

MATHEMATICAL MODEL OF A FILTER FOR CO₂ REMOVAL OF CONTINUOUS GAS SUPPLY FOR AN ALKALINE MEMBRANE FUEL CELL

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ARTICLE INFO

Keywords: fuel cell, CO₂, AMFC.

Received: Sept 01, 2024

Reviewed: Sept 03, 2024

Accepted: Sept 20, 2024

ABSTRACT

The pursuit of new technologies capable of generating clean and sustainable energy has been growing over the years, not only for high-power generation systems but also to support the change from the world's dependence on internal combustion engines powered by fossil fuels. The Paris Agreement and the United Nations Sustainable Development Goal 2030 are examples of the international interested in switching their energy matrices to sustainable sources and reducing the use of fossil fuels. Fuel cells (FCs) have emerged as potential replacements for internal combustion engines and fossil fuel-powered generators. Due to their economic viability to continuously and safely generate electricity, FC's are used in stationary energy generation systems and electric vehicles. Fuel cells are devices that convert chemical energy into electricity through a spontaneous redox reaction between two gases, hydrogen, and oxygen. The products of that reaction are the electric energy and water. There are two types of low-temperature fuel cells mainly used, the Proton-Exchange Membrane Fuel Cell (PEMFC) and the Alkaline Membrane Fuel Cell (AMFC). They differ in the chemical reactions that take place in the electrodes and in the membrane, that can be either acid or alkaline. The membrane allows the ions (cations or anions) from one side of the FC to reach the other side completing the overall chemical reaction. PEMFC is most commonly used due to its versatility, as it can be applied in stationary and mobile systems. However, the AMFC has superior efficiency and similarity in membrane manufacturing, although its use is restricted when CO₂ is present in the air supply. When the CO₂ coming in with the air supply reacts with the KOH present in the membrane, it forms potassium carbonates (K₂CO₃) in the membrane, which impairs the efficiency of the AMFC. The present study proposes a mathematical model of the CO₂ capture through a filter composed of a container with KOH solution placed between the air supply and the fuel cell. The goal is to quantify the capture of carbon dioxide the efficiency of the filter through mass balance, inhibiting CO₂ and allowing only the passage of air without the presence of substances that contaminate the alkaline membrane. We also consider the need of filter replacement or replenishment, ensuring a constant pure air flow to the AMFC. The control of CO₂ passage to the alkaline membrane would provide increased reliability in the use of fuel cell technology and result in higher efficiency.

1. INTRODUCTION

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With the increasing global population that has been extending over the years, it is estimated that the population will reach 9 billion people by 2037, representing an increase of up to 56% in the planet's energy consumption. This will demand even more energy and a higher utilization of the planet's natural resources to meet this substantial energy demand (UN, 2022). Energy consumption and CO_2 emissions will also rise, potentially leading to intensified climate changes and increased greenhouse effect, primarily due to the majority of countries dependence on fossil fuels and non-renewable energy sources (Masnadi et al., 2015).

The growing awareness of climate change and the recurring oil crises have sparked increased research and investments in renewable energy sources and clean energy technologies, aiming to mitigate the greenhouse gas effect and achieve the target of limiting global warming to the 1.5 °C scenario by 2100 (Wang et al., 2011).

Fuel cells (FC) have emerged as a promising technology that can significantly contribute to the transformation of energy conversion approaches on a global scale. However, certain technological challenges continue to hinder their widespread adoption, notably the reliance on expensive and rare catalysts (Pan et al., 2017). The use of hydrogen fuel cells is attractive for stationary stacks and some mobile applications like automotive vehicles because their high energy conversion efficiency of up to 60% and their lack of CO_2 emissions (Negro et al., 2018).

Despite their higher cost, fuel cells have found application in various domains, including vehicles, stationary power plants, trains, and hybrid systems, primarily due to their high energy conversion efficiency, straightforward operation, and environmentally friendly characteristics, such as low noise and absence of harmful emissions (Benipal et al., 2018).

Among the early developments in fuel cell technologies using hydrogen for electricity generation, the alkaline fuel cell (AFC) stands out. AFCs employ a potassium hydroxide (KOH) solution as the liquid electrolyte, facilitating the movement of ions (Gülzow and Schulze, 2014). However, the presence of CO_2 in the system leads to undesirable reactions forming potassium carbonate, resulting in the "poisoning" of the cell (Merle; Wessling and Nijmeijer, 2011). To overcome this issue, efforts have been directed towards incorporating anionic exchange membranes (AEM) to effectively replace the liquid electrolyte in AFCs.

