Microwave Plasma Enhancing Mg-Based Hydrogen Storage: Thermodynamics Evaluation and Economic Analysis of Coupling SOFC for Heat and Power Generation

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Hydrogen, as a kind of green and efficient energy, plays an increasingly important role in current social development. Hydrogen storage technology is considered to be one of the main bottlenecks in limiting the large-scale application of hydrogen energy. The solid-state hydrogen storage technology based on Mg-based materials has received extensive attention due to its advantages of high hydrogen capacity, good reversibility, and low cost, but there are still shortcomings such as high reaction temperature, large energy consumption, and slow reaction kinetics. In order to solve these problems, this article proposes a new method of using microwave plasma to ionize hydrogen into H^- ion. The possible activation mechanism of microwave plasma to improve the hydrogen storage properties is put forward. Based on the activation mechanism, the thermodynamic performance of Mg-based hydrogen storage is evaluated using density functional theory. It is concluded that the reaction temperature is significantly reduced from 339°C to 109°C with the help of microwave plasma. In addition, the comparison between the conventional heating hydrogen storage process based on MgH_2 and microwave enhanced advanced hydrogen storage process based on MgH_2 systems coupled with solid oxide fuel cells for heat and power generation is conducted to evaluate the economic feasibility. The results show that the energy consumption cost of the proposed microwave plasma enhancing hydrogen storage system is approximately 1.71 \$/kgH$_2$, which is about 50% of the energy consumption cost of the conventional system.

Keywords: hydrogen storage, magnesium hydride, thermodynamics, microwave plasma, reaction temperature

INTRODUCTION

In recent years, the low-carbon process of energy has attracted national strategic attention (Hepburn et al., 2021; He et al., 2022). Hydrogen has been widely popular as a potential alternative energy source. Among the hydrogen energy full chain, hydrogen storage and transportation technology are the key points to realizing the commercial application of hydrogen energy (Ding et al., 2022). Among

GRAPHICAL ABSTRACT |
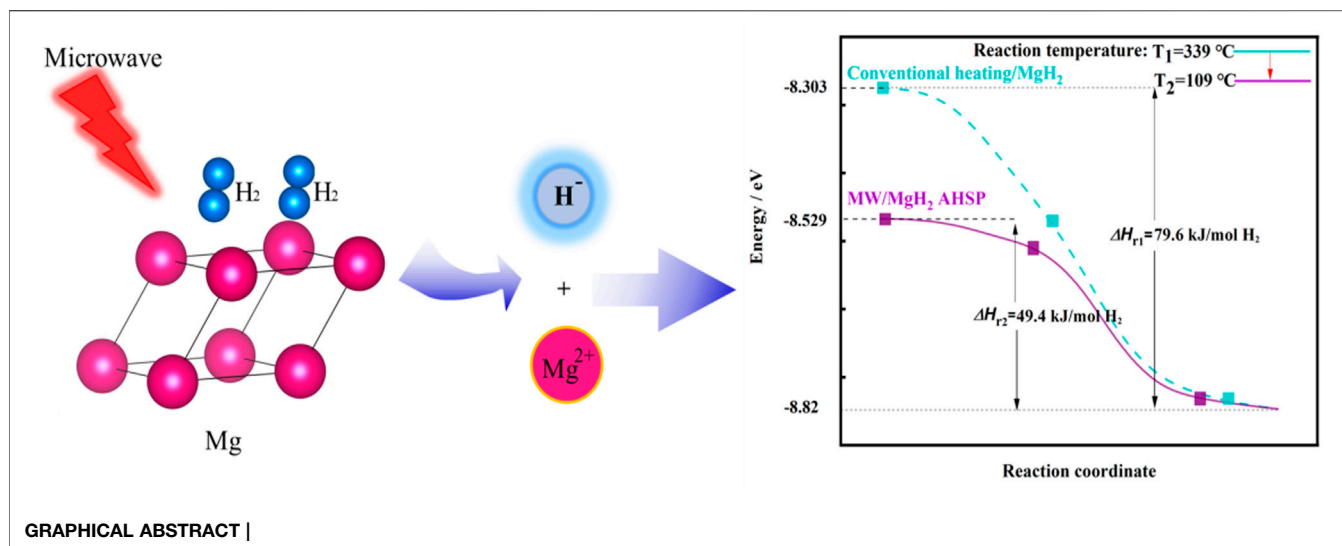
various hydrogen storage methods, the solid-state hydrogen storage method could realize high-capacity storage of hydrogen by reversible adsorption/desorption of solid adsorption materials.

Figure 1 indicates the key role of hydrogen in the renewable energy system. The hydrogen sources from many kinds of renewable energy include wind energy, solar energy, nuclear energy, and others. Green hydrogen is produced by electrolyzation of water (Song et al., 2021), using an alkaline electrolyzer, proton exchange membrane (PEM) electrolyzer (Wang et al., 2021), or solid oxide electrolysis cell (SOEC). Hydrogen is transported by vehicle carriers and stored through high-pressure storage, low-temperature storage, and solid-state storage. Soon afterward, hydrogen needs to be used to supply power residents live and industry. The released hydrogen from storage materials is returned to the fuel cell to generate electricity and heat. It is a benefit for the continuous operation of the renewable energy system to store energy through hydrogen media. At the same time, hydrogen also realizes the recycling when accompanying renewable energy utilization.

MgH_2 is regarded as one of the most promising solid hydrogen storage materials due to its high hydrogen storage density, abundant resource, low cost, high safety, excellent reversible cycle, long life, and mature preparation process (Hirscher et al., 2020). The reaction enthalpy change could even reach as high as 85 kJ/mol H_2 (Bogdanović et al., 1999). The operating temperature is too high. For example, the dehydrogenation temperature is as high as 350°C (Reiser et al., 2000), which is much higher than the vehicle-mounted hydrogen release temperature requirement of the U.S. Department of Energy (DOE) (lower than 85°C) (Department of Energy and United States of America, 2017). The ideal hydrogen storage process is that the reaction process has a low reaction temperature and fast kinetic characteristics.

