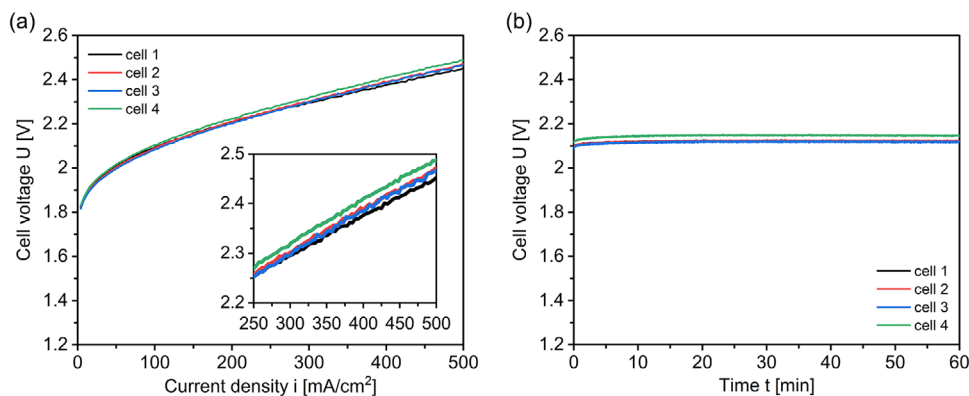
electrodes (RE). The electrical connections for connecting the potentiostat to the CCs are on the rear of the cell.

The sealing concept of the flow cell is based on industry standards for stacked flow cells. Each reactor rack is equipped with a piston press. With this press, all four cells in each reactor rack are axially compressed. This approach facilitates rapid installation of the flow cells into the system and uniform force distribution for improved electrode contacting. Furthermore, it enables the application of elevated system pressures.

The housing, screw fittings and frames for electrode area adjustment are made of inert polymers. The use of inert polymers allows the application of highly corrosive electrolyte solutions and thus increases the process variability. The CCs are made of titanium grade 5, but can also be fabricated from other materials such as nickel alloys or stainless steel. A coating of the CC surfaces can be applied optionally. For optimized experimental results, the CC and optional surface coating material must be matched to the electrolysis process.

The developed electrochemical cell was intensively tested during the single-cell prototyping phase. For this purpose, cyclic voltammogram (CV) measurements were conducted in gap configuration with a 3-electrode setup for AWE. Polarization curves were recorded in zero-gap configuration with a 2-electrode setup for AWE and anion exchange membrane (AEM) water electro-

lysis. Figure 5 shows the results of the single-cell prototyping. The 3-electrode setup is an important approach for catalyst and electrode research. The setup was tested here with a Hg/HgO RE, Ni foils and Zirfon UTP 220 in 6 M KOH. Figure 5 shows the CVs of the Ni foil at potentials in the range of the hydrogen evolution (Figure 5a) and the oxygen evolution (Figure 5b). The CVs were performed at different scan rates from 5 to 20 mV/s and a step size of 0.2 mV. Both CVs show the expected oxidation and reduction peaks: Ni(0) to Ni(II) and Ni(II) to Ni(0) at the cathode and Ni(II) to Ni(III) and Ni(III) to Ni(II) at the anode. As the scan rate increases, the magnitudes of the peak currents decrease. Below  $-1.1 V_{\text{Hg/HgO}}$  the hydrogen and above  $0.55 V_{\text{Hg/HgO}}$  the oxygen evolution reaction starts at the cathode and anode, respectively. The CV results measured with our developed modular flow cell are in good agreement with literature values produced with commercially available flow cells under similar conditions [49, 50]. The zero-gap configuration was tested in a 2-electrode setup. Two different systems were used for this purpose. Figure 5c shows the polarization curves at different cell temperatures using AWE with Ni meshes and Zirfon UTP 220 in 6 M KOH. The installation of MEAs in the zero-gap configuration was validated using AEM water electrolysis (Figure 5d). For this test, PiperION AEMs of different thicknesses were tested in 1 M KOH with Ni-based catalyst-coated gas diffusion layers provided by Dioxide Materials [51]. The surface area of the electrodes was reduced to  $5 \text{ cm}^2$  in both setups. Increasing the process temperature in the AWE (Figure 5c) and reducing the membrane



