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RECENT PROGRESS ON HYDROGEN PRODUCTION FROM SOLAR-THERMAL WATER SPLITTING

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ABSTRACT

The development of renewable energy is a huge opportunity and challenge in the energy field. As the ultimate energy source, solar energy is one of the most reliable energy sources among the renewable energies. Solar water splitting for hydrogen production is one of the most promising technologies of solar energy utilization, in recent decades. As an ideal clean energy source, hydrogen has made a significant breakthrough in transportation and storage technologies and hydrogen is also of great significance for applications such as chemical synthesis and energy storage. This paper reviews the basic methods and development of hydrogen production from solar water splitting, including photovoltaic, photoelectrochemical, and photothermal methods. In view of the photothermal method-solar thermochemical hydrogen production technology, this paper analyzes the thermodynamics, summarizes its future development perspectives. Furthermore, this paper also points out some new ideas for the realization of solar-thermal water splitting for hydrogen production.

Keywords: Solar energy; water splitting; hydrogen production; perovskite; density functional theory.

1. INTRODUCTION

Renewable energy, hydrogen[1], is one of the research topics of the energy field. With the rise of new energy sources, the use of water as a raw material for large-scale hydrogen production by using solar energy or nuclear energy has become a promising energy conversion method. Among all the technologies, solar water splitting for hydrogen production is an essential international frontier, and it is a hot research topic of solar thermal utilization. Solar energy thermally decomposes water or CO2 into H2 and CO, which is then converted into a high-density liquid fuel such as methanol. It not only converts low-density solar energy into high-density fuel chemical energy but also converts water and carbon dioxide into energy products of industrial value.

The method for hydrogen production by solar water splitting mainly includes photovoltaic, photo electrochemistry, photothermal method, and the like. The direct solar thermal [2] splitting method (referred to as "one-step method") refers to the direct heating of water or water vapor by concentrating solar energy. When the temperature reaches the equilibrium temperature of the water splitting reaction, the water splitting reaction is promoted to generate hydrogen and oxygen.

In the standard state, the thermochemical equation of the water splitting reaction is shown as:

$$H_2O(l) \rightarrow H_2(g) + \frac{1}{2}O_2(g) \qquad (1)$$

ΔH=285.84 kJ/mol; ΔG=237.19 kJ/mol;
ΔS=0.163 kJ/mol

It can be seen from the above equation that the Gibbs free energy of the reaction is zero at a temperature of 4427 ° C. That means that the reaction can occur spontaneously at that temperature. It was found that when the temperature reached 2227 ° C, water began to decompose, but the efficiency was very low. Due to the impurities in water, the combination of gas products, the choice of catalysts and the high-temperature solar reactor materials, the water splitting efficiency is

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deficient, which restricts the development of the method. Kogan[3] calculated that the water can hardly be decomposed under the conditions of 1727 ° C and 5 kPa. When the temperature is raised to 2227 ° C, the conversion rate of water splitting is about 25%, and at 2527 ° C, the conversion rate is increased to 55%. For the one-step method, in addition to the problems of high reaction temperature and low conversion rate, there is also a problem of gas separation. The product of the direct water splitting reaction is a mixed gas of hydrogen and oxygen. How to separate hydrogen safely and efficiently is a great challenge in the direct water splitting process.

By adding a catalyst to the water, the water is decomposed into multiple steps, reducing the temperature required for water splitting, shown as fig. (1). In this view, the ΔG of the overall reaction of the thermochemical cycle is consistent with water splitting. By controlling a certain reaction temperature, ΔG = 0 of one step. The heat does not need to be concentrated to supply the water splitting reaction at a very high temperature but is supplied to the water splitting system containing the intermediate medium at different temperatures at different stages. The redox cycle can be represented by the following reaction equation, equation (2) is a metal oxide reduction reaction, equation (3) is a hydrogenation process in which a reduced metal reacts with water, and equation (4) is a total (net) reaction equation of water splitting. Therefore, the process is a cyclic reaction, which only consumes water and can be continuously circulated.