2. EXPERIMENTAL SETUP

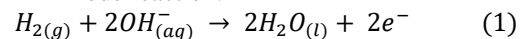
Alkaline Membrane Fuel Cells (AMFCs) have shown to deliver high power densities and operational stability. They operate at lower temperatures and exhibit relatively higher efficiency compared to other types of fuel cells, making them a worthy competitor

to Proton Exchange Membrane Fuel Cells (PEMFCs) (Gottesfeld et al., 2018). Additionally, AMFCs have the potential to achieve considerable lifespan, further solidifying their position as a formidable rival to PEMFCs.

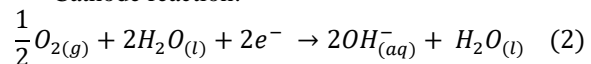
For a Fuel Cell (FC) to produce electrical energy, it must be continuously supplied with a fuel and an oxidant. This feeding of reactants is particularly crucial when the current intensity is high, as the rate of reactant consumption becomes rapid and can become a limiting factor for the overall reaction. For this reason, controlling the oxygen input is necessary. Additionally, they are well-known for their efficient oxygen reduction reaction (ORR) electrochemical kinetics, as well as their ability to minimize corrosion due to the alkaline environment they operate in. Another advantage of AMFC is their utilization of various electro-catalyst materials beyond noble metals (Hamada et al., 2023).

The overall reactions that illustrate the functioning of an AMFC are given by:

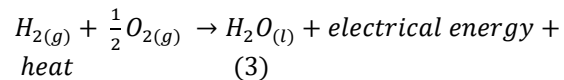
Anode reaction:



Cathode reaction:



Overall Cell reaction:

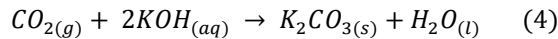


The performance of the AMFC is influenced by various parameters, including temperature, pressure, and relative humidity of both the membrane and gas. The membrane plays a crucial role in the AMFC's operation, allowing the exchange of ions while preventing the transfer of hydrogen from the anode to the cathode. Besides possessing good ionic conductivity, the membrane also offers some mechanical stability (Fan et al., 2021).

An important advantage of the AMFC is that it utilizes oxygen gas, which allows the use of atmospheric air as a feed source, making it economically viable and favorable for application in mobile systems. This characteristic enhances its practicality and attractiveness for various portable and mobile energy applications (Bidault et al., 2011).

2.1 Electrolyte poisoning

KOH stands out as a favorable electrolyte for implementation in AMFCs due to its exceptional ionic conductivity. However, it exhibits a high reactivity with CO_2 present in the surrounding air, leading to the formation of carbonates through a chemical reaction with hydroxide ions (Schulze and Gülzow, 2004). This reaction can be represented as follows:



The sensitivity of KOH solution to CO_2 necessitates careful handling and measures to mitigate its impact on the AFC's performance (Gouérec, et al., 2004).

The primary issue arising from the formation of carbonates is the detrimental impact on the FC's performance. When large metal carbonate crystals, such as K_2CO_3 , precipitate, they alter the electrolyte's composition and significantly reduce its ionic conductivity. This diminishes the efficiency and effectiveness of the fuel cell.

Furthermore, carbonate precipitation can lead to blockages in the gas diffusion layer, impeding the proper flow of reactants and products within the cell. This obstruction further hampers the fuel cell's functionality and can result in decreased overall performance (Laminier and Dicks, 2002). These challenges highlight the importance of managing carbonate formation in AMFCs to ensure their long-term stability and optimal performance.

The adoption of membranes instead of liquid electrolytes has proven beneficial in mitigating the detrimental effects caused by CO_2 contamination in fuel cells (Liang, et al., 2022). By using anion exchange membranes, the formation of precipitated carbonates and large crystals that could block the gas diffusion electrodes is prevented. However, this shift to membranes does come with the trade-off of limiting the use of KOH, despite its superior ionic conductivity for fuel cell performance.

Apart from mitigating CO_2 -related issues, anion exchange membranes offer other advantages such as volumetric stability and ease of handling. Their use also results in reduced size and total weight of the fuel cell, expanding the range of potential applications. Additionally, this technology improves overall efficiency and slows down performance degradation over time. It is worth noting that achieving these benefits might require more rigorous processing and treatment of the membrane material (Ziv et al., 2018).