Actually, a lot of efforts have been made to improve the Mg-based hydrogen storage performance by nanocrystallization, element doping, catalytic modification, and other hydrogen

storage methods. Zaluska et al. (Zaluski et al., 1997) found that the hydrogen storage capacity and kinetic properties will be significantly improved when the grain size is reduced to 50 nm. Zhu et al. (Cao et al., 2015) doped In, Al, and Ti elements into Mg-based hydrogen storage materials. The reaction enthalpy is changed by the affinity of doped elements. As a result, the reaction enthalpy is reduced to 65.2 kJ/mol H_2. In addition, Shaw et al. (Zhong et al., 2016) formed $Li_2B_{12}H_{12}$ intermediate product and changed the reaction path of MgH_2 to achieve reversible hydrogen absorption and desorption at 150°C by compositing MgH_2, $LiBH_4$, phase and C powder. However, these methods cannot simultaneously guarantee the high performance of thermodynamics and kinetics. The improvement of thermodynamics is often at the expense of kinetics and hydrogen storage. The Mg_4NiPd alloy with a BCC-based CsCl-type structure could achieve hydrogen storage at near room temperature but with the hydrogen capacity of 0.7 wt% only (Edalati et al., 2018). Therefore, it is significant and essential to developing a new hydrogen storage approach with good thermal and kinetic properties and also low energy consumption.

Recently, attention in different fields of chemistry has been increasingly paid to using microwave energy to improve reaction properties. Omran (Omran et al., 2020) investigated the effect of microwave energy on the carbothermic reduction of zinc oxide. It was found that the required temperature is reduced from 950°C to 550°C. This indicates that microwave energy has the potential for improving the thermodynamic performance of the reaction process. In addition, microwave plasma is often used as an effective auxiliary way to enhance mass transfer and activate reactions due to its strong penetration, non-contact effect, and various special effects caused by highly active plasma. This is because that microwave plasma can stimulate the reaction substances to produce plasma containing rich active particles, which is especially suitable for adverse reactions of thermodynamics or kinetics. For example, ammonia synthesis generally requires
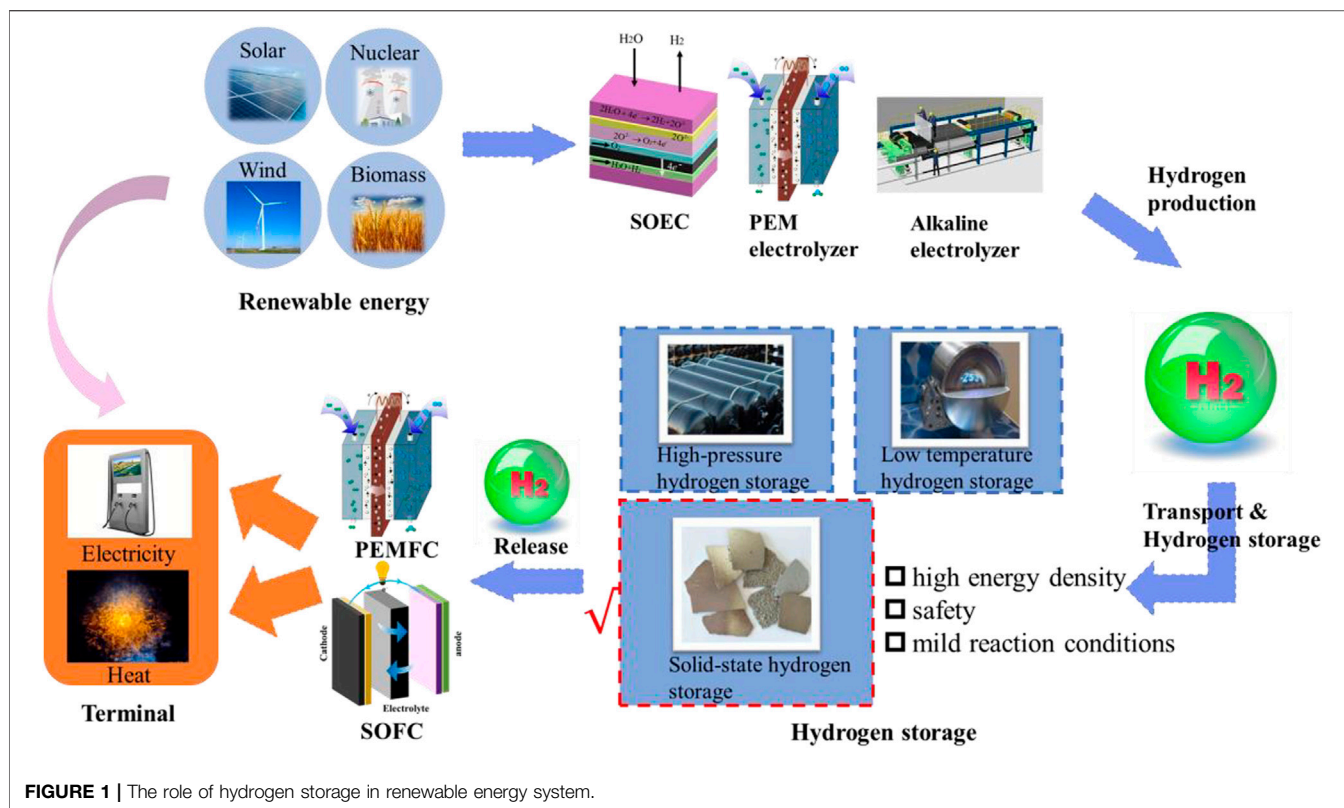
FIGURE 1 | The role of hydrogen storage in renewable energy system.

