Modeling and simulation of graphene/palladium catalyst reformer for hydrogen generation from waste of IC engine

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Abstract. A small amount of hydrogen made by on-board reformer is added to the normal intake air and gasoline mixture in the vehicle's engine could improves overall combustion quality by allowing nearly twice as much air for a given amount of fuel introduced into the combustion chamber. This can be justified based on the calorific value of Hydrogen (H₂) 141.9 MJ/kg while the gasoline (C₆H₁₁) is 47MJ/kg. Different weight % of Pd and GO uses for the reformer model and has conducted simulation by COMSOL software. The best result found for the composition of catalyst (palladium 30% and graphene 70%). The study shows that reformer yield hydrogen 23% for the exhaust temperature of 600-900°C and 20% for 80-90°C. Pumping hydrogen may boost the fuel atomization and vaporization at engine idle condition, which could enhances the fuel combustion efficiency. Thus, this innovative technology would be able to save fuel about 12% and reduce the emission about 35%.

1. Introduction

Hydrogen is a clean fuel and contributes as a combustible fuel of the IC engine due to wide range of flammability, prompt ignition, and high flame speed at stoichiometric ratios, high diffusivity, higher calorific value, and lighter and lower density. One of the systems for on-board hydrogen production is steam reforming process. Steam reforming process is an endothermic process, which can convert unburned hydrocarbon of engine into hydrogen and carbon monoxide reaction with steam in presence of catalyst. The catalyst promotes the oxidation of the carbon to produce carbon dioxide.

Wyszynski et al. [1] stated that exhaust gas of fuel reforming with high temperature in the range of 800-900 °C produces high hydrogen yield, which is over 30% in the reformed gas. At exhaust temperature in the range of 500 – 700 °C, the hydrogen production can be obtained about 20% in the reformed gas [2,3]. The authors stated that a portion of hydrogen can be directly fed back to fuel input to increase the temperature of the 'hydrogen enriched mixture’ for better fuel efficiency.

The hydrogen can be obtained from the reforming the exhaust gas by using catalyst reformer. The exaust gas purification apparatus and method has patented [4,5]. The inventor has claimed that the composition of partial oxidation of exhaust over nickel catalyst hydrogen can be generated. The system results for partial oxidation of gasoline obtained 78.5% thermal efficiency with hydrogen and carbon monoxide yields of Vol.21.6% and Vol.23.6% respectively. In his experimental result, the reformed fuel
having hydrogen and carbon monoxide levels of 32.2% and 19.6% has observed at a reactor gas inlet with
temperature setting of 1223 K and gas hourly space velocities (GHSV) of 1000 h⁻¹.

Catalysis is the hidden gem in environmental and chemical industries. While, the use of catalysts is
most commonly known in automotive catalytic converters, they are routinely used in other
applications including gasoline, automobile lubrication and its additives and the synthesis of fine
chemicals. In addition to their economic impact, catalysts play a critical role in developing sustainable
and environmentally friendly processes. The fifty highest-produced chemicals by volume in the US correlate with a substantial carbon footprint via the emission of carbon dioxide and other harmful
gases into the earth’s atmosphere. Currently thirty of these processes have employed catalysts to reduce the emissions released and waste generated [6]. Despite catalysis emerging in the forefront of developing sustainable and efficient processes, industries still struggle to successfully develop such processes. The E-factor, one of the most straight-forward and useful green chemistry metrics, is defined as kilograms of waste produced per kilograms of product [7]. While catalysts have proven to assist in product selectivity, future advancements in process efficiency by developing immobilized catalysts [8,9,10]. Over half of all chemicals produced are still made using traditional batch reactions, yet continuous reactions offer unique advantages including intensified mixing, smaller volume to heating ratio, and safer operating conditions [11,12,13]. The highly reproducible nature and therefore the robust application of continuous processes in the production of hydrogen from the exhaust of internal combustion engine have led to an increased interest in developing the proposed technology.

Hydrogen engines have demonstrated efficiencies (BTE) in excess of today’s gasoline engines, NOₓ is the only regulated tailpipe pollutant resulting from hydrogen combustion, and carbon dioxide is a non-issue, at least in terms of the driving (Tank-to-Wheels) component of the lifecycle [14]. Although current efforts by Ford and BMW reflect early stage development, both BMW and Sandia outlined technology approaches for subsequent generations of H2ICE systems that could deliver significant improvements in fuel economy and emissions reduction, while delivering additional power (Green car Symposium).