Despite the challenges, researchers have dedicated numerous studies to enhance the durability and competitiveness of anion exchange membranes, aiming to achieve comparable performance to PEMFCs. Through continued research efforts, the ultimate life of these fuel cells is being extended, positioning them as a compelling alternative in the fuel cell landscape (Arges and Zhang, 2018).

3. METHODS

As mentioned earlier, Alkaline Membrane Fuel Cells (AMFCs) typically utilize ambient air as the oxidant for the electrochemical reaction that generates electricity, which avoids the need for high-purity O_2 and enhances practicality and economic viability.

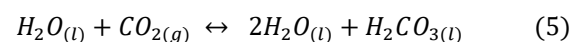
The formation of carbonates in AMFCs can be a concern, but it has been reported that this carbonation reaction can be managed to a tolerable extent without significantly compromising the fuel cell's efficiency and durability. To achieve this, it is essential to reduce the CO_2 concentration in the system to levels of 5 parts per million (ppm) or lower (Gottesfeld et al., 2018). By minimizing the presence of CO_2 to such low scales, the negative impact on the fuel cell's performance can be effectively mitigated, allowing for the continued operation and longevity of the AMFC.

Significantly reducing the carbon dioxide concentration in the ambient airflow before supplying it to the cathode of air-fed AMFCs is of utmost importance. Recent research efforts, such as those conducted by Raimundo et al. (2020) and Liang et al. (2022), have been focused on exploring practical applications that involve chemistry reactions and membrane systems to mitigate CO_2 contamination.

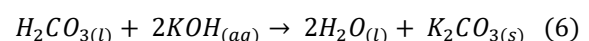
To contribute to the ongoing research on containing carbon dioxide to prevent its reaction with the alkaline membrane, this study aims to model and simulate a purifier that filters carbon dioxide as it passes through a KOH solution along with the air. Building upon the previous work by Sapucaia et al. (2022), it is already established that the filter has the capacity to capture up to 95% of the CO_2 present in the air. However, over time, the concentrations of H_2O and KOH decrease due to the conversion of carbon dioxide into potassium carbonate, necessitating the development of an efficient control system.

This control system must be capable of replenishing the filter with high rates of CO_2 capture while maintaining a low flow of KOH to replenish the system. The successful implementation of such a system will ensure the sustained effectiveness of the purifier and facilitate its practical application in air-fed AMFCs, addressing the challenges associated with CO_2 contamination and preserving the performance and longevity of the fuel cells.

The formation of carbonate occurs through a two-step process. Initially, carbon dioxide reacts with H_2O to form the intermediate and unstable acid, carbonic acid (H_2CO_3), as shown in Eq. (5) (Pocker and Bjorkquist, 1977). It is important to note that this reaction is reversible, meaning carbonic acid can further decompose into CO_2 and H_2O under certain conditions.



The second step involves the reaction of carbonic acid (H_2CO_3) with potassium hydroxide (KOH), resulting in the formation of carbonate (K_2CO_3). This carbonate compound has the tendency to precipitate within the liquid solution, as depicted in Eq. (6).



The choice of using reaction (7) in this study is well-suited to achieve the research objective, primarily due to its irreversible nature. As a result, the produced carbonate (K_2CO_3) will remain confined within the KOH solution and will not be transported to the membrane alongside the air. For the purpose of this work, the chemical species K_2CO_3 resulting from reaction (7) will not be incorporated into the calculations. The focus of the analysis centers on the mass transfer that occurs during the entry and exit of the purifier. In accordance with the study's objectives, it is assumed that the potassium carbonate remains in the solution without influencing the modeling and simulation of the purifier.

The capture of CO_2 through the use of a KOH filter results in the formation of potassium carbonates, which is a substance with low environmental impact, non-toxic and cheap, as has been advocated by Gohla (2021) for applications in extracting microplastics in environments. K_2CO_3 have been studied for domestic heat storage applications on in heat batteries (Sögütöglu et al., 2018) and an alternative to carbon capture tool in emission areas (Chai, Ngu and How, 2022).