**FIGURE 6** | Reproducibility test with the 4-cell high-throughput (HT) prototype under nominally identical conditions. All cells were operated with alkaline water electrolysis in zero-gap configuration using Ni meshes and Zirfon UTP 220 in 6 M KOH at room temperature. Polarization curve with 50 mA/s scan rate and 20 mA step size (a) as well as chronopotentiometry at 100 mA/cm<sup>2</sup> for one hour (b).

thickness in the AEM WE (Figure 5d) lead to the expected reduction in cell voltage. Onset potential and cell resistance are lower with the AEM water electrolysis than with the AWE. The presented results confirm the high quality of the experimental data generated with the developed flow cell. In the next step, the use of gas diffusion electrodes is to be tested in order to validate the applicability of the cell for CO<sub>2</sub> electrolysis. Experiments on CO<sub>2</sub> electrolysis have not yet been performed, however this application has been included in the system design and its physical evaluation.

### 3.3 | 4-Cell Prototype System

A prototype was built with a single reactor rack, providing four cells in parallel operation (cf. 3.1. High-Throughput Test System). The purpose of performing preliminary tests on the HT prototype was to test the technological practicability and basic functions. Thereby, issues or necessary optimizations to the system, cell, or software could be identified and resolved at an early stage. In addition, the prototype was used to validate the reproducibility of the results generated with four cells operating in parallel. For this purpose, all cells were operated under nominally identical conditions, that is, with AWE in zero-gap configuration using Ni-Meshes and Zirfon UTP 220 in 6 M KOH.

Figure 6 shows the results of the 4-fold reproducibility test. First, polarization curves up to 500 mA/cm<sup>2</sup> (scan rate 50 mA/s and step size 20 mA) were measured in all cells (Figure 6a). The current density range was selected based on state-of-the-art KPIs of AWE [7]. Since the shape and values of the measured polarization curve depend on the reaction kinetics, ohmic resistance, and mass transport losses, this method is well suited to derive first conclusions on the reproducibility [52]. The cell voltages of cell 2 and cell 3 show minimal deviation over the entire current density range and thus excellent reproducibility. The average cell voltage at 500 mA/cm<sup>2</sup> is 2.471 V with a standard deviation of 14.7 mV. Deviations from this value are -20.1, 1.9, -3.0 and 21.1 mV for cell 1, cell 2, cell 3 and cell 4 respectively. Figure 6a shows a higher onset potential in cell 4, which shifts the voltage curve marginally upwards. The polarization curve of cell 1, on the other hand, has a lower slope leading to slight deviations at high current densities. Figure 6b shows a subsequent chronopotentiometry at

100 mA/cm<sup>2</sup> for 1 h. The data show a negligible deviation between cell 1, cell 2 and cell 3 at 100 mA/cm<sup>2</sup>, however, a shift of about 25 mV for cell 4, confirming the observations described above.