high temperature and pressure, which inevitably results in large energy consumption and a poor economy. In Kunimori's scheme, the nature of plasma is used to atomize nitrogen (Kunimori et al., 1992). The high-energy particles and catalysts interact with each other with the help of plasma to improve the reaction kinetics by the E-R (Eley-Rideal) mechanism. Accordingly, the ammonia synthesis with the increase of ammonia yield by 6.5 times can be obtained at room temperature and atmospheric pressure (Hong et al., 2018). Eun et al. (Lee et al., 2019) used microwave plasma to remove the solvent that coordinates and fills the gaps in metal-organic frameworks (MOF) and finally achieved a good activation effect. It takes only 30 min to complete the whole activation process, which greatly improves efficiency. In the reaction of methane catalytic conversion to hydrocarbon and hydrogen, microwave plasma forms highly active free radicals by interacting with the catalyst surface, thus changing the theoretical reaction path, but the actual energy efficiency is only 5.5% (Suib and Zerger, 1993). These facts show that microwave plasma enables to strengthen the chemical reaction process by virtue of its own heating effect, plasma effect, and other properties.

In this article, the effect of microwave plasma on improving the thermodynamic performance of Mg-based hydrogen storage is investigated by using density functional theory (DFT) calculations. The activation mechanism of microwave plasma on Mg-based hydrogen storage is uncovered. In addition, the economic feasibility of this kind of newly enhanced technology is evaluated based on the potential application of supplying
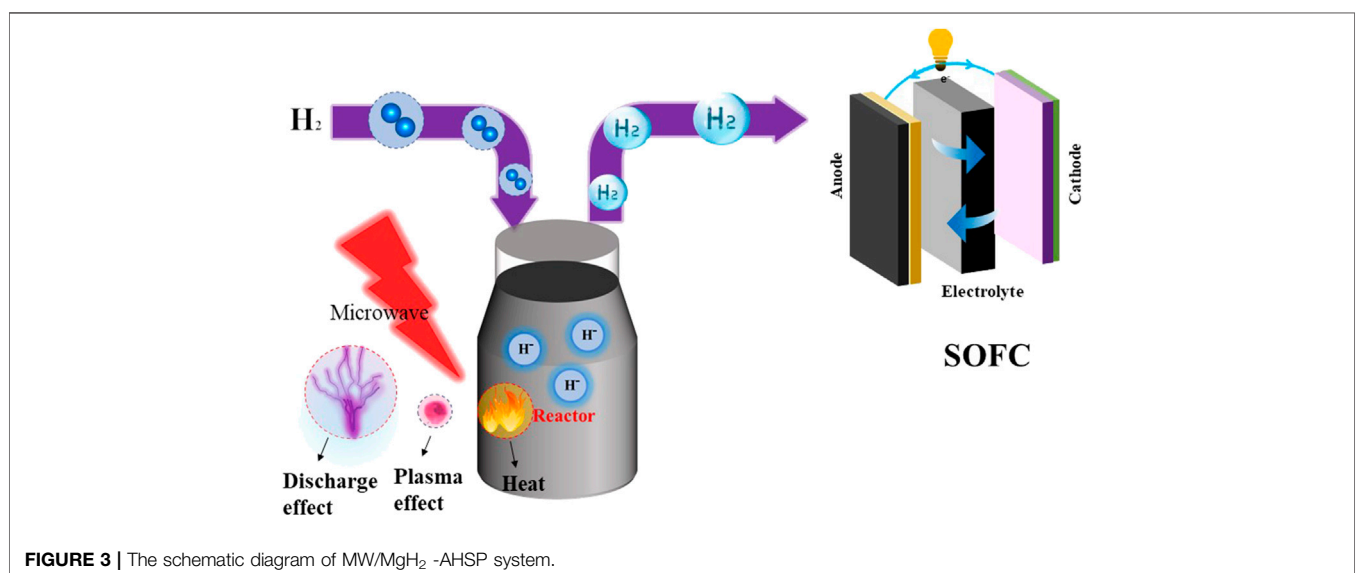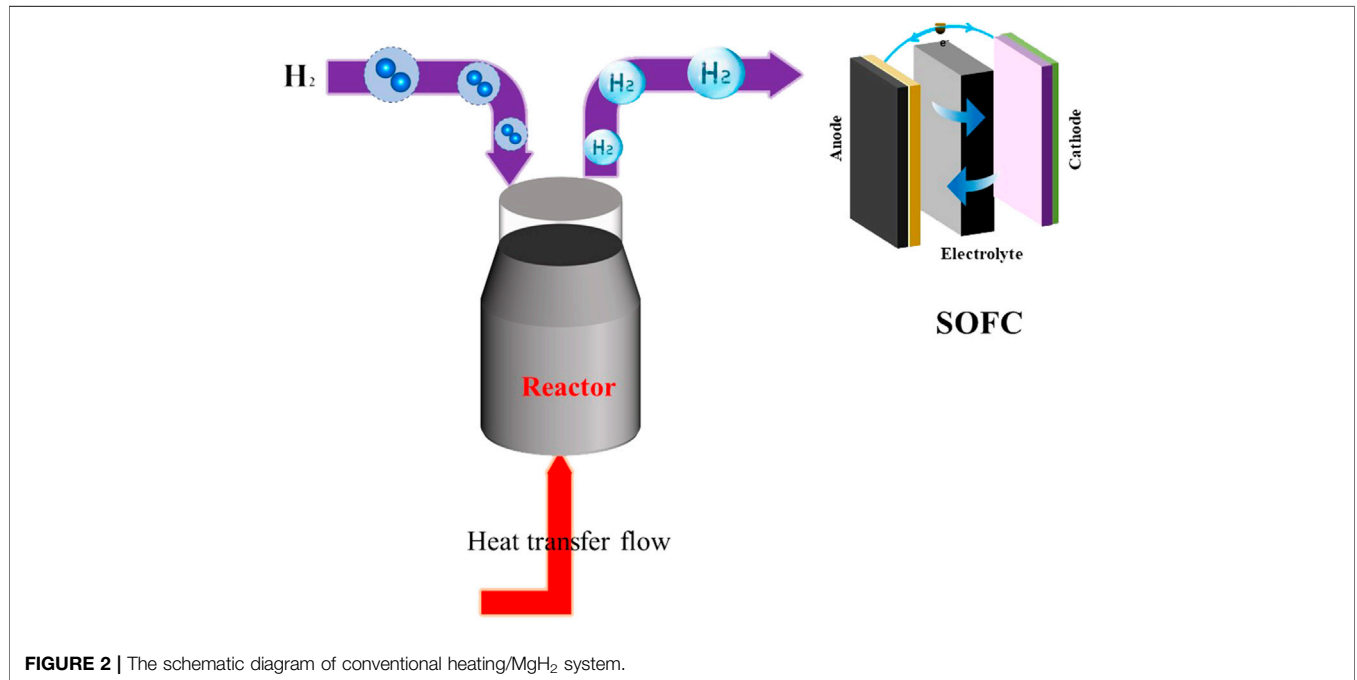
hydrogen to solid oxide fuel cells (SOFC) for heat and power co-generation.