4. MATHEMATICAL MODELING

Using Henry's Law to describe the reaction presented in the Eq. (5), we obtain:

$$\frac{d[CO_2]}{dt} = \frac{k_L A_1 ([CO_2] - [CO_2^*])}{V_L} - \frac{k_L A_2 [CO_2^*]}{V_L} - k_1 [CO_2^*] + k_2 [H_2CO_3] + Q_L (-[CO_2^*]) \quad (7)$$

Where:

$[CO_2]$ is the inlet carbon dioxide concentration, mol/mL;

$[CO_2^*]$ is the carbon dioxide concentration in the alkaline solution, mol/mL;

$[H_2CO_3]$ is the carbonic acid concentration, mol/mL;

Q_L is the volume flow, mL/s;

k_L is the mass transfer coefficient by convection, cm/s;

k_1 is the carbon dioxide kinetic rate, s^{-1} ;

k_2 is the carbon acid kinetic rate, s^{-1} ;

A_1 and A_2 are the inlet and outlet area, respectively, in square centimeters (cm^2);

V_L is total volume of the reactants, mL.

The Henry's law coefficient and partial pressure were utilized to determine the carbon dioxide concentration in the air that enters the filter, as described in the study by Seader et al. (2016).

$$[CO_2] = H_e P_{CO_2} \quad (8)$$

where, H_e is the Henry's law coefficient, mol/mL.atm and P_{CO_2} is the carbon dioxide partial pressure, atm.

Eq. (9) represents the reaction of CO_2 into H_2CO_3 in the alkaline solution.

$$\frac{d[H_2CO_3]}{dt} = k_1 [CO_2^*] - k_2 [H_2CO_3] - k_3 [H_2CO_3][KOH] + Q_L (-[H_2CO_3]) \quad (9)$$

Where k_3 is the potassium hydroxide kinetic rate, s^{-1} ; $[KOH]$ is the solution concentration of potassium hydroxide, mol/mL.

Eq. (10) represents the reaction of KOH with H_2CO_3 , shown in the Eq. (6), generating potassium carbonate (K_2CO_3). Potassium carbonate is not taken into account in the Eq. (10) because it is a final product of the reaction and does not generate any other product, therefore, the Eq. (6) it is an irreversible reaction.

$$\frac{d[KOH]}{dt} = -2k_3 [H_2CO_3][KOH] + Q_L ([KOH]^o - [KOH]) \quad (10)$$

The filter efficiency was defined as follows:

$$\eta = 100 - 100 \left(\frac{k_L A_2 [CO_2^*]}{k_L A_1 ([CO_2] - [CO_2^*])} \right) \quad (11)$$

The filter's efficiency is defined as the ratio of the CO_2 entering the system to the carbon dioxide exiting the system. The carbonates formed during the reaction are not taken into account due to their irreversibility and lack of presence in the filter's effluent.

To determine the concentrations $[CO_2^*]$, $[H_2CO_3]$ and $[KOH]$, a system of second and third-order differential equations is solved, consisting of equations (7), (9), and (10), considering the initial conditions and geometric parameters specified in Table 1.

Table 1. Parameters fixed to simulate equations

Symbol	Value	Reference
A_1 and A_2	1.0 cm^2	---
k_L	0.04 cm/s	Han et al., 2013
$[KOH]^o$	0.0001 mol/mL	---
H_e	0.000034 mol/mL.atm	Pandis and Seinfeld, 1989
k_1	0.04 s^{-1}	Han et al., 2013
k_2	18 s^{-1}	Stumm and Morgan, 1996
k_3	2050 s^{-1}	Stumm and Morgan, 1996
P_{CO_2}	0.00039	Pandis and Seinfeld, 1989
V_L	50 mL	---

*Values entered by the authors.

5. RESULTS AND DISCUSSION

Through the previously presented reactions (05) and (06), it was possible to mathematically model the functioning of a CO_2 capture filter with the objective of limiting the passage of carbon dioxide and implementing a KOH flow control to replenish the filter as it produces potassium carbonate.

This control mechanism proved necessary because previous calculations showed high carbon capture efficiency, but limited durability. By calculating the time and amount of KOH to be added to the liquid filter under a constant air inlet flow or while the AMFC is in operation, two main issues can be mitigated: membrane poisoning and filter maintenance during operation.

In Figure 1, the first test in the system was conducted with a control range between 90% - 95% efficiency, where the KOH refill was triggered when the efficiency dropped below 90%, maintaining a volumetric flow rate of 0.01 mol/mL. However, as observed, due to the low flow rate, the system could not sustain the 95% efficiency, indicating the need to increase the volumetric flow rate to maintain stable control.