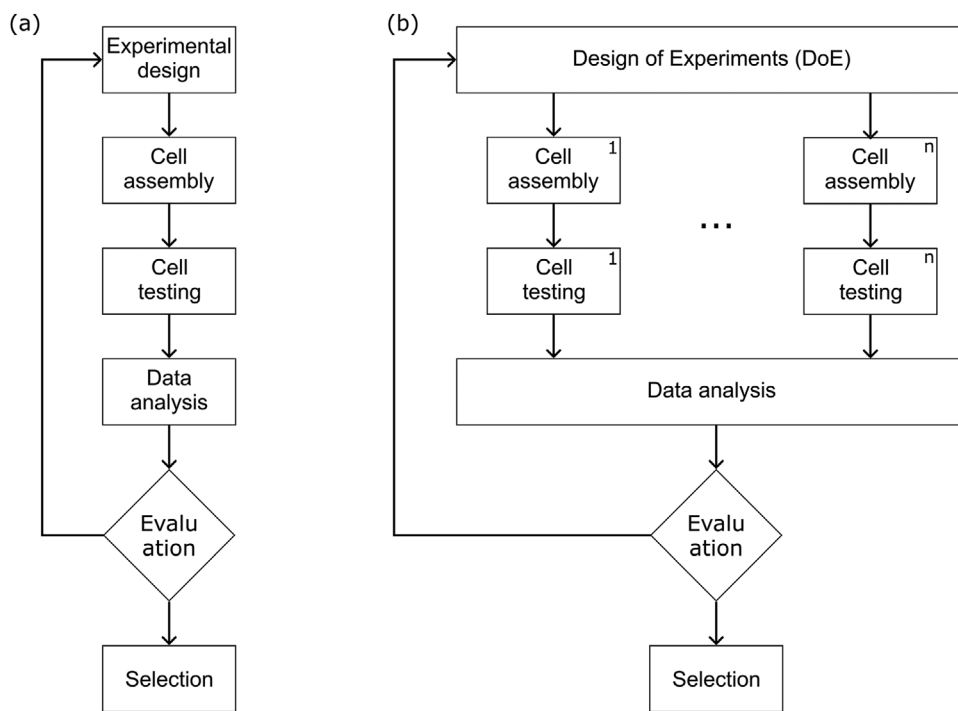
Titanium tends to form a characteristic passivation layer on the surface. Passivation layers increase the required onset overvoltage of the electrons at the phase transitions [53–55]. The deviating results for cell 1 and cell 4 are therefore potentially related to the passivation layers on the surface of the CCs. Two approaches could address this issue. First, the CCs can be manufactured from other materials (e.g. nickel alloy or stainless steel) that have a lower tendency to build a passivation layer on the surface. Second, the CC surfaces can be coated with a thin layer of precious metal (e.g. Au on the cathode and Pt on the anode) or other materials with sufficient conductivity.

The temperature control units of the 4-cell prototype were not used during the prototype experiments. Without active control, temperature fluctuations in the laboratory or external energy inputs (e.g. from the pump, neighboring cells, or waste heat of other devices) can cause temperature deviations between the individual electrolyte cycles. This might affect the results measured with the 4-cell prototype. With commissioning the multifactorial temperature control (cf. 3.1. High-Throughput Test System), temperature fluctuation-related errors in the final HT system should be precluded.

All cells were operated under nominally identical conditions. However, other effects such as slight variations in electrode activity or differences in electrode spacing due to local overlap of electrode and gasket are still theoretically possible. Such material or experimental deviations would also influence the reproducibility of the results. The results presented highlight the challenge of ensuring high reproducibility under nominally identical conditions. The reproducibility of the data can be further improved with the measures discussed.

## 4 | Design of Experiments

The previous sections have discussed, how the four key principles, i.e., parallelization, automation, modularization, and standardization (cf. 3 System Design Approach), were implemented in



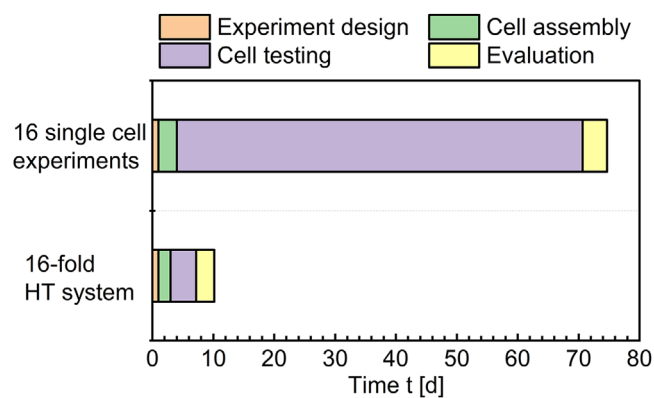
**FIGURE 7** | Workflow for a one-dimensional sequential single-step experimentation approach (a) and a Design of Experiments (DoE) workflow implemented in a high-throughput (HT) system with  $n$  parallelized cells (b).