The aim of this study to develop a green technology to generate hydrogen from the waste of IC engine which would be able to reduce the fuel consumption and reduce emission.

2. Experimental procedures
Laminar flow physics is added to the heating pipes of the reformer for the steam flow. The
wall velocity is set as zero and inlet velocity for heating tubes is set as 1 m/s. The flow properties
are defined [5] by the following equation (1):

\[
L_{entr} \bar{V}_t \ p_l + \ \bar{V}_u + (\bar{V}_u)^T \ \frac{2}{3} \ l(\bar{V}_u)^\frac{1}{2} = np_{entr}
\]  

(1)

Then the outlet flow properties are defined [5] by the following equation (2):

\[
\tau_n \ p_l + \ (\bar{V}_u + (\bar{V}_u)^T) \ \frac{2}{3} \ l(\bar{V}_u) = p_o
\]

with \( p_0 = p_{o,u,t} = 0 \)

According to Darcy’s Law, \( \bar{V}.(u)=Q_m \)

\[
u = \frac{k}{\bar{V}_p}
\]  

(2)
The mass flow rate of air, hydrogen and exhaust gas can be estimated by using the equation (3):

\[
\dot{m}_{a} = \frac{C_{d,a}A_{a}P_{a}}{\sqrt{R_{g}T_{a}}} \left( \frac{P_{a}}{P_{a}} \right)^{\frac{1}{2}} \left( \frac{2\gamma_{a}}{\gamma_{a} - 1} \left[ 1 - \left( \frac{P_{a}}{P_{a}} \right)^{\frac{\gamma_{a} - 1}{\gamma_{a}}} \right]^{\frac{1}{2}} \right)
\]

\[
\dot{m}_{h} = \frac{C_{d,h}A_{h}P_{h}}{\sqrt{R_{g}T_{h}}} \left( \frac{P_{h}}{P_{h}} \right)^{\frac{1}{2}} \left( \frac{2\gamma_{h}}{\gamma_{h} - 1} \left[ 1 - \left( \frac{P_{h}}{P_{h}} \right)^{\frac{\gamma_{h} - 1}{\gamma_{h}}} \right]^{\frac{1}{2}} \right)
\]

\[
\dot{m}_{e} = \frac{C_{d,e}A_{e}P_{e}}{\sqrt{R_{g}T_{e}}} \left( \frac{P_{e}}{P_{e}} \right)^{\frac{1}{2}} \left( \frac{2\gamma_{e}}{\gamma_{e} - 1} \left[ 1 - \left( \frac{P_{e}}{P_{e}} \right)^{\frac{\gamma_{e} - 1}{\gamma_{e}}} \right]^{\frac{1}{2}} \right)
\]

where, \( C_{d,a}, C_{d,h}, \) and \( C_{d,e} \) are coefficient of discharge for air, hydrogen and gasoline, respectively. \( A_{a}, A_{h}, \) and \( A_{e} \) are restrictions flow areas for air, hydrogen and exhaust gas, respectively. \( P_{a}, P_{h}, \) and \( P_{e} \) the air, hydrogen, and exhaust pressures, and \( \gamma_{a}, \gamma_{h}, \) and \( \gamma_{e} \) the air, hydrogen, and exhaust specific heat ratios, respectively.

The mixture of exhaust gases and steam pass to the catalyst cells and form the chemical reaction as shown in figure 1 and generate the hydrogen (H\(_{2}\)), carbon dioxide (CO\(_{2}\)) and water (H\(_{2}O\)). The hydrogen pumps to the engine cylinder, CO\(_{2}\) uses to heat the product of reformer H\(_{2}O\) and recycled to the reformer. Therefore, the rate of mass change inside this capacity can be given \([15]\) by the continuity equation (4):

\[
\frac{dm_{m}}{dt} = \dot{m}_{i} - \dot{m}_{o}
\]

with \( \dot{m}_{i} = \dot{m}_{co_{2}} + \dot{m}_{H_{2}} + \dot{m}_{NOx} + \dot{m}_{UHC} + \dot{m}_{steam(H_{2}O)} \) and

