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OPTIMIZATION OF A LOW-COST ELECTROLYTIC CELL FOR HYDROGEN PRODUCTION TO BE USED IN AN INTERNAL COMBUSTION ENGINE

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ABSTRACT

Hydrogen has important physicochemical and combustion properties that can be used to improve performance and reduce pollutant emissions by motor vehicles. However, storage difficulties can limit its use in internal combustion engines. So, in this research, hydrogen gas was produced and consumed immediately in order to eliminate the need for a storage device. First, a cell for alkaline water electrolysis was built to produce hydrogen and use it as a complementary fuel in an internal combustion engine. Aiming to investigate the energy efficiency and the capacity of this cell, three different electrolytes (LiOH, NaOH and KOH) were analyzed. The experimental tests occurred at concentrations of 1 to 10 mol/L of each electrolyte. The highest hydrogen flow rate was obtained using 7 mol/L of potassium hydroxide as electrolyte and a current of 23.5 A. Hydrogen produced under these conditions was introduced into the motor at three distinct rotation speeds (1573, 2692 and 3879 rpm). According to the results, the emissions of unburned hydrocarbons and carbon monoxide decreases with the addition of hydrogen. It can be concluded that the use of hydrogen as a supplementary fuel showed satisfactory results in reducing pollutant emissions.

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INTRODUCTION

Concerns about energy consumption and pollutant emissions has been increasing in recent years (Ji and Wang, 2009). In this aspect, hydrogen has shown to be one of the most promising alternative fuels for vehicle engines (Ji and Wang, 2009; Karogoz *et al*., 2015 and Kahraman *et al*., 2007). Hydrogen is a clean fuel, whose combustion does not produce emissions of carbon compounds such as hydrocarbons, carbon monoxide and carbon dioxide. In addition, hydrogen has excellent physicochemical and combustion properties, which can beused to promote improvements in engine performance (Karogoz *et al*., 2015; Wang *et al*., 2011; Wang *et al*., 2011; White *et al*., 2006; Wang, *et al*., 2016 and Verhelst & Wallner, 2009). Hydrogen can be used in vehicles through the employment of fuel cells or through its feed in internal combustion engines. Although fuel cells have many advantages, their high cost and short life span have become the concerns of their wide application (Ji *et al*., 2009 and Shudo *et al*., 2007). The internal combustion engines, on the other hand, are based on a well-known technology, so that the use of hydrogen in these engines can occur more simple and quicker.

In addition, the current costs of adding hydrogen to internal combustion engines are considerably lower than the costs of acquiring fuel cells (White *et al*., 2006; Winter, 2009 and Sopena *et al*., 2010). Hydrogen has a very low density, so it must be stored in tanks with high pressures (Karogoz, 2015 and Bar *et al*., 2010). Intending to eliminate this problem, hydrogen can be used as a supplementary fuel. The advantage of hydrogen-supplemented fuel is that it requires a smaller amount of gas, which considerably reduces the problems related to hydrogen storage (Karogoz *et al*., 2015; Yousufuddin *et al*., 2008 and Olavson *et al*., 1984). A better performance of engines fueled by hydrogen as additional fuel has been reported for some years. The effect of adding hydrogen on improving the performance and emissions of a gasoline motor was investigated by Wang *et al*. (2011). The unburned hydrocarbons and carbon monoxide emissions decreased 72.43% and 32.00%, respectively, when the hydrogen flow rate increased from 0 to 4.3 L/min. Another study conducted by Wang *et al*. (2010) analyzed the performance of a motor powered by hydrogen-enriched ethanol. The fuel energy consumption rate was reduced by 20% when the volume fraction of hydrogen in the air feed was increased from 0 to 6.38%. On that same line, Ceviz *et al*. (2012)