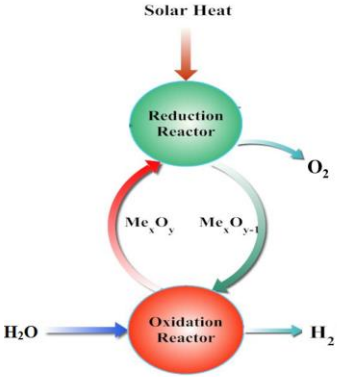

Fig 1 Schematic diagram of solar thermochemical cycle for hydrogen production.

$$MeO \rightarrow \mathrm{Me} + O_2(g) \tag{2}$$

$$\mathrm{Me} + H_2O \rightarrow MeO + H_2(g) \tag{3}$$

$$H_2O \rightarrow H_2(g) + \frac{1}{2}O_2 \tag{4}$$

2. THERMODYNAMICS ANALYSIS

In order to understand the principle of thermochemical hydrogen production, it is necessary to analyze the thermodynamic basis of direct one-step splitting and two-step splitting of water. At any temperature T, for the reversible process to proceed in the forward direction, it is necessary to provide work equivalent to ΔG_T and equal to $\mathrm{T}\Delta S_T$ heat. The theoretical work provided by splitting 1 g of molecular water at the oxidation-reduction temperatures of temperatures T1 and T2 is:

$$W_{T_1} = \Delta G_{T_1}^0 = \Delta H_{T_1}^0 - T_1 \Delta S_{T_1}^0 \tag{5}$$

$$W_{T_2} = \Delta G_{T_2}^0 = \Delta H_{T_2}^0 - T_2 \Delta S_{T_2}^0 \tag{6}$$

Here， $\Delta H_{T_1}^0$ $\Delta H_{T_2}^0$ $\Delta S_{T_1}^0$ $\Delta S_{T_2}^0$ represent the enthalpy and entropy hcange at T1 and T2 for water splitting， assuming that the ΔH^0 and ΔS^0 is independent of temperature, thus, $\Delta H_{T_1}^0 = \Delta H_{T_2}^0 = \Delta H^0$, $\Delta S_{T_1}^0 = \Delta S_{T_2}^0 = \Delta S^0$. Since the overall process is carried out under normal pressure,

$$W_{T_2} = \Delta H_{T_1}^0 - T_2 \Delta S_{T_1}^0 \tag{7}$$

(7)-(5):

$$W_{T_2} = \Delta G_{T_1}^0 - (T_2 - T_1)\Delta S^0 \tag{8}$$

The minimum amount of heat required to break down water at temperatures T1 and T2 is:

$$Q_{T_1} = T_1 \Delta S_{T_1}^0 \tag{9}$$

$$Q_{T_2} = T_2 \Delta S_{T_2}^0 \tag{10}$$

(10)-(9):

$$Q_{T_2} = Q_{T_1} + (T_2 - T_1)\Delta S^0 \tag{11}$$

(8) and (11) indicate the work and heat necessary for water splitting at any temperature. According to equation (8), if you want to reduce the power consumption of water splitting, it is required to increase the temperature. This requires consideration of materials, cost, and efficiency. And other issues. These problems limit the further development of direct water splitting.

At this point, we change the idea of dividing the direct water splitting process into multiple reactions, and controlling the temperature and other factors, so that the entropy becomes positive and the reaction proceeds at a higher temperature so that more heat is extracted from the heat source. The reaction that causes the entropy to become negative is performed as much as possible at a lower temperature to reduce power consumption.

Here, the thermochemical cycle of water splitting is composed of N reactions. To allow any one reaction i to proceed, the theoretical work and heat that must be provided by the outside can be expressed by the following equation:

$$W_{(i)T_2} = G_{(i)T_1} - (T_{(i)2} - T_1)\Delta S_{(i)} \quad (12)$$

$$Q_{(i)T_2} = Q_{(i)T_1} + (T_{(i)2} - T_1)\Delta S_{(i)} \quad (13)$$

the total work and heat of the cycle are：

$$\mathrm{W} = \sum_{i=1}^{N} \langle \Delta G_{(i)T_1} - \Delta S_{(i)}(T_{(i)2} - T_1) \rangle \quad (14)$$

$$\mathrm{Q} = \sum_{i=1}^{N} \langle Q_{(i)T_1} + \Delta S_{(i)}(T_{(i)2} - T_1) \rangle \quad (15)$$

Since the total reaction is water splitting, other substances are only cyclically unchanged, then there is:

$$\mathrm{W} = \Delta G_{T_1} - \sum_{i=1}^{N} \Delta S_{(i)}(T_{(i)2} - T_1) \quad (16)$$

$$\mathrm{Q} = \sum_{i=1}^{N} \Delta S_{(i)} T_{(i)2} \quad (17)$$

(16) and (17) respectively represent the work and heat required for the multi-step reaction, in contrast to (8) (11). The difference between the two is that in the direct water splitting process, when T2 is determined, the required work has been determined, and it is necessary to increase the temperature in order to further reduce the power consumption. The multi-step reaction process can avoid this situation by multiple reactions. From (16), in order to reduce power consumption, it is possible to increase the temperature or select different reaction paths to form different chemical reaction sequences. At this point, the work and heat required for thermal chemical water splitting no longer rely solely on the control of thermodynamic parameters (temperature).