the HT environment. In order to exploit the full potential of HT experimentation, both experimental design and data analysis must be automated and streamlined to close the circle of a self-consistent, highly accelerated screening workflow (Figure 7). The full automation, process control, online analysis, and feedback loops of the HT setup (cf. 3.1. High-Throughput Test System) improve the availability, reproducibility, and cost-effectiveness of electrolyzer data considerably. On the other hand, the parameter space of the degrees of freedom, which determine the performance of the electrolyzer, is high-dimensional, complex, and very often correlated. This parameter space comprises process parameters (temperature, pressure, electrolyte properties, flow rate, etc.), electrochemical parameters (current, voltage, electrode potential, etc.), and cell component parameters (electrodes, membranes, porous transport layers, etc.). Even with the introduced HT system, the feasible amount of generated data is still too small for systematic screening of the parameter space and orders of magnitude below the required big data for machine learning approaches for data analysis and system modelling. However, this is where the adaptive, statistic DoE approach holds a lot of potential.

DoE reduces experimental data with statistical methods, extracts the maximum amount of information from the data, and determines the optimal set of parameters for new experiments to be conducted. A DoE approach requires three things: First, a defined research goal, second, one or multiple descriptors, and third, a defined parameter space. Based on the research goal, two different areas of application for DoE are defined. The objective is either to optimize the experiment or to gain maximum information about the process throughout the parameter space corresponding to the exploration-exploitation dilemma in decision-making with minimal experimental effort. The latter case strives to elucidate the mechanisms of the experiment and

to describe them with mathematical models. In both cases, one or multiple descriptors, which describe the performance of the experiment in a robust, concise way, are required to quantify either the optimality or characteristic features of the process [56–58].

In the context of our electrochemical HT approach, iterative DoE strategies are the method of choice. Based on data collected from experiments in previous iterations of the DoE approach, DoE determines the most promising experiment (or set of experiments) to be conducted next in order to reach the research goal, such as optimizing important KPIs (cf. Figure 1), in the most efficient way. By exploiting previous knowledge and making an informed decision based on statistical probabilities, adaptive DoE strategies surpass systematic, sequential as well as random sampling and scale orders of magnitude better with the dimensionality of the parameter space [59–61]. More recently, the coupling of DoE with large language models (LLM), such as GPT-4, has been demonstrated. Simultaneously to active learning from the experiments conducted, the DoE is enabled to exploit the vast resources of both interdisciplinary and domain knowledge by LLMs. While additional care has to be taken with respect to the risks of hallucinations by LLMs and the limited comparability of previous literature knowledge with the current, innovative cell design, LLMs promise a significant step towards fully autonomous self-driving labs for electrochemical high-throughput screening [62–64]. The combination of DoE strategies with the developed HT system will amplify their individual potential. Together they have the potential to accelerate the research and development process of new technologies while minimizing the required resources. DoE will be an indispensable tool of the HT system and guarantee its optimal use for maximum gain and output.



**FIGURE 8** | Comparison of workflow timeframes for 16 iterations of a 100 h experiment in a single cell setup vs. 16 cells running for 100 h in a 16-fold high-throughput (HT) system. The calculation is based on an 8-h work day. Non-working days were not considered for the calculations.

## 5 | Discussion

### 5.1 | Acceleration Potential

One of the main objectives in developing the presented HT system is the acceleration of electrolysis research activities. In a conventional single-cell setup, the individual standardized workflow steps are carried out sequentially (Figure 7a). Due to the complexity of the parameter space in electrocatalysis, the workflow typically must be iterated several times, which is exceedingly time-consuming. Hereby, the operator laboriously works his way through the parameter space and slowly gains a deeper understanding of the material or process. The advantage of a parallelized HT system combined with a DoE approach over a one-dimensional sequential workflow is clear and becomes particularly attractive when a large number of parameter combinations need to be varied (cf. 4 Design of Experiments). By implementing DoE, a HT system can fully exploit its acceleration potential compared to one-dimensional strategies. The DoE workflow for a parallelized  $n$ -fold HT system is shown in Figure 7b.