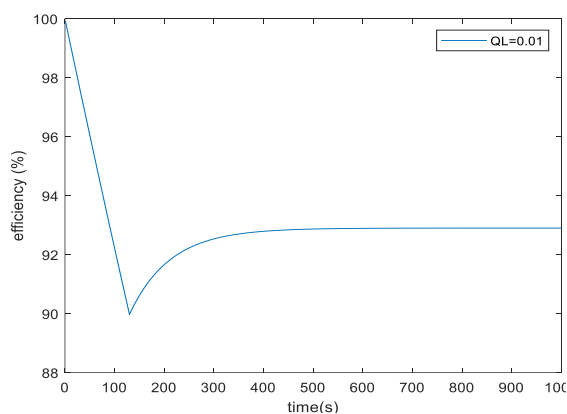


Figure 1. Control system comparing efficiency over time with a flow rate of 0.01 mol/mL.

In Figure 2, the control system is shown to be functioning effectively, with adjustments to the minimum flow rate enabling the system to regain its maximum efficiency and effectively control the capture of carbon dioxide present in the air. Mathematically, the system demonstrates good performance and consistent adjustments, ensuring the safe and reliable operation of the setup.

Figure 3 explores another point of improvement, highlighting the opportunity to increase efficiency by making parametric adjustments that allow for further enhancement of the filter's performance. This enhancement comes at the cost of a higher amount of KOH being introduced into the system, but it results in a notable increase in the filter's efficiency.

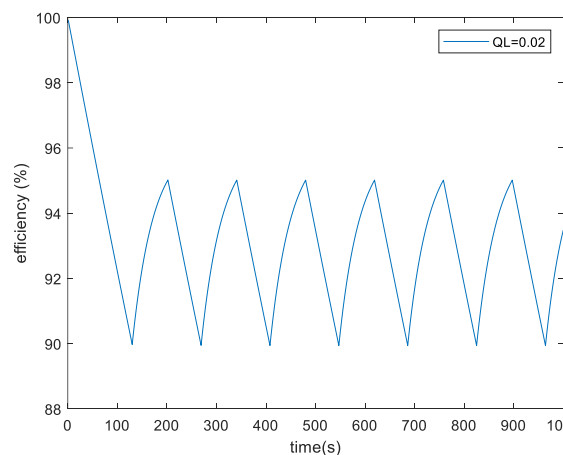


Figure 2. Control system comparing efficiency over time with a flow rate of 0.02 mol/mL.

In Figure 3, the control range was expanded to 95% - 99% with the aim of achieving maximum system efficiency while effectively regulating the passage of CO_2 to the KOH membrane and preventing its poisoning through the formation of potassium carbonate.

By setting a minimum flow rate (QL) of 0.08 mol/mL, the system was stabilized, and regular, symmetrical adjustments were achieved, resulting in an efficiency exceeding expectation based on previous studies. This successful performance opens up the possibility for highly effective applications in membrane contamination containment. Moreover, the developed control system proves to be a simple and cost-effective tool for achieving these results.

Overall, the study's outcomes demonstrate the feasibility and effectiveness of the control system in enhancing the AMFC's efficiency and preventing membrane contamination, thereby presenting a valuable contribution to the field of CO_2 capture and fuel cell operation.

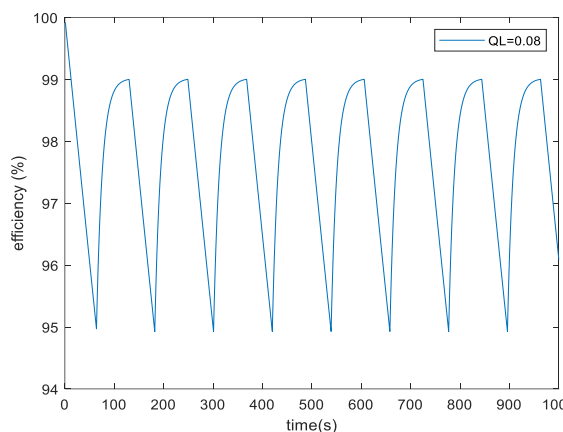


Figure 3. Stability in CO_2 containment within a high-efficiency control range.