\[
\dot{m}_{o} = \dot{m}_{a} + x\% \cdot \dot{m}_{H_{2}(f)} + (1 - x\%) \cdot \dot{m}_{C(f)}
\]

where, \( \dot{m}_{m} \) is the intake manifold mass content, \( \dot{m}_{i} \) is the input mass flow to the intake manifold, and \( \dot{m}_{o} \) is the outflow from engine cylinders, \( \dot{m}_{UHC} \) is the outflow of unburn hydrocarbon, \( x\% \) is the percentage of H\(_{2}\) pumps to the engine cylinder, \( \dot{m}_{o(i)} \) is the mass rate of conventional fuel (gasoline).

The mass-balance equations (5) for the model are the Maxwell-Stefan diffusion and convection equations at steady state:

\[
\mathbf{w}_{i}^{\rho} = \mathbf{w}_{i}^{\rho} - \mathbf{D}_{ij}^{\rho} \mathbf{\nabla x}_{j} + \left( \mathbf{x}_{j}^{\rho} \mathbf{w}_{j}^{\rho} \right) \mathbf{\nabla R}_{i}^{\rho}
\]

where, \( \rho \) is the density, kg/m\(^3\), \( \omega_{i} \) the mass fraction of species \( i \), \( x_{j} \) the molar fraction of species \( j \), \( D_{ij}^{\rho} \) is the \( ij \) component of the multicomponent Fick diffusivity, m\(^2\)/s, \( D_{ij}^{T} \) the thermal diffusion coefficient, kg/(m\(^3\)·s), \( T \) is the temperature, K, and \( R_{i}^{\rho} \) the reaction rate kg/(m\(^3\)·s). The flow of heating gas in the tubes is described by the Navier-Stokes equation (6) at steady-state:

\[
\rho \mathbf{u} \mathbf{\nabla u} = \mathbf{\nabla} \left( -\mathbf{p} + \eta \left( \mathbf{\nabla u} + (\mathbf{\nabla u})^{\top} \right) \right) -
\]

\[
(2\eta/3)(\mathbf{\nabla u} \mathbf{\nabla u})^{\top}
\]
where \( \rho \) is the density \( \text{kg/m}^3 \), \( u \) the velocity, \( \text{m/s} \), viscosity, \( \text{kg/(m·s)} \), \( \dot{V} \) the flow rate, \( \text{kg/m}^3 \) and \( p \) the pressure of the mixture in the tubes, \( \text{Pa} \). At the outlet, viscous stresses are ignored and the pressure is set to 1 atmosphere.

![3D exhaust reformer](image)

**Figure 1.** 3D exhaust reformer [5].

### 2.1 Estimation Of Exhaust Gas Temperature

Rahman et al. [15] carried out the effect of engine speed (rpm) to the heat transfer in the cylinder wall and volumetric efficiency by using GT-Suite software as shown in figure 2(a) and 2(b). At 6000 rpm, there will be less time for fuel intake and combustion, which caused the temperature of the engine higher and increase in unburn hydrocarbon. Reitz (2012) found that about 30% of the fuel energy is used by transferring heat energy to the cylinder wall. Therefore, the exhaust temperature becomes higher in a range of 300-900 °C as shown in figure 2(b) and it will affect the volumetric efficiency of the engine as the heat transfer at the cylinder wall of the engine becomes less at high speed. The result of the simulation literature for exhaust gas temperature by GT-Suite is the pre-requirement for modelling of steam reformer as the exhaust temperature is crucial input in the simulation of exhaust reforming. For this project, average exhaust temperature of 900 K is used for the inlet of the steam reformer.

![Affect of exhaust gas into engine cylinder at different crank angle](image)

**Figure 2.** Affect of exhaust gas into engine cylinder at different crank angle [15]; heat transfer (a) and temperature (b).