3. METAL OXIDE CATALYST

More than 350 thermochemical cyclic metal oxides have been studied to date, and most materials can theoretically achieve 40%[4, 5]. Nakamura et al. [6] first proposed the iron oxide system, and the oxides of iron with different valence states will release oxygen during the conversion process, thereby having the ability to reduce water to hydrogen. Steinfeld et al. [7] first proposed the Zn/ZnO high-temperature two-step hydrogen production system and carried out a lot of research on this basis, such as system analysis and theoretical calculation, recycling material development, solar high-temperature reactor design. The definition of solar fuel conversion efficiency and ideal efficiency are, respectively (18) and (19). When the concentration ratio is 5000, the maximum efficiency of solar energy conversion is 75% at the optimum temperature of 1227 °C. Daniel et al. [14] used this equation to indicate that when IDNI is 1 kW/m2, concentration ratio C=3000, TH is 1500 °C, and TL is ambient temperature, the ideal efficiency of solar fuel is 68%, although this is not as high as the concentration ratio. The ideal efficiency, but 50% higher than the ideal efficiency of a multijunction optical absorption electrochemical cell.

$$\eta_{solar-to-fuel} = \frac{-\Delta G}{Q_{solar}} \quad (18)$$

$$\eta_{solar-to-fuel,ideal} = \left[1 - \left(\frac{\sigma T_H^4}{IC}\right)\right] \times \left[1 - \left(\frac{T_L}{T_H}\right)\right] \quad (19)$$

Weidenkaff et al. [8] analyzed the influence of reaction temperature, oxygen partial pressure and inert gas flow rate on the reaction kinetics of Zn/ZnO system in the splitting of water to produce hydrogen; Perkins et al. [9] made a lot of research on thermochemical method. And find the spontaneous reaction cycle at low temperatures, such as Mn2O3/MnO, Zn/ZnO, Fe3O4/FeO[10]. In addition, Fe3O4/FeO, TiO2/TiOx, Mn3O4/MnO, Co3O4/CoO[11], etc. have been experimentally verified. For composite metal oxides, scholars have also made a lot of attempts, such as (Fe1-xMx)3O4/(Fe1-xMx)1-yO, M=Co, Mn, Mg, ZnFe2O4/Zn/Fe3O4, all of which have achieved Good results. The above metal oxide oxygen carriers are all stoichiometric oxides, and the products before and after the reaction are all present in the natural environment. Researchers have subsequently proposed the use of non-stoichiometric oxides to form recycled materials, such as CeO2/CeO2-δ [12], which have a wide range of choices and the amount of oxygen released can vary depending on conditions. When the temperature is low, and the partial pressure of oxygen is high, the amount of released oxygen δ still exists, but it is small. When the temperature rises and the partial pressure of oxygen decreases, the number of released oxygen increases. Table 1 gives the reaction equations, temperatures, and efficiencies of the main thermochemical cycle materials.

Table 1. Thermochemical cyclic water splitting reaction

Redox cycle	T	η
ZnO→Zn+1/2 O_2 (g)	1800/ 420	45

$Zn+H_2O \rightarrow ZnO + H_2$ (g)	[13]	
$Fe_3O_4 \rightarrow 3FeO + 1/2\ O_2$ $3FeO + H_2O \rightarrow Fe_3O_4 + H_2$ $MxFe_{3-x}O_4 \rightarrow xMO + (3-x)FeO + 1/2\ O_2$ $xMO + (3-x)FeO + H_2O \rightarrow MxFe_{3-x}O_4 + H_2$ $M_xFe_{3-x}O_4+3Al_2O_3 \rightarrow (3-x)FeAl_2O_{4-x}+xMAl_2O_4+yO_2$ $(3-x)FeAl_2O_{4-x}+xMAl_2O_4+yH_2O \rightarrow M_xFe_{3-x}O_4+3Al_2O_3+yH_2$	1300/ 1000	-
$CeO_2 \rightarrow CeO_{2-\delta} + \delta/2\ O_2$ $CeO_{2-\delta}+\delta H_2O \rightarrow CeO_2 + \delta H_2$ (g) $M_xCe_{1-x}O_2 \rightarrow M_xCe_{1-x}O_{2-\delta} + \delta/2\ O_2$ $M_xCe_{1-x}O_{2-\delta}+\delta H_2O \rightarrow M_xCe_{1-x}O_2 + \delta H_2$ (g)	1500/ 900[14]	4
$SnO_2(s) \rightarrow SnO(g)+ 1/2\ O_2$ $SnO(g)+H_2O \rightarrow SnO_2(s)+ H_2$ (g)	1500/ 600[15]	-
$CoFe_2O_4 \rightarrow CoFe_2O_3+1/2O_2(g)$ $CoFe_2O_3+H_2O \rightarrow CoFe_2O_4+H_2(g)$	1200/ 1000	19
$ABO_3 \rightarrow ABO_{3-\delta} + \delta/2\ O_2$ $ABO_{3-\delta}+\delta/2\ H_2O \rightarrow ABO_3 + \delta/2\ H_2$	<1200	-