Upon reaching an optimal control range with minimal flow during the KOH adjustments, it was also possible to analyze the consumption that would occur

in the reservoir when continuously feeding the filter. By maintaining the same concentration and varying only the KOH flow rate into the system, the goal was to identify an optimal point of low potassium hydroxide consumption and the minimum required flow rate to maintain the system's optimal CO_2 capture rate.

Figure 4 presents the graph illustrating this analysis, comparing different flow rates in control ranges that demanded high efficiency due to the intervals of KOH adjustments and the quantity of KOH entering the system. The graph does not follow a linear curve, but rather exhibits variations in the KOH consumption, making a QL of 0.08 mol/mL less efficient in solution economy compared to a higher QL of 0.1 mol/mL. This finding highlights the importance of considering both efficiency and solution economy when designing the control system for the CO_2 capture filter in the air-fed AMFC.

Based on these results, it can be stated that a 50mL filter with a flow rate of 0.01 mol/mL would achieve an AMFC with 99% efficiency in containing CO_2 during its operation. Moreover, this filter would demonstrate high efficiency with low solution consumption for its maintenance over extended periods.

These findings showcase remarkable filter performance, indicating the feasibility to proceed with experiments to validate the model and eventually apply it physically in Fuel Cells. The achieved success in efficiency and solution economy makes the filter a promising solution for CO_2 capture in AMFCs, paving the way for advancements in the development of more efficient and sustainable systems for electricity generation.

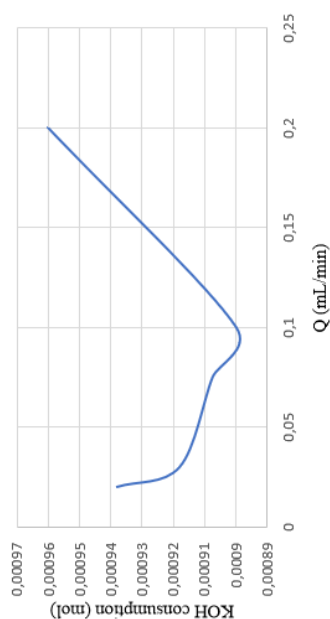


Figure 4. Comparison of KOH consumption concerning the refill flow rate in various control ranges.

6. CONCLUSIONS

In conclusion, the mathematical modeling of the CO_2 capture filter presented through reactions (05) and (06) has proved to be a crucial tool in controlling and optimizing the filter's efficiency. By implementing a KOH flow control to replenish the filter as it produces potassium carbonate, the system addresses the challenges of high carbon capture efficiency but limited durability. Through systematic calculations of the time and amount of KOH needed under different conditions, the control system effectively mitigates membrane poisoning and filter maintenance issues during AMFC operation.

The presented graphs demonstrate the feasibility of developing KOH-based filters as viable alternatives for the application of KOH-based AMFCs in mobile systems, thereby broadening their range of applications and utilization. The advantage of the alkaline membrane not requiring platinum as the main component in the electrode, opens up scope for the development of cheaper electrodes that can make fuel cells viable in systems that traditionally use non-renewable fuels, such as vehicles that use internal combustion engines and boats.

The filter that captures CO_2 to produce K_2CO_3 can encourage new research with the capacity to contain CO_2 present not only in the air, but in systems that emit higher concentrations into the environment.

Overall, the study provides valuable insights into the design and optimization of CO_2 capture filters for AMFCs, with a 50mL filter at a flow rate of 0.01 mol/mL attaining 99% efficiency in carbon dioxide containment. These findings open the pathway for conducting physical experiments and validating the model's performance in real-world applications, making the filter an attractive solution for improving the efficiency and sustainability of fuel cell technology.

7. ACKNOWLEDGEMENTS

Wellington Balmant. A great researcher, advisor and friend. Rest in peace.

To the Brazilian National Council of Scientific and Technological Development - (CNPq) - [projects 407198/2013-0, 403560/2013-6, 407204/2013-0, 430986/2016-5, 443823/2018-9, 313646/2020-1, 310708/2017-6, 308460/2020-0, 446787/2020-5, no. 25/2021, and to project 300093/2022];

CAPES; Ministry of Education (MEC), Brazil - (projects 062/14 and CAPES-PRINT-UFPR-88881.311981/2018-01);

Araucaria Foundation of Parana, Brazil (project 115/2018, no. 50.579 – PRONEX).

To FUNDEP, Renault and UFPR for the funding and the development of the project called "ROTA 2030".

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