studied the effect of adding different volumetric fractions of hydrogen (0, 2.14 and 5.28%) to a gasoline engine with constant velocity (2000 rpm). Their hydrocarbon emissions decreased 13% when 5.28% of hydrogen was added to the mixture. Concerning the obtainment of hydrogen, it can be produced by water electrolysis, which eliminates the need of a device for its storage. When electrolytic cells are used in vehicles, hydrogen can be produced only when the engine is being operated, thus being consumed simultaneously (Karogoz *et al*., 2015 and Wang *et al*., 2011). The electrolysis of water can occur by several processes, the most common being alkaline water electrolysis, proton exchange membrane (PEM) and solid oxide electrolyzer cell (SOEC) (Bhandari, *et al*., 2014 and Carmo, 2013). Among these three methods, alkaline water electrolysis has the advantage of being a simple process, since its implementation does not need equipment (or experimental unit) with moving parts (Karogoz *et al*., 2015 & Zeng and Zhang, 2010). In the literature, most studies are concentrated on investigating the effect of adding hydrogen, which is provided by high pressure tanks, on the emissions and the performance of engines. In addition, the studies that refer to the optimization of the electrolyzer for hydrogen production, in general, are not tested in engines as a supplementary fuel source (Karogoz *et al*., 2010 and Rajeshwar *et al*., 2020). In this context, the aim of this study was to build a low-cost electrolytic cell for the production of hydrogen through alkaline water electrolysis and to determine the optimal conditions for its operation, in terms of capacity and energy efficiency. Then, the hydrogen produced by this cell was used as additional fuel in a spark ignition engine in order to investigate the effect of adding hydrogen gas on engine performance and emissions.

MATERIALS AND METHODS

Construction of the electrolytic cell: The electrolytic cell built for this work is shown in Figure 1 (a).

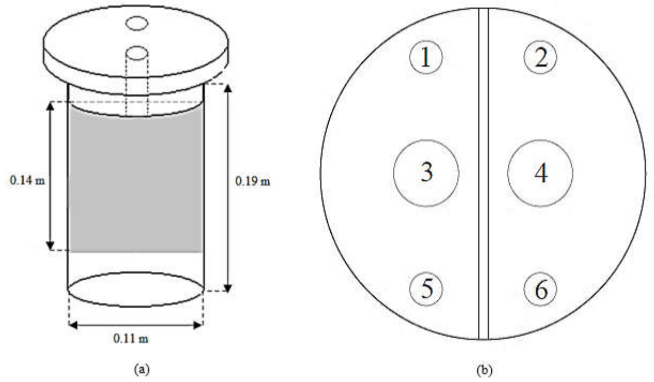

Figure 1. The electrolytic cell built: (a) Front view and (b) Upper view with: (1) H_2 gas output, (2) O_2 gas output, (3) cathode; (4) anode; (5) water inlet and (6) thermometer input

Figure 1. The cell was built in acrylicand in a cylindrical format with a volume of approximately 1805 cm^3. The upper end of the cell was designed to receive the necessary electrical connections as well as to contain the fluid passage ducts. Thus, two 340 stainless steel cylindrical electrodes, diameter of ¾ inch, were placed in the center of the electrolyticcell, through threaded fittings on the upper part of the cell. Two ducts, with internal diameter of 0.4 cm, were adjusted parallel to the two electrodes, in order to allow the output of each of the gases produced. All these components can be observed in Figure 1 (b), which shows the upper end of the cell in greater details. Hole 6, shown in this figure, was added to allow the introduction of a thermometer inside the electrolytic solution. A 14.0 cm long acrylic plate was placed in the center of the electrolytic cell, between the two stainless steel electrodes, aiming to prevent the mixing of the gases produced. However, as the electrolysis occurs, there is a need for ionic transfer between the electrodes. So, the plate had a length smaller than the height of the electrolytic cell, allowing the ionic contact in the lower part of the cell and avoiding the mixing of the oxygen and hydrogen gases.

Planning and construction of the experimental unit: With a focus on evaluating the performance of the electrolytic cell in the production of hydrogen, it was necessary to plan an experimental unit for the flow measurement of the gas produced by the cell. This unit is shown in Figure 2.