The last item in Table 1 is a perovskite-type metal oxide circulating material, and the perovskite-type oxide (ABO3) has a stable octahedral, cubic mixed crystal structure. Since the perovskite type oxide carries an oxygen atom and has good oxygen and oxygen loss ability, it can be used as an oxygen carrier material in the chemical chain. In the perovskite structure, the A site is a rare earth metal, an alkaline earth metal or an alkali metal, which plays a role of supporting the skeleton and changing the valence state of the B site; the B site is usually a transition metal element having a small radius, which determines the catalytic performance of the material. When the tolerance factor of the A and B elements is in the range of 0.75 to 1.0, the oxygen ion coordination of the A and B elements can be achieved to form a perovskite configuration. The tolerance factor (t) is defined as $\mathrm{t} = (r_A + r_O)/\sqrt{2}(r_B + r_O)$, Wherein rA, rB, rO are the A site, the B site ion and the oxygen ion radius, respectively. The special crystal structure of the octahedron and the cube is beneficial to improve the oxygen carrier's property, anti-carbon deposition property, and reduce the reduction reaction temperature. The physicochemical properties of the material can be improved by the doping of different metals at the A and B sites. It is getting more and more attention from domestic and foreign scientific research units and has broad research prospects.

The advantage of the solar thermal chemical cycle is that the temperature required for the reaction of the process is lower than that of the direct thermal splitting method, and there are no by-products in the process, and the environmental problems are not considered, and the products H2 and O2 are generated stepwise, so there is no high-temperature gas separation. However, the reaction in the high-temperature zone is still limited to about 1500 °C. The requirements for the solar concentrating reactor and material reactivity are high, and the thermal radiation loss is large. The actual total efficiency of the system is far from the ideal efficiency. An effective method of the current thermochemical method is to find a metal/metal oxide system which is suitable for splitting at a lower temperature and has good reactivity and cyclability.

4. PEROVSKITE CATALYST

Figure 2. is a schematic diagram of the thermochemical cycle of perovskite as an oxygen carrier material. The reduction reaction absorbs the heat of the solar energy, releases oxygen, and simultaneously obtains a reduced state of the perovskite material. This part of the material reacts with water or carbon dioxide during the oxidation process to produce solar fuel (H2 or CO). The reaction equation is as follows (20) (21):

$$\mathrm{ABO_{3-\delta}} \rightarrow \mathrm{ABO_{3-\delta}} + \delta/2\ \mathrm{O_2} \quad (20)$$

$$\mathrm{ABO_{3-\delta}} + \delta/2\ \mathrm{H_2O} \rightarrow \mathrm{ABO_{3-\delta}} + \delta/2\ \mathrm{H_2} \quad (21)$$

A.H.BorK et al. [23] investigated the reactivity of La0.7Sr0.3CrO3-δ 、 La0.7Sr0.3CoO3-δ 与 CeO2-δ ， Ce0.95Zr0.05O2-δ Ce0.80Zr0.20O2-δ. The amount of oxygen release increases when the CeO2-δ ， Ce0.95Zr0.05O2-δ, Ce0.80Zr0.20O2-δreaches the temperature of 1000 ° C. At 400 ° C, the amount of oxygen release for the La0.7Sr0.3CoO3-δ has increased significantly, and it is four times that of conventional oxides. The ability to decompose H2O and CO2 can be determined by the Gibbs free energy of the reaction. Taking water splitting as an example $\Delta g_{rxn} = \Delta g_{oxd} - \Delta g_{H_2o}$, where Δg_{oxd} is the Gibbs required for oxidation of the material. Free energy, Δg_{H_2o} is the Gibbs free energy which is required for hydrogen oxidation to water. When $\Delta g_{rxn} < 0$ indicates that the hydrolysis reaction can be carried out, and the larger the negative value is favorable, this also indirectly indicates that the larger the negative value of Δg_{oxd}, the more favorable the splitting reaction of water and carbon dioxide.