Figure 8 exemplifies the temporal comparison of two comparative studies that yield an equal amount of experimental data. It shows the total time required to carry out the entire workflow once in a 16-fold HT system (Figure 7b) compared to 16 times in a single-cell setup (Figure 7a), based on a duration of 100 h per experiment. The estimated time required for the experiment design, cell assembly, and data analysis is based on practical experience. For the estimated time frames, it was considered that experiment design, cell assembly, and data analysis could only be carried out within working hours. The experiments (cell testing) can run 24 h a day without the presence of the operator. Experiment design takes the same time for both workflows, regardless of the system. During cell assembly and data evaluation, synergies can already lead to little time savings when using the HT system. The most significant time savings are clearly achieved by carrying out the cell tests in parallel. The example calculation for a test period of 100 h results in a total time saving of approximately 86%. The demonstrated comparison shows the great acceleration potential through considerable time savings when operating a

16-fold HT system. However, studies in a 16-fold system require careful experiment planning to make optimal use of the system's capacity and thus exploit its full acceleration potential (cf. 4 Design of Experiments).

### 5.2 | Variability Potential

High variability is one of the key characteristics of the introduced HT system, which distinguishes it from previous, mostly application-specific HT platforms. The system is intended to enable research into a wide range of research applications and the operation of various electrocatalytic processes.

#### 5.2.1 | Application Variability

The most important applications are component screening as well as process and electrochemical parameter screening. In the case of component screening, different variants of a component are assembled into the flow cells. The modular design of the flow cell enables the assembly of all components commonly used in electrochemical research (cf. 3.2 Modular Electrochemical Flow Cell). Different variants of a component can be tested simultaneously under identical, precisely adjustable process and electrical parameters. The high level of comparability and reproducibility of the system ensures that nominally equal test parameters result in identical test conditions (cf. 3.3 4-Cell Prototype System). Therefore, reliable and high-quality data can be generated, allowing any performance disparities observed during screening procedures to be directly attributed to the components themselves.

In contrast, parameter screening involves testing identical components in all flow cells. Due to the high degree of automation and the sophisticated sensor and controller equipment of the HT system, most of the process and electrical variables of the complex parameter space can be controlled automatically. Therefore, the parameter space can be efficiently screened for the predefined set of components. The aim of parameter screening can be either the exploration of the parameter space or the discovery of optimum process parameters (cf. 4 Design of Experiments).

The HT system can be used in many other applications. Maintenance and replacement of degraded system components are very costly. To minimize these costs, system lifetime is one important KPI for industrial applications (cf. Figure 1). Accelerated stress testing is a suitable method to efficiently and quickly induce the aging of system components, thus enabling lifetime investigations, e.g., in the development of new materials at an early stage. The high degree of system automation provides ideal conditions for well-defined accelerated stress testing procedures.

Currently, electrolyzers can still be operated at a constant load due to the availability of fossil fuels. The constant availability of the energy carrier will end with the expansion and transition to renewable energies. Future electrolysis systems must therefore be able to operate well under dynamic loads. The behavior under transient operation is another important KPI (cf. Figure 1) that must be considered during process development. A transient operation can be easily investigated with the developed HT system due to the sophisticated potentiostat setup.

Standard operation of the HT system involves the use of 16 single flow cells. The modular subdivision into four reactor racks allows the electrical coupling of the cells within one rack. With minor modifications to the cells, the CCs of neighboring cells can be electrically interconnected via bipolar plates. Thereby, up to four single cells could be transformed into one bipolar short stack. This would provide the unique opportunity to test parallelized bipolar short stacks in a highly automated HT environment.