### 2.2 Hydrogen Generation Process
The exhaust gas from the engine is passed to the turbocharger for boosting the flow rate of the exhaust gas. Figure 3 shows the hydrogen generation process from the exhaust gas and steam combination. The exhaust controller adjusts the amount of exhaust gas which distributes between the water tank and the reformer. About 5~7% of high temperature exhaust gas is considered to be utilized to heat the water of the tank and make steam which feeds into the reformer. The temperature of the hydrogen gas produced would be around 250~300 °C. In order to reduce the high temperature of hydrogen, the heat exchanger is placed immediately after the reformer to cool the hydrogen. When the desired temperature of 70~90 °C is achieved, the produced hydrogen is stored in the tank for usage in the IC engine as an alternative fuel source to save fuel consumption. The amount of hydrogen supplied to the engine about 7% of total fuel supply. The amount of hydrogen supplied to the engine is limited by the interior design of the engine. Using a crankshaft rpm sensor, the hydrogen supplied to the engine is controlled. Theoretically, the hydrogen production by the reformer increases for the engine operating rpm of 4000 rpm and above. The temperature of supplied hydrogen to the engine is expected to be 90 °C. The main reason for this temperature is to make the fuel atomized and vaporized in the engine cylinder, which helps the engine to achieve 95% fuel combustion. In the conventional engine, the amount of fuel combustion in the engine is about 80-90%. A heat exchanger is used to maintain the temperature of hydrogen at 90 °C [10]. The amount of hydrogen supplied to the engine is limited by the interior design of the engine. It is recommended that the hydrogen supply to the engine should not be more than the 7% of fuel.

Figure 3. Hydrogen generation process.

2.3 Microwave Assisted Pd/GO Synthesis
Microwave assisted synthesis of GO/Pd is proposed based on the study [12,13]. Graphite oxide (1 to 8 mg) has been dissolved in 400 ml of water until a homogenous suspension is obtained. Palladium nitrate (210 μL) has been added to the homogenous mixture and agitated using a magnetic stir plate. The homogenous mixture then placed in an Emerson conventional kitchen microwave where 100 μL of hydrazine hydrate solution (25% in H₂O) has been added. The solution then immediately microwaved on full power (1000 W) in 30-second cycles (on for 10 s, off and stirring for 20 s) for a total reaction time of 60 seconds. The resulting black solution has been centrifuged at 5000 rpm for 20 minutes and dried under vacuum until black flakes obtained. The chemical reaction that incurs into
the reformer can be presented as:

\[
\text{Exhaust}(0.5\% \text{H}_2, 5\% \text{CO, 11}\% \text{CO}_2, \text{NO}_x, 21\% \text{HC and H}_2\text{O}) + \text{Steam}(\text{H}_2\text{O})
\]

\[
Pd/\text{GO Catalyst}
\]

\[
23\% \text{H}_2 + 10\% \text{CO}_2 + 5\% \text{N}_2 + \text{H}_2\text{O}
\]

3. Results and Discussion

The reformer has been developed with two zones: mixing zone and reforming zone as shown in figure 1. At the inlet of the reformer, exhaust and steam mix together to form the mixing zone. Immediate to the mixing zone is the reforming zone where the exhaust and steam reform throughout the graphene-palladium honeycomb composite catalyst to generate hydrogen. At the outlet, water will pass downwards hydrogen will be led upwards and remaining exhaust plus carbon dioxide will go straight through the respective ducts. The length of the catalyst is 150 mm with 60 mm diameter and 5mm holes are cut through the entire length equally to make honeycomb shape. Steam, water and hydrogen ducts are 19mm in diameter and 25 mm in length.

![Figure 4. Darcy’s velocity magnitude.](image)

![Figure 5. Pressure along the catalyst bed.](image)

The flow of gases in the catalyst bed is simulated by the Darcy’s velocity magnitude in figure 4. For the flow of gases in the catalyst bed, the flow started as 1m/s as defined by the input and steadily increased along the reactor. At the reformer exit, the velocity of the gases is about 2 m/s. For the steam flow in heating tubes, the initial velocity is \(2 \times 10^{-2}\) m/s at the very beginning of the tubes and decreased to \(1.4 \times 10^{-2}\) m/s throughout the length of the heating tubes. The pressure at the inlet of the reformer is 75 Pa as defined by the input and it is found that the pressure dropped uniformly throughout the length of the catalyst bed. At 0.05m, the input pressure dropped to about 50 Pa as shown in figure 5. At 0.1m of catalyst bed, the pressure again dropped to about 30 Pa. However, the outlet pressure of the catalyst bed is close to 0.
The surface temperature of the heating tubes begins with 700 K at the inlet as defined by the input. From there it decreased slightly along the heating tubes and increased to about 740 K just before the outlet of the heating tubes as shown in figure 6(a). The surface temperature of catalyst bed at the inlet is 900 K as defined by the parameter and that of insulated foam is about 800 K. However, the temperature dropped drastically to almost 620 K at 0.05m of catalyst bed length as shown in figure 6(b). This happened because the hydrogen formation reaction absorbed heat. However, due to the heating from the heating tubes, the temperature climbs back to 660 K at the outlet of the reformer.