PRINCIPLES OF MICROWAVE PLASMA

In recent years, there are more and more research focusing on a microwave-assisted method to enhance chemical reactions. Microwave is a kind of non-ionic radiation energy. It is generally believed that, under the action of microwave, the energy levels of polar molecules are matched with the rotational energy levels of polar molecules. The microwave promotes the polar conversion movement of substances, including the rapid rotation of molecules, the fracture of chemical bonds, and the friction and collision between the molecules, thus increasing the energy of activated molecules. As a result, the reaction activity of substances is greatly improved (Sun et al., 2016). Owing to the special "plasma effect" and "discharge effect", microwave-induced metal discharge technology has been widely concerned (Chehade et al., 2020). When the metal satisfies certain conditions, the electromagnetic field produced by the microwave enriches the charge around the material. When the charge reaches a certain strength, it will produce electric discharge and break down the ambient gas, showing the "discharge effect" and "plasma effect". Therefore, the microwave plasma has the characteristics of high ionization degree and high energy density. This contributes to producing active components which have a certain catalytic potential for chemical reactions. The active components are

Working substance	Helium	Argon	Hydrogen	Oxygen	Air	Aluminum oxide	Distilled water	Benzene	Toluene	Silicon dioxide
Dielectric strength (MV/m)	0.15	0.18	0.50	0.92	0.97	13.4	65–70	163	199	470–670

FIGURE 2 | The schematic diagram of conventional heating/MgH$_2$ system.

FIGURE 3 | The schematic diagram of MW/MgH$_2$ -AHSP system.

even able to trigger physical and chemical reactions that could not be completed under the normal operating conditions. That's to say, microwave plasma enables to stimulate the active media of chemical reaction and to self-produces the heat in the meanwhile. By comparison, for the common methods, the catalyst is always used as active media to accelerate the reaction. However,

FIGURE 4 | The crystal structure model of Mg and MgH_2.
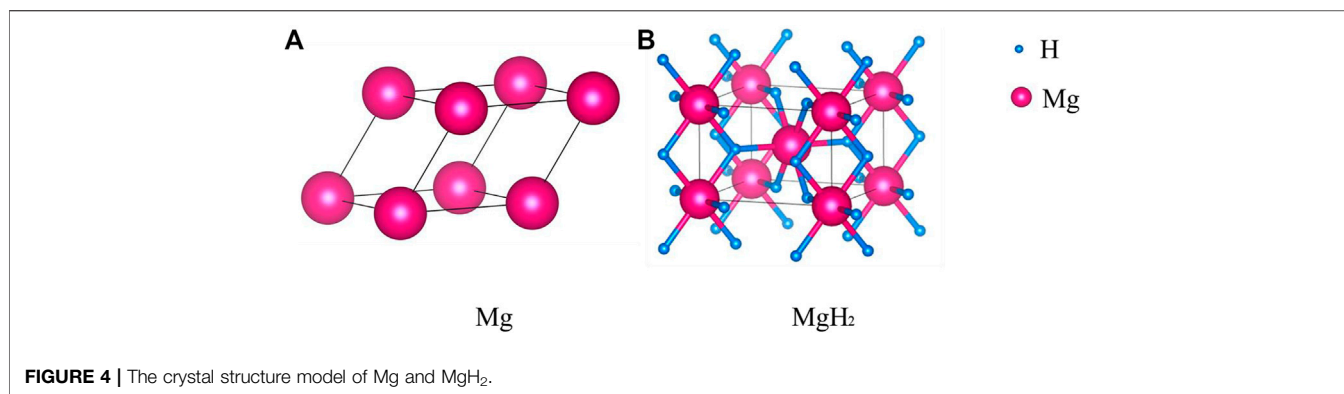
auxiliary heating equipment should be used to provide heat energy.

The plasma effect requires that the working fluid can be ionized at least. In fact, hydrogen is the best easily ionized or broken-down gas except for inert helium and argon. The required energy for ionizing hydrogen is also small due to the low dielectric strength of only 0.5 MV/m. The dielectric strength coefficient of common working fluids is summarized in **Table 1**. Moreover, the electron escape ability from Mg metal is not high. The work function of the electron from the Mg element is about 3.66 eV, which is lower than those from transition metals such as Fe, Cu, Ni, and so on. That is to say, the electron is easy escape from the metal surface of Mg after external stimulation.

In this article, two kinds of MgH_2 hydrogen storage processes are analyzed. One is a conventional heating hydrogen storage process based on MgH_2 (marked as conventional heating/MgH_2 thereinafter) with a conventional heat source as reaction energy. The other one is microwave enhanced advanced hydrogen storage process based on MgH_2 (marked as MW/MgH_2 -AHSP thereinafter).