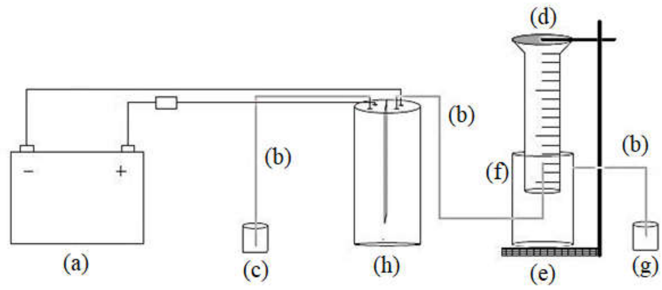

Figure 2. Experimental unit for the flow measurement of hydrogen with: (a) automotive battery; (b) silicon ducts to output of the gases; (c) beaker containing water for O_2 gas bubbling;(d) graduated cylinder; (e) retort stand; (f) beaker containing water; (g) beaker to receive the water supernatant from the graduated cylinder and (h) electrolytic cell

Figure 2. In an effort to obtain higher currents and to work in conditions similar to a vehicle, a 12 V and 45 A/h automotive battery (Fig. 2a) was used as a direct current source. The positive pole of the battery was connected to the anode of the cell, and the negative pole, in turn, was connected to the cathode of the cell. Silicon ducts (Fig. 2b) were coupled to the pipelines allowing the output of the gases produced inside the cell. The oxygen, produced in the anode, when passing through one of these ducts was bubbled in a beaker containing water (Fig. 2c), being discarded into the environment. The hydrogen produced at the cathode was collected in a 500 mL graduated cylinder (Fig. 2d), inverted and filled with water and coupled in a retort stand (Fig. 2e). The graduated cylinder was immersed in a beaker containing water (Fig. 2f). The beaker contained a spillway at the top, where the overflow of water was collected as the hydrogen gas displaced it from the graduated cylinder. Through this system, it was possible to observe the volume of water displaced in the graduated cylinder by the hydrogen in a timed time, obtaining, in this way, the volumetric flow rate of the hydrogen gas, calculated by Equation 1:

$$Q_V = V_d \Delta t$$

where Q_V is the volumetric flow rate of the hydrogen gas in m^3/s, V_d is the volume of water displaced by the hydrogen in the graduated cylinder (m^3) and Δt is the time duration of the experiment (s).

Capacity and efficiency tests of the electrolytic cell: With the planning and the construction of the experimental unit, experiments were conducted to evaluate the hydrogen production capacity of the electrolytic cell. As a mean to observe and compare the effect of different electrolytes in the behavior of the electrolytic reaction and in the production of hydrogen,tests were performed with solutions of lithium hydroxide (LiOH), sodium hydroxide (NaOH) and potassium hydroxide (KOH). The experimental tests with each electrolyte occurred at concentrations of 1 to 10 mol/L, with a variation of 1 mol/L and with the concentration of each electrolyte being evaluated in triplicate. In each experiment, the electrical currents of the process, the voltage supplied by the battery, the initial and final temperature of the electrolytic solution inside the cell and the volumetric flow rate of hydrogen were measured. With the data collected from the experiment, the efficiency of the electrolytic cell was calculated in the different concentrations of electrolytes. The efficiency was obtained through the production of hydrogen against the total electric energy applied to the system. Thus, the efficiency of the electrolysis was calculated as follows:

$$\eta_{H_2} = D_e Q_V P_E \quad (2)$$

where η_{H_2} is the efficiency of the electrolytic cell, D_e is the energy density of hydrogen in J.m^{-3} and $P_E$is the electric power consumed by the cell (W). In addition, it was necessary to evaluate the efficiency of the electrolytic cell seal, preventing the mixing of the gases produced by electrolysis. For this, gas samples collected at the cathode were analyzed on the GC-2014 gas chromatograph (Shimadzu, Japan). After the analysis of the results, the best conditions of hydrogen production by the cell were determined. The hydrogen obtained under these conditions was then added to the internal combustion engine.