### 5.2.2 | Process Variability

One further important objective was to cover a wide range of different processes. Primarily, the system will be run with water and CO<sub>2</sub> electrolysis (cf. 3.1 High-Throughput Test System and 3.2 Modular Electrochemical Flow Cell). For water electrolysis, our focus is mainly on AWE and AEM water electrolysis. For CO<sub>2</sub> electrolysis, we are focusing on the processes CO<sub>2</sub>-to-CO, to-formic acid and to-ethanol. Beyond this, the system can be used to investigate other processes such as proton exchange membrane water electrolysis or organic electrochemistry. With minor system modifications, it is technically possible to switch to other electrochemical systems such as redox flow batteries. However, any practicable process change must be accompanied by thorough cleaning procedures in order to avoid contaminations from previous processes. The high process variability is facilitated by various aspects.

Different electrolysis processes require various cell concepts. The modular design of the flow cell enables its adaptability to all common cell concepts and, thus, ensures a high degree of process variability. Process variability also requires the HT system to be operated with different electrolytes, including solutions with high chemical corrosion activity. To avoid undesired interference (cf. 3.1 High-Throughput Test System), all system components in contact with the electrolyte during operation are made of chemically inert polymers. All commonly investigated electrolyte solutions can therefore be used in the system.

For various electrolysis processes, toxic (e.g. CO) or flammable gases (e.g. H<sub>2</sub>) are required as feedstocks or are generated as products. Appropriate gas management to prevent the formation of dangerous concentrations of toxic or flammable gases is therefore important. All educt and product gases are released directly to the exhaust (no recirculation). Furthermore, the HT system is installed in an enclosure, in which the air is continuously circulated and vented to the exhaust. Therefore, no gas leaks into the laboratory. In addition, sensors for the most important hazardous gases are located inside the enclosure to detect dangerous gas concentrations. In the case of a gas leakage in the system, the detection signal triggers appropriate safety measures depending on the hazard level.

## 6 | Conclusion

The development of current or new electrolysis processes is a complex challenge. Research and development efforts must be carried out at all system levels, from material testing to stack optimization. At the same time, the performance of electrolysis

technologies needs to be optimized in multiple dimensions, considering various relevant key performance indicators. High-throughput (HT) test environments have the potential to be the ideal tool to efficiently tackle such complex multi-level and multi-dimensional research challenges and to significantly shorten technology development times.

Over the past four years, an innovative HT test system and HT electrochemical flow cell for electrolysis research have been developed in collaboration between IET-1 and hte GmbH (Heidelberg, Germany) [44]. The parallelization of 16 electrolyzers will significantly accelerate testing compared to conventional single-cell test setups. Its high degree of automation and development in line with commercial standards improves the reproducibility and comparability of the data generated. Furthermore, the modular design supports quick modifications to the system and the flow cell enabling the operation of various processes and research from material to short stack level. To exploit the full HT potential, the entire equipment - from upstream and downstream to analytics and the electrical setup - has been designed to optimize the performance of electrolysis technologies regarding the most important key performance indicators.

Experimental prototyping included single-cell testing and a reproducibility study performed in a 4-fold HT prototype system. The results demonstrate that the developed cell enables the investigation of many applications relevant to electrochemical research and the operation of different processes through modular modifications. In a 4-fold prototype system, the data from four parallel-operated cells under nominally identical conditions were obtained and compared with each other. Overall, good reproducibility was demonstrated. It is hypothesized that the reproducibility can be further improved by optimizing the current collector surfaces and operation with active temperature control.

The full automation, process control, online analysis, and automated feedback loops of the HT system provide perfect conditions for Design of Experiment (DoE) strategies. By exploiting prior knowledge and making an informed decision based on statistical probabilities, adaptive DoE strategies surpass conventional testing approaches and scale orders of magnitude better with the dimensionality of the complex parameter space in electrochemistry. DoE will therefore be an indispensable tool for the HT system. The implementation of DoE strategies will further amplify the acceleration potential, minimize the required resources, and guarantee the optimal use of the HT system for maximum gain and output.

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### Conflicts of Interest

The authors declare no conflicts of interest.

## Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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