![Figure 6](image_url)

**Figure 6.** Pressure (a) and temperature (b) of heating tubes.
Figure 7. Mass fraction of gas species Pd 100% (a), Pd 70% Graphene 30% (b), Pd 50% Graphene 50% (c), 30% Pd and 70% Graphene (d).

At the inlet of the catalyst bed, the surface density of reacting gases 0.27 kg/m$^3$. As the chemical reaction occurred, the density increased to 0.29 kg/m$^3$ at 0.05m. After that point, the density decreased gradually throughout the reformer length until 0.19 kg/m$^3$. The mass fraction of gas species is shown in figure 7(a) – 7(f) for the different composition of palladium and graphene. By considering only Palladium as catalyst, mass fraction of hydrogen is 0.1 and that of carbon dioxide is about 0.85 at the outlet as shown in Fig.7a. Although the mass fraction of methane is 0 at the outlet which means that all methane gases are consumed in this reaction, the mass fraction of water is 0.05. Therefore, there will be some steam left in the outlet products. Considering the Pd 70% and Graphene 30% as catalyst, the mass fraction of H$_2$ increased to almost 0.13 as show in in Fig.7b. This might be the effect of graphene in the catalyst. Also in this case, all the methan are consumed throughout the length of the catalyst bed. Temperature distribution by the energy exchange between heating pipes and reformed catalyst is investigated at 900 K and 1100 K. It is studied that the steam enters the heating pipes at 900 K and exit at 632 K roughly. However, the exhaust gas enters at 900 K reaching minimum temperature first and eventually exits at average temperature of 680 K. For the 1100K, it can be seen in figure 7(f),
the mass fraction of hydrogen increases with increasing the reformer heating pipes length, while the other decreases with increasing the length of the reformer.

Figure 8 shows the conversion of each species along the entire length of reformed catalyst. As the exhaust gas travels further along the catalyst bed, the deposition of hydrogen is increased because of faster reaction rate. In the plot for mass fractions, it can be seen that the entire reactor length is active in converting methane. Hydrogen concentration increased as the species goes into the entire reactor. However, the deposited mass fraction of hydrogen is less compared to CO$_2$ and H$_2$O due to the molecular mass of each species. The amount of hydrogen production for different catalyst concentration has presented in table 1.

![Figure 8. GO/Pd catalyst based steam reformer over the conventional reformer.](image)

**Table 1.** Amount of H$_2$ produced and their energy.

| Catalyst Concentration | H$_2$ Production (g) (T=900K) | Energy (Btu) |
|------------------------|--------------------------------|-------------|
| Pd 100%                | 8                              | 908.304     |
| Pd 70%/GO30%           | 10                             | 1135.38     |
| Pd 50%/GO50%           | 11.2                           | 1271.625    |
| Pd30%/GO70%            | 14.4                           | 1634.95     |
| GO 100%                | 8.8                            | 999.13      |
Table 2. Performance comparison with reformer and without reformer.

| Fuel Type                        | Performance   |   |
|----------------------------------|---------------|---|
|                                  | Mileage (km/litre) | Energy Content (Btu) |
| (i) without reformer             |               |               |
| CNG/kg                           | 22            | 44589.6       |
| Diesel/litre                     | 20            | 34214         |
| Petrol (without reformer)/litre  | 16            | 30118.89      |
| (ii) with reformer               |               |               |
| Pd 100%                          | 20.8          | 39201.93      |
| Pd 70%/GO30%                     | 22.1          | 414726.9      |
| Pd 50%/GO50%                     | 22.7          | 42835.14      |
| Pd30%/GO70%                      | 24.7          | 46468.39      |
| GO 100%                          | 21.3          | 40110.19      |

3.1 Model validation
The proposed GO/Pd catalyst based steam reformer simulation performance has been compared with the car of compact natural gas (CNG), diesel engine and petrol engine with and without reformer in terms of mileage, and energy contents. The comparison results has been presented in table 2. By utilizing the reformer with appropriate catalyst composition, the mileage of petrol-type vehicle can be improved by 8.685 km, which is 54.28% of the normal mileage per litre of petrol. By selecting the Pd 30%/GO70% catalyst composition, the amount of hydrogen obtained is 14.4 g and the energy content is 1634.95 Btu per mol of CH₄.