Tests in the internal combustion engine: The hydrogen produced by the electrolytic cell was added to the internal combustion engine without any modification. The experimental study was performed on an alternating four-stroke (4 T) spark ignition and gasoline engine, Fiat Palio ELX. The tests were performed with the objective of observing and comparing the effects of adding hydrogen as a supplementary fuel on the emission values of the engine. For this, three engine speeds were evaluated: 1573, 2692 and 3879 rpm. To maintain the engine at the desired rotational speed for the experiment, an inertial roller dynamometer (operating range: 1000 to 6500 rpm) was used. In continuous load mode, the dynamometer used the electromagnetic brake to keep the motor in a constant rotation, specified in each experiment. Figure 3 shows the general configuration of the experiments performed on the internal combustion engine. The hydrogen produced by the electrolytic cell (a) was sent to a bubbler (b). This reservoir was used as a safety measure, since, in the absence of it, the electrolytic solution contained inside the cell could be sucked directly into the engine. Thus, the outlet duct of the hydrogen gas was connected to the bubbler and another duct was also connected to this vessel, allowing only the hydrogen gas to leave while traces of solution would remain in the reservoir.

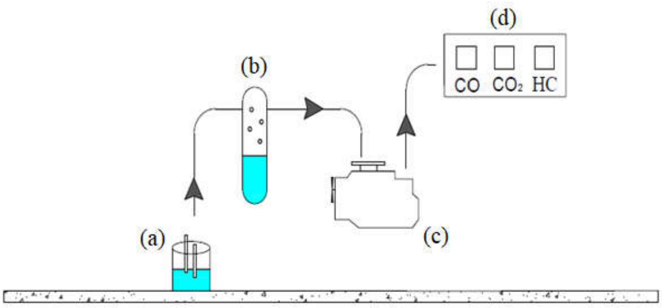

Figure 3. General configuration of the experiments performed on the internal combustion engine with: (a) electrolytic cell; (b) bubbler; (c) engine; (d) gas analyzer

Figure 3. Subsequently, the hydrogen was routed directly to the air intake manifold of the engine (c) through a silicone duct. This duct was attached to the intake manifold to prevent the hydrogen from escaping during the experiments. The emissions of unburned hydrocarbons, carbon monoxide and carbon dioxide were measured by the PC Multigas gas analyzer (d) using the non-dispersive infrared (NDIR) method.

RESULTS AND DISCUSSION

Capacity and efficiency of the electrolytic cell: To analyze the perfect functioning of the electrolytic cell in the separation of the gases produced by electrolysis, gaseous samples collected at the cathode were analyzed by means of chromatography. The results showed that the samples contained hydrogen with purity of 99.03 ± 0.37%, indicating the good sealing of the cell with respect to the mixture of the gases produced by electrolysis. The concentration and type of electrolyte are very important in the electrolysis process due to the ionic transfer in the electrolytic solution. Therefore, different alkaline electrolytes were investigated as a mean to evaluate the best working conditions of the electrolytic cell. Different concentrations of solutions of lithium hydroxide (LiOH), sodium hydroxide (NaOH) and potassium hydroxide (KOH) were tested for hydrogen production. Figure 4 shows the behavior of the electric current (*I*) as a function of the change in electrolyte concentration (*C*) for the different electrolytes. It was possible to verify by this figure that in all the concentrations analyzed, the highest currents were obtained for the solutions of potassium hydroxide. The lower electrical currents, on the other hand, were obtained using lithium hydroxide.