Figure 2 shows the schematic diagram of a solid-state hydrogen storage system with conventional heat source as reaction energy (conventional heating/MgH_2 system). **Figure 3** illustrates the schematic diagram of solid-state hydrogen storage system with microwave as the auxiliary approach. This system is MW/MgH_2 -AHSP system. Both of the hydrogen storage systems are connected to SOFC as terminal applications of hydrogen. The heat of the conventional heating/MgH_2 system is mainly supplied by boiler steam, while the microwave hydrogen storage system is supplied by microwave energy. At the same time, due to the special effect of microwaves, a large number of plasma containing highly active radicals, ions, and molecules are produced. In addition, plasma can regulate the gas velocity, making the most reactions involving gas media work. These advantages usually lead to the reduction of reaction time, which is also beneficial to reducing reactor size and manufacturing cost.

The process of hydrogen storage assisted by microwave plasma is shown in three basic stages in this model:

(1) Microwave generated by controlling magnetrons

TABLE 2 | Energy of species involved in the reaction (Park et al., 2009).

Species	Mg	H_2	MgH_2	Mg^{2+}	H^-
Energy/eV	−1.49	−6.77	−8.82	−2.11	−1.96

(2) The propagation of microwave
(3) Ionizing hydrogen in the Mg/MgH_2 system, inducing the formation of H^- ion

In the second process, due to the special nature of the microwave, the material of the reactor that transmits the microwave must be guaranteed to make the microwave easy to penetrate. The reactor material is suggested to use ceramics, plastics, and glass with a small dielectric loss coefficient, which contributes to minimizing microwave losses.
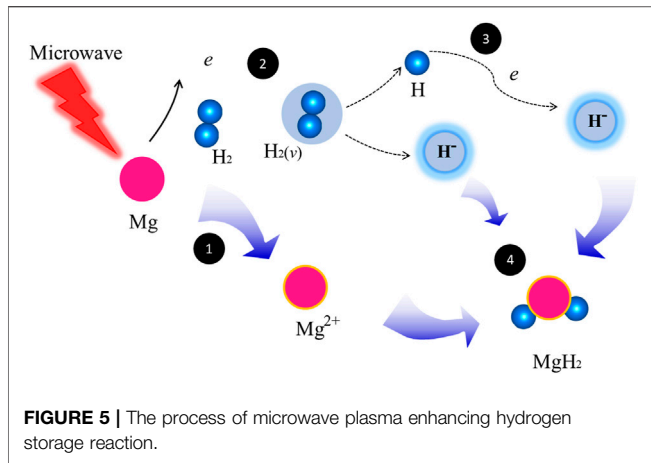
In the third process, appropriate high-frequency electromagnetic fields can ionize the reaction medium, and produce high-energy electrons, activated H atoms, hydrogen ions, and other high-energy particles.

THERMODYNAMIC PERFORMANCE EVALUATION

DFT Calculations

Theoretical calculation based on DFT is widely used to study the reaction mechanism of the hydrogen storage system. Because this study involves microwave field and H^- ion, it is difficult to carry out experiments at this stage. Therefore, the crystal structure and energy change of the reaction system are calculated by simulation calculation in the first place. Based on the DFT calculations, the change in thermodynamic properties of the reaction before and after the introduction of the microwave field could be deduced. It is essential to study the activation mechanism of microwave plasma to improve the thermodynamics of Mg-based hydrogen storage from the theoretical aspect.

Before the calculations of energy and properties, the geometry optimization of the unit cell should be first performed to achieve the most stable structure of the crystal. The projection augmented wave method (PAW) was employed to describe the interaction between ions and electrons in the simulation calculation. The

FIGURE 5 | The process of microwave plasma enhancing hydrogen storage reaction.

FIGURE 6 | Reaction enthalpy comparison between Mg-based hydrogen storage with and without the microwave plasma.

iteration stops when the force on all atoms during the ion relaxation process is less than 0.02 eV /Å, and the total energy variation between the two iterations in the electron relaxation process is less than 10^{-5} eV. For the calculation of MgH$_2$ and H$_2$ energy, the cut-off energy is 400 and 350 eV, respectively.

As for free hydrogen molecules, the gas molecule is put into a huge cubic unit cell ($a = b = c = 10$ Å) under the periodic boundary condition for isolating these molecules. As a result, the interaction between different unit cells of the H$_2$ molecules could be ignored. Moreover, the Monkhorst-Pack mesh k-point for the free gas molecules was set as $3 \times 3 \times 3$.

The structure models of magnesium and magnesium hydride are constructed as shown in **Figure 4**. **Figure 4A** is the crystal structure of Mg, whose space group is P1(#1–1) with the lattice constant of $a = b = c = 3.186$ Å, $\alpha = \beta = \gamma = 60°$. **Figure 4B** shows the crystal structure model of magnesium hydride with 2 Mg atoms and 4 H atoms in the cell, whose space group is P42/mn (#136–1) with lattice constant of $a = b = 4.517$ Å, $c = 3.02$ Å, $\alpha = \beta = \gamma = 90°$.

Comparison of Thermodynamic Properties

Mg and hydrogen molecular generally react in the process of storage as shown in the following equation:

$$Mg + H_2 \rightarrow MgH_2 + \Delta Hr_1 \qquad (1)$$

The reaction enthalpy change (ΔH_{r1}) is calculated by analyzing the energy of reactants and products. Based on the previous DFT energy calculation results, the energies of all the species involved in the hydrogen storage reaction are calculated and summarized in **Table 2**. Therefore, the reaction enthalpy ΔH_{r1} of the conventional heating/MgH$_2$ process could be calculated as follows:

$$\Delta H_{r1} = E(MgH_2) - [E(Mg) + E(H_2) + ZPE(H_2)], \qquad (2)$$

where $E(MgH_2)$ represents the total energy of MgH$_2$, $E(Mg)$ and $E(H_2)$ represent the energies of Mg and H$_2$, respectively. $ZPEH2$ represents the zero-point energy of hydrogen molecule. This is because the energy of light elements in DFT calculation is inaccurate due to ignoring the contribution of vibration. The ZPE
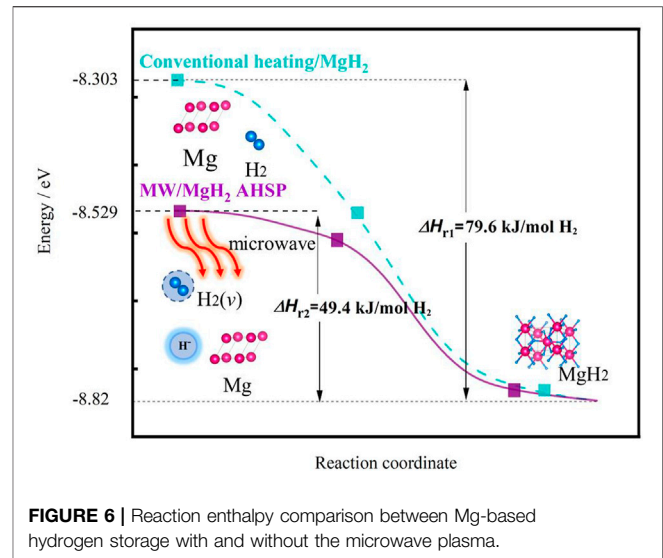
for H$_2$ is calculated to be 0.27 eV in the previous study (Wu et al., 2018). Accordingly, the reaction enthalpy ΔH_{r1} is 79.6 kJ/mol H$_2$, which agrees well with the reported data of 73–85 kJ/mol H$_2$ (Sun et al., 2016; Hirscher et al., 2020).

As is well known, the reaction could happen and reach the equilibrium when the Gibbs free energy change is $\Delta G = 0$. The relationship between ΔH and reaction temperature T is expressed in the following equation:
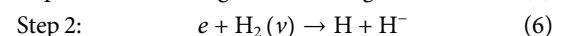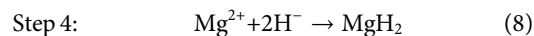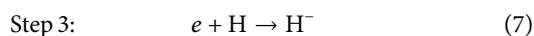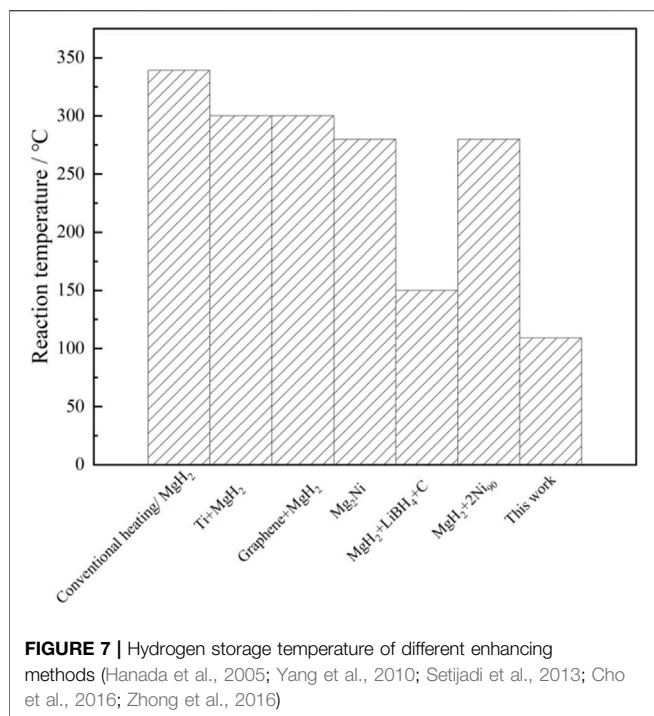
$$\Delta G = \Delta H - TS, \qquad (3)$$

$$T = \frac{\Delta H}{\Delta S}. \qquad (4)$$

The corresponding value of reaction entropy change ΔS is 130 J/(mol·K) (Bogdanović et al., 1999). As a result, the reaction temperature of conventional Mg-based hydrogen storage without the microwave plasma is calculated to be about 339°C, which is also close to the reported reaction temperature of 350°C (Bogdanović et al., 1999). The calculation error is approximately 3.2%, indicating that the prediction results by DFT are reasonable to evaluate the thermodynamic properties of the MgH$_2$ hydrogen storage system.

Figure 5 shows the detailed mechanism of the MW/MgH$_2$ -AHSP system. When the microwave works, the reactant is stimulated to produce high-energy electrons and other high-activity particles. Next, high-energy electrons in the plasma collide with the vibration-state hydrogen molecules, thus producing the excited H$^-$ ions and H atoms (Janev et al., 1987). The produced H atoms further capture low-energy electrons in plasma to generate H$^-$ ions. That is to say, H$^-$ ions are provided in two ways. Finally, H$^-$ ions spread along the gap in the lattice, and combine with Mg^{2+} ions freely.

In this case, the hydrogen storage in plasma is channeled by vibrational and electron excitation as follows:

Step 1: $\qquad Mg + MW \rightarrow Mg^{2+} + 2e \qquad (5)$

Step 2: $\qquad e + H_2(v) \rightarrow H + H^- \qquad (6)$

FIGURE 7 | Hydrogen storage temperature of different enhancing methods (Hanada et al., 2005; Yang et al., 2010; Setijadi et al., 2013; Cho et al., 2016; Zhong et al., 2016)

Step 3: $\qquad e + H \rightarrow H^-$ $\hfill (7)$

Step 4: $\qquad Mg^{2+} + 2H^- \rightarrow MgH_2$ $\hfill (8)$

Microwave irradiates on nano-scale magnesium particles with sharp tips, giving rise to a large energy focus in the microwave region. At the same time, the nano-scale surface roughness further enhances the plasma effect, making Mg convert into Mg^{2+} and releasing active electrons. Subsequently, H_2 and H are ionized as H^- via vibrational and active electron excitation.