Antonie et al. [16] calculated 5329 perovskite materials using high-throughput density functional (HT-DFT) and initially screened 383 stable perovskite oxides for two steps. The water splitting reaction was carried out to calculate the stability, oxidation state, and structural properties of the above materials. Figure 9 shows the comparative analysis of oxygen vacancy formation energies of several lanthanide perovskites. It can be seen from the figure that the calculation results of HT-DFT are in good agreement with the experimental results, and the oxygen vacancy formation energy of LaNiO3 is the lowest. Figure 10 shows the distribution of perovskite materials suitable for two-step water splitting. The abscissa is the stability of the structure, and the vertical axis is the oxygen vacancy formation energy. Only when the stability and oxygen vacancy formation energy is < 0.5eV, respectively. It can be applied to the two-step water splitting reaction in the range of 2.5~5 eV.

5. *MULTI-STEP METHOD*

The calculation of the two-step method has been explored in the field of experiment and calculation. However, the reaction-to-materials that can be used on a large scale have not been found so far, and the temperature cannot reach below 1000 °C. Some scholars have explored the multi-step method to further reduce the temperature. Xu et al. [17] studied the boundary problem of thermodynamic properties of metal oxides and obtained the isentropic line and isosceles distribution map of the reduction-oxidation reaction. It can be seen from the figure that the blue region is a thermodynamically achievable reflection region. The boundaries of the area are all above 1000 °C. The study shows that there is a bottleneck in the study of the two-step method. Xu proposed to expand the manganese oxide material and add sodium carbonate material to the system.

6. *HYBRID ENERGY*

The first two sections are all related to the study of water splitting without the introduction of fossil fuels. Nowadays, based on the concept of Chemical Looping Combustion proposed by Ishida and Jin [18] in the 1970s, some researchers applied this concept to the hydrogen production reaction, which is called chemical chain reforming technology.

He et al. [19] prepared perovskite oxides LaFeO3 and La09Sr0.1FeO3 by combustion method. Under the reaction temperature of 900 °C in the thermogravimetry, methane was used as the reducing gas, and the Sr-doped oxygen carrier reduction peak shifted to the low-temperature direction. It shows that the oxygen carrier has enhanced oxygen release capacity at low temperature after doping. Magnus Ryden et al. [20] of the Department of Energy and Environment, Chalmers University of Technology, Sweden, studied the application of perovskite-type oxides in chemical chain combustion and chemical chain reforming and selected the La0.5Sr0.5Fe0.5Co0.5O3-δ perovskite oxide was studied.

Nalbandian L et al. [21] applied La1-xSrxMyFe1-yO3-δ perovskite oxides to chemical chain reforming. In the results of chemical chain reforming performance of La0.7Sr0.3M0.05Fe0.95O3 (M=Ni, Cu, Cr, Co). during the reaction of Cr or Cu doped oxygen carrier, the conversion rate of methane and water vapor is obviously improved, the conversion rate is slightly increased during Ni doping, and the Co doping conversion rate is deteriorated, indicating that Co inhibits fuel conversion. In summary, among the four doping elements, Cr is the most effective. Källén M et al. [22] studied CaMn0.9Mg0.1O3-δ and found that the oxygen release amount of the oxygen carrier increases as the temperature of the fuel reactor increases. When the temperature rises to a certain condition, and the air flow rate is appropriate, an oxygen carrier can release oxygen to 35%. And CaMn0.9Mg0.1O3-δ has excellent resistance to sintering and abrasion.

7. *CONCLUSIONS*

Hydrogen production from solar water is a clean, widely technology of fuel production and energy utilization. Solar thermal hydrogen production and solar photocatalytic hydrogen production are two development ideas. For solar thermal chemical hydrolysis of hydrogen, the choice of circulating media has always been the focus of research and exploration. With the development of solar thermal chemistry technology, the future development trend may be summarized in the following aspects:

(1) Improve the thermodynamic analysis of solar thermal chemical water splitting and find out the maximum utilization efficiency of thermodynamic theory.

(2) Calculate and analyze a variety of doped perovskite-type metal oxides by high-throughput density

functional theory to supplement the vacancies of thermodynamic properties.

(3) Develop a high-efficiency solar reactor suitable for solar thermal chemical hydrolysis to produce hydrogen.

(4) Experimentally develop the perovskite-type metal oxides with excellent performance as a circulating medium for solar thermal chemical hydrogen production.

(5) Further experimental analysis of the cyclic nature of the cyclic carrier in the solar reactor provides experimental support for further industrialization.

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