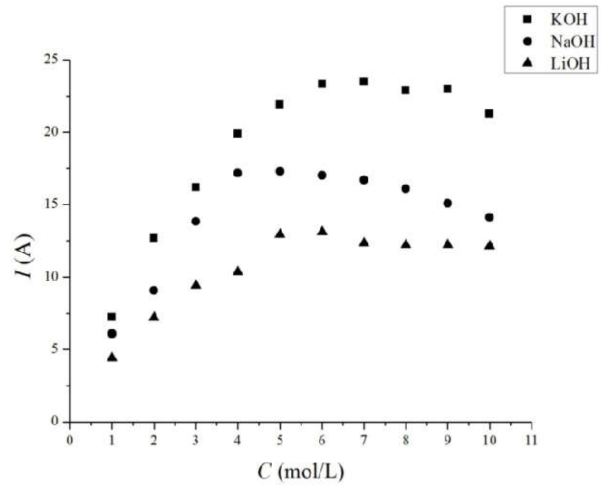

Figure 4. Electric current (*I*) as function of the concentration (*C*) of KOH (■), NaOH (●) and LiOH (▲)

Figure 5 shows the behavior of hydrogen gas flow rate produced (Q_V) as a function of the change in electrolyte concentration (*C*) for the different electrolytes. As occurred for the current, the higher hydrogen flow rates were obtained in the KOH tests. On the other hand, electrolysis with NaOH as an electrolyte produced higher amounts of hydrogen than with LiOH.

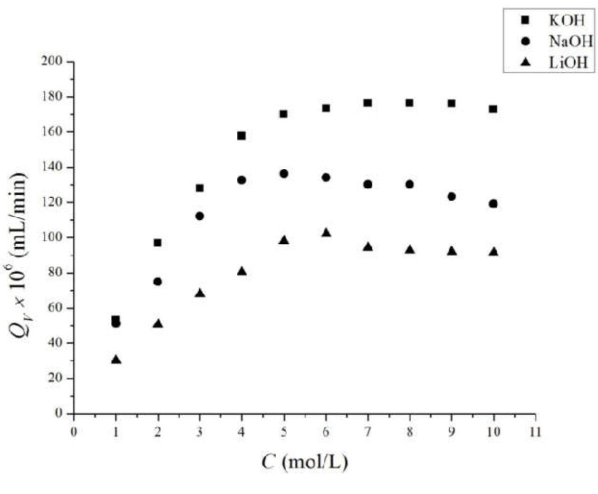

Figure 5. Hydrogen gas flow rate (Q_V) as a function of the concentration (*C*) of KOH (■), NaOH (●) and LiOH (▲).

Figure 5. One of the main justifications for the behavior shown in Figures 4 and 5 is related to the electrical conductivity presented by these compounds. Potassium hydroxide has higher electrical conductivity than sodium hydroxide and, in turn, has higher values of electrical conductivity than lithium hydroxide (Smedley, 1980 and

Wendt & Kreysa, 1999). The capacity of the electrolyte to conduct the ions present in solution during electrolysis is an extremely important factor for process efficiency (Ticianelli, 2005). This is because the increase of the ionic conductivity of the electrolyte favors the kinetics of the reaction due to the smaller activation and ohmic overpotentials (Karogoz, 2015 and Zeng & Zhang, 2010). Thus, as expected, the higher conductivity presented by KOH was determinant for the higher production of hydrogen gas. The maximum production of hydrogen with this electrolyte was 176.5 mL/min (2.94×10^{-6} m^3/s), with a continuous current of 23.5 A and a concentration of 7 mol/L. Through the analysis of these figures, it was possible to notice that the curves of electric current and hydrogen flow rate presented similar behavior. In this way, the current density was decisive for the greater obtaining of hydrogen, being in agreement with the work of Zeng and Zhang (2010). The temperature was monitored to evaluate the ohmic losses of the electrolytic process. There was an increase of approximately 6°C in the solution temperature during the experiments. It was observed that with the increase in current, the average temperature variation rose by about 1° C. The result is in agreement with the studies of Zeng and Zhang (2010), which stated that the ohmic loss increases with the current density. The average efficiency of the electrolytic cell was 10.94 ± 0.51% for LiOH solutions, 11.85 ± 0.36% for NaOH and 11.33 ± 0.36% for electrolysis with KOH as the electrolyte, asthe cell operated at current densities ranging from 0.06 to 0.30 A/cm^2.