That is to say, the active components produced in the atmosphere of microwave plasma change the reaction path. Accordingly, the overall reaction equation under microwave plasma is deduced as follows:

$$Mg^{2+} + e + H + H^- + E_A \rightarrow MgH_2 + \Delta H_{r2}, \qquad (9)$$
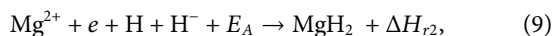
where e is a low-energy electron and E_A is external energy to overcome energy potential barriers, whose value is 0.75 eV. Species activated by microwave plasma require different energy levels to overcome the different barriers of ion hop during diffusion. Furthermore, when electrons combine with the hydrogen atom, the surface electrons need to break through a potential barrier. It is considered that microwave provides energy (E_A) to overcome this potential barrier.

The reaction enthalpy ΔH_{r2} of MW/MgH$_2$ - AHSP could be calculated in the following formula:

$$\Delta H_{r2} = E(MgH_2) - \left[E(Mg^{2+}) + \frac{1}{2} E(H_2) + E(H^-) + E(e) + E_A \right]. \qquad (10)$$

It is noteworthy that the energy of the low-energy electron is assumed to be -0.1 eV (Janev et al., 1987). In addition, the hydrogen anion is difficult to obtain energy values directly by DFT calculations. Hence, the energy of hydrogen anion is calculated by the following formula:

$$E(H^-) = E(H_{ground\text{-}state} - Reference(H), \qquad (11)$$

where $E(H_{ground\text{-}state})$ represents the ground state energy of hydrogen atom, Reference(H) is the value of H in POTCAR file, which is calculated as -1.96 eV.

As a result, the reaction enthalpy ΔH_{r2} of Mg-based hydrogen storage with the help of microwave plasma is significantly reduced to 49.4 kJ/mol H$_2$. The comparison of reaction enthalpy between the Mg-based hydrogen storage with and without the use of microwave plasma is illustrated in **Figure 6**. Accordingly, the hydrogenation temperature is also reduced to about 109°C, which is close to the vehicle-mounted hydrogen release temperature requirement of DOE. **Figure 7** also compares the reaction temperature of Mg-based hydrogen storage between different enhancing methods, such as MgH$_2$+LiBH$_4$+C (Zhong et al., 2016), graphene + MgH$_2$ (Cho et al., 2016), MgH$_2$+2Ni$_{90}$ (Yang et al., 2010), Ti + MgH$_2$ (Setijadi et al., 2013), and Mg$_2$Ni (Hanada et al., 2005). It can be clearly seen that the microwave plasma activating hydrogen storage method has the lowest reaction temperature, which strongly indicates that this new modification method is feasible to reduce the hydrogen storage temperature of magnesium hydride.
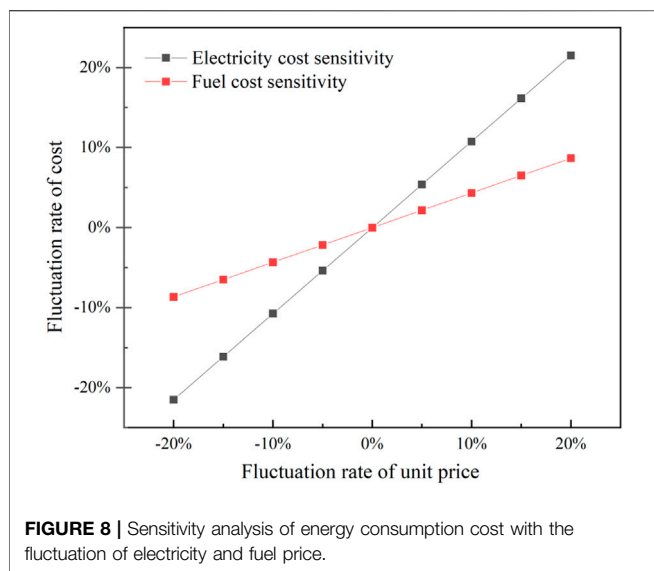
ECONOMIC FEASIBILITY EVALUATION

Comparison of Energy Consumption

Based on the previous analysis, it can be seen that a high ionization and dissociation are needed to ensure the production of active particles. It was reported the microwave power of 6 kW is stable for methane transformation (Zamri et al., 2021). Since the dielectric constant of hydrogen is 0.5 MW/m only, which is much lower than that of methane, hydrogen will be more easily ionized by microwave to produce plasma, so the microwave power of 5 kW is supposed to be enough for completing the reaction of producing high-energy hydrogen species.

In the Mg-based hydrogen storage system, the supply of heat is indispensable for the release of hydrogen. Therefore, it is necessary to provide thermal energy for the whole system with the help of external heat sources. Generally, heat is generated by steam via the boiler. Such heat sources need a long-term operation to ensure the stability of the reaction. Usually, the annual operation number is about 8,000 h. By comparison, the process of microwave plasma activating reaction does not require so much time. Specific time is ascertained based on reaction systems, microwave power, and other factors in experiments.

Therefore, it is hard to accurately calculate the reaction time. Some systems need several minutes (Ricard et al., 2001), while some need a few hours (Wongjaikham et al., 2021). But it is certain that the microwave plasma takes much less time than the conventional heating type. In a project funded by DOE, a pulsed microwave catalytic reactor was mentioned, which did not

FIGURE 8 | Sensitivity analysis of energy consumption cost with the fluctuation of electricity and fuel price.

FIGURE 9 | Comparison of energy consumption cost between system 1 and 2.

continuously supply energy. Only when the catalyst and reaction need an energy supply, does the microwave work. It was concluded that the pulsed microwave reactor almost doubles the reaction conversion and the product yield (Hu, 2018). This shows that the microwave can achieve a good cost-performance ratio of input to output.

In addition, the microwave promotes the generation of plasma owing to its own thermal effect. The existence of hot spots caused by microwave plasma makes the reactants vulnerable to activation. Microwave energy can be selectively transmitted between the active site and the reaction intermediate, and will not lose to the surrounding environment, thus improving the energy utilization efficiency (Tiwari et al., 2020).