3.2 Influence of Pd/GO reformer on engine performance
Typically hydrogen engines are designed to use about twice as much air as theoretically required for complete combustion. At this air/fuel ratio, the formation of NOx is reduced to near zero. Unfortunately, this also reduces the power output to about half that of a similarly sized gasoline engine. To make up for the power loss, hydrogen engines are usually larger than gasoline engines, and/or are equipped with turbochargers or superchargers. The turbocharger has been proposed for the engine with GO/Pd Catalyst based steam reformer (figure 2). This study focuses to turn the waste energy into wealth by converting exhaust into hydrogen. Without interrupting the engine size the hydrogen is pumped to the engine about 7% of total gasoline injected into the engine. It is expected that the 7% of hydrogen, which not only reduces the fuel demand it also improve the combustion of air-fuel mixture by making the fuel atomization and vaporization into the combustion chamber. It is reported that the engine performance can be enhance about 7-10% by atomizing and vaporizing the fuel (Rahman et al., 2015 and Biplah, 2013). The combustion reaction of air/fuel mixture if the hydrogen is supplied 7% of C₈H₁₈ (gasoline), the A/F (including 93% of C₈H₁₈ and 7% of H₂):

\[
0.93C_8H_{15} + 1.6H_2 + 11.75O_2 + 11.75(3.76)N_2
\]

\[
8CO_2 + 7.5H_2O + 11.75(3.76)N_2
\]

where, \( x \) is the percentage of reduction of fuel with adding \( x \) percentage of H₂. The calorific value of gasoline (C₈H₁₈) is 47 MJ/kg while hydrogen (H₂) is 141.9 MJ/kg. The 7.9 g of gasoline can be replaced with 3.2 g of hydrogen (1.6 moles of H₂). The air fuel ratio can be estimated:
0.93 x 8 moles of C  12 g/moles + 0.93 x 7.5 moles of H₂  2 g/moles = 103.23 g
1.6 moles of H₂ = 1.6 moles of hydrogen  2 g/moles = 3.2 g
Mass of fuel = 103.23 + 3.2 = 106.43 g
Mass of air = (1 + 3.76)(11.75)(29) = 1621.97 g
The air/fuel (gasoline + hydrogen) = \frac{Mass of air}{Mass of fuel} = \frac{1621.97}{106.43} = 15.24

The 1.6 mole of hydrogen pumping to the engine increases the air intake into the engine, which makes the stoichiometric A/F ratio 15.27:1 while for 100% gasoline it is 14.67:1. This indicates that the hydrogen pumping can make the engine energy efficient by saving the engine fuel pumping losses and enhance the vehicle mobility with improving emission [17].

4. Conclusions
The proposed GO/Pd catalyst steam reformer can promote the production of clean fuel hydrogen from reforming of raw exhaust gas of automobiles and steam mixture. The principle of the exhaust reforming process can be utilized the hydrogen generation from waste of engine which can be used as clean fuel into IC engines, thus improving the fuel consumption, exhaust emissions and efficiency of the engine.

- Pd/GO catalyst steam exhaust reformer be able to generate hydrogen 23-33% for the exhaust temperature in the range of 300-1100 °C.
- The temperature distribution of the catalyst bed is in the range of reasonable working temperature 680-1100 K.
- Pumping H₂ about 7% of total fuel at temperature about 80-90 °C may boosted the fuel atomization and vaporization at idle condition could enhance the fuel combustible efficiency and engine efficiency in turns to increase the engine fuel consumption and reduces emission.
- The emission of NOx could be formed into the engine due to complete combustion. The temperature of the reformer will drop due to the presence of steam and the oxidation in presence of GO catalyst the NOx will be turn into N₂ and 3O₂.
- The catalyst Pd and GO both are expensive for the reformer. However, it is very much reasonable to consider them to make the reformer efficient and compact.

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