In this work, an automotive battery was used as a source of continuous voltage, which provided about 12 V to the process, much higher than that is used in ordinary alkaline cells. In spite of that,the process has not supply sufficient hydrogen productions to promote increased efficiency. This fact can be explained by the conversion of electric energy into heat, evidenced by the temperature increase of the solution during the experiments and that is probably related to the limitations in the transport of ions existent in the electrolytic cell. Moreover, it can be noticed that there were not great variations in the efficiency of the process in the different electrolytic solutions analyzed. However, the production of hydrogen was significantly altered by modifying the electrolyte. Thus, it was decided to use potassium hydroxide solutions of 7 mol/L in the alkaline electrolysis carried out in the tests with the internal combustion engine, since the greatest hydrogen production was obtained in this type of solution.

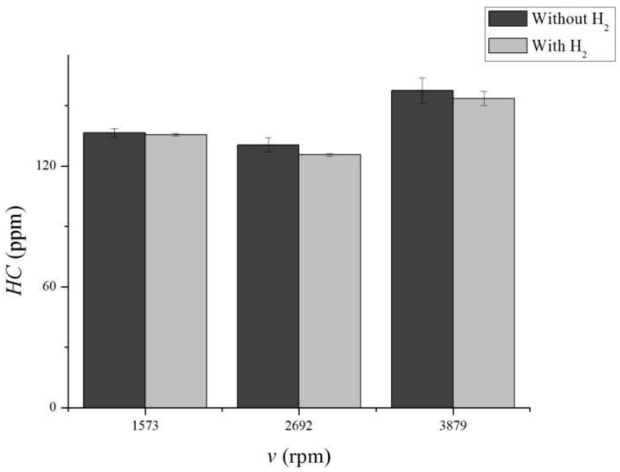

Figure 6. Variation of the emissions of unburned hydrocarbons (*HC*) in function of engine speed (*v*)

Emissions: Figure 6 shows the mean emissions of unburned hydrocarbons (*HC*), as a function of engine speed (*v*), with the respective uncertainties of the measurements.

Figure 6. Through this figure, it can be seen that the addition of hydrogen caused a decrease in the emission of these pollutants at all rotation speeds. There was an average reduction of 0.73, 3.83 and 2.54%, in relation to the operation without hydrogen, at speeds of 1573, 2692 and 3879 rpm, respectively. The quenching distance for hydrogen is lower than that for gasoline (Rajeshwar *et al*., 2008; Greenwood *et al*., 2014 and Yilmaz *et al*., 2010). Thus, the addition of hydrogen allows the air-fuel mixture to extend closer to the cylinder wall before being extinguished, thus promoting a more complete combustion, with lower hydrocarbon emissions (Ceviz *et al*., 2012). The diffusion coefficient of hydrogen is higher than those of other fuels and, for this reason, it can be claimed that hydrogen increases the homogeneity of the air-fuel mixture. A more homogeneous mixture provides a faster and more efficient combustion (Ji and Wang, 2009; Karogoz *et al*., 2015; Kahraman *et al*., 2007; Wang *et al*., 2011 and Heywood, 1988). As a result, there is an increase in engine efficiency and a lower production of soot and unburned hydrocarbons resulting from the incomplete combustion process (Zhao, 2010). The average emissions of carbon monoxide (*CO*), as a function of engine speed (*v*), in the different hydrogen flow rates analyzed, are presented in Figure 7. According to Figure 7, the addition of hydrogen reduced *CO* emissions in two of the analyzed flow rates. The decreases were 12.24% at 2692 rpm and 14.03% at 3879 rpm. Hydrogen assisted gasoline combustion emits less carbon monoxide, because the hydrogen does not include any carbon compounds in its structure. In addition, the hydrogen properties, such as high flame velocity, high diffusion coefficient and wide flammability limit increase the combustion efficiency. In the same way, hydrogen provides higher pressures and temperatures in the cylinder, which improves the oxidation reaction and consequently generates less *CO* emissions () (Almeida *et al*., 2015; Yuksel & Ceviz, 2003 and Wang *et al*., 2014). Figure 8 shows the mean values of carbon dioxide emissions (CO_2), as a function of engine speed (*v*), in the different hydrogen flow rates analyzed and with the uncertainties of the measurements.