The hydrogen storage system mentioned in this article is a magnesium hydride reactor coupled with a 300 kW SOFC for a heat and power co-generation system. The difference is that the conventional heating/MgH$_2$ system employs the boiler steam heated by natural gas or coal as the external heat source to provide energy for the whole reaction process. However, the microwave plasma is employed to activate and heat the hydrogen storage reaction as the external energy supply. On one hand, the thermal effect of microwave is used to make the reactants quickly reach the reaction temperature. On the other hand, the generated plasma activates the hydrogen ion and improves the reaction thermodynamics.

In the following analysis, system 1 represents the conventional hydrogen storage system without microwave plasma (conventional heating/MgH$_2$ system), while system 2 is the proposed microwave plasma activating hydrogen storage system (MW/MgH$_2$ -AHSP system). For the two reactor systems, it is assumed that the target hydrogen production is 100 kg. The hydrogen production rate of the magnesium hydride reactor is 40 NL/min (de Rango et al., 2016). The steam boiler generating hot fluid by combustion of natural gas fuel (0.33 \$/m^3) is used to supply energy for system 1. The efficiency of combustion converting to heat is usually 30%. The energy supply of generating microwave for system 2 is consuming electric energy
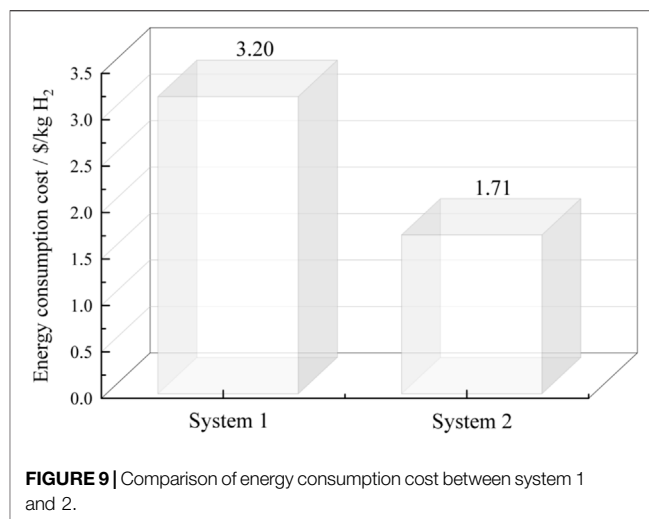
with the price of 0.15 \$/kWh. The reaction time of the pulse microwave reactor is assumed to be half of the conventional reaction time.

Figure 8 shows the comparison of energy consumption between conventional heating/MgH$_2$ system and MW/MgH$_2$ -AHSP system when coupling with SOFC for heat and power co-generation. It can be seen that the energy consumption cost per unit hydrogen mass of system 1 is approximately 3.20 \$/kg H$_2$, which is almost twice as large as that of system 2 (1.71 \$/kg H$_2$). The comparison indicates that the microwave is cost-optimal and energy-saving for the large-scale hydrogen storage system.

Sensitivity Analysis

Sensitivity analysis is further carried out with the change of electricity unit price and natural gas fuel unit price as the conditions. Assuming that the fluctuation range of fuel unit price is −20%~+20%, the corresponding change range of total fuel cost is −8.7%~ +8.7%. For the same fluctuation range of unit price of electricity, the total electricity cost changes from −21% to +21%. By comparison, the sensitivity of fuel or electricity price on the energy consumption cost of systems 1 and 2 could be illustrated as shown in **Figure 9**. It can be seen that the electricity price is more sensitive for system 2 compared with the fuel price for system 1. Therefore, for the application of microwave plasma enhancing hydrogen storage systems, the decision-makers should pay attention to the impact of the fluctuation of electricity prices on the economic benefits of the project.

CONCLUSION

In this article, microwave plasma is introduced to enhance Mg-based hydrogen storage performance. The hydrogenation reaction enthalpy of the MW/MgH$_2$ –AHSP system is about 49.4 kJ/mol H$_2$, which is much lower than the value of 79.6 kJ/mol H$_2$ of the conventional heating/MgH$_2$ system. Accordingly, the reaction temperature is significantly reduced from the initial 339°C–109°C. Compared with the previously reported other

enhancing methods, the reaction temperature of microwave plasma activating Mg-based hydrogen storage is also the lowest. Moreover, the economic feasibility of this kind of newly enhanced technology is evaluated based on the potential application of supplying hydrogen to SOFC for heat and power co-generation. The energy consumption cost is only 1.71 \$/kg H_2, which is nearly half of the energy consumption cost without the microwave plasma. These results show that the proposed microwave plasma activating Mg-based hydrogen storage is feasible and promising to achieve the low-energy-consumption solid-state hydrogen storage in the future.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material; further inquiries can be directed to the corresponding author.

AUTHOR CONTRIBUTIONS

HW: simulation operation, investigation, and writing—original draft. HY: model development and investigation. JR: investigation. BL: review and editing. SNN: review and editing. ZW: conceptualization, supervision, and writing—review and editing.

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NOMENCLATURE

Abbreviation

PEM Proton exchange membrane

SOEC Solid oxide electrolysis cell

DOE U.S. Department of Energy

E-R mechanism Eley-Rideal mechanism

MOF Metal-organic frameworks

DFT Density functional theory

SOFC Solid oxide fuel cell

Conventional heating hydrogen storage process based on MgH$_2$ Conventional heating/MgH$_2$

Microwave enhanced advanced hydrogen storage process based on MgH$_2$ MW/MgH$_2$-AHSP

PAW Projection augmented wave

Symbols

ΔH The change of reaction enthalpy

E Energy

ZPE The zero-point energy

ΔG The change of the free Gibbs energy

T Reaction temperature

E_A The external energy

Subscripts

r1 The conventional hydrogen storage reaction

r2 Microwave plasma enhancing hydrogen storage reaction

ground-state Ground state