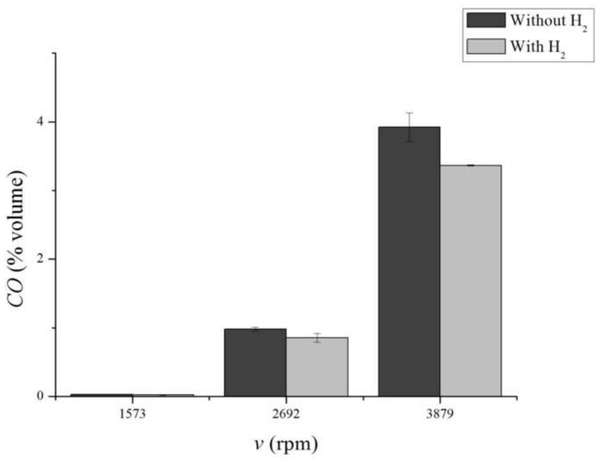

Figure 7. Variation of the emissions of carbon monoxide (*CO*) in function of engine speed (*v*)

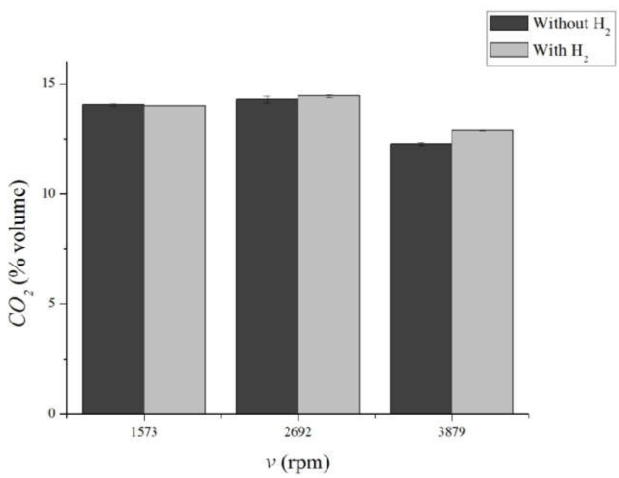

Figure 8. Variation of the emissions of carbon dioxide (CO_2) in function of engine speed (*v*)

According to Figure 8, it is possible to see that there was an increase in the emission of carbon dioxide with the addition of hydrogen. Combustion with hydrogen as a supplementary fuel contributes to the lower emission of carbon compounds, since hydrogen has no carbon atoms in its structure. However, this expected reduction in carbon dioxide emissions may have been counterbalanced by the oxidation of *CO* to CO_2due to the more complete combustion provided by the addition of hydrogen, evidenced by the reduction of the carbon monoxide, as shown previously.

CONCLUSION

With respect to the capacity in the production of hydrogen by the electrolytic cell under the operating conditions used, it can be concluded that the cell was able to promote the proper separation of the gases produced by electrolysis since hydrogen was obtained with purity of approximately 99%. Concerning the efficiency of the process, among the analyzed electrolytes, potassium hydroxide (KOH) presented the best results regarding the amount of hydrogen produced, indicating the influence of ionic conductivity on alkaline electrolysis. The maximum production with this electrolyte was 176.5 mL/min, with a current of 23.50 A. In general, the addition of hydrogen in the engine presented satisfactory results in reducing the emissions of pollutants, as it was observeddecreases in the emissions of hydrocarbons and carbon monoxide when hydrogen was injected in the test engine.

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