Effects of Ceria Concentration in the Electrode on Water Decomposition Efficiency

K. Isobe and T. Yamanishi

Tritium Technology group, Japan Atomic Energy Agency, Tokai, Ibaraki, 319-1195, Japan

The highly tritiated water is expected to be produced in the vacuum chamber and in a bleeding blanket of fusion reactor system. A ceramic electrolysis would be only a method to recover tritium from such highly tritiated water. Aiming at improving the water decomposition efficiency of this process, we have developed effective electrode containing cerium oxide (ceria). In this study, the effect of ceria concentration in the electrode on the efficiency has been studied. The current density increased with ceria concentration in the electrode and reached the value of high density 100mA/cm^2 for 30% ceria-containing electrode at 1.5V. This current density is one order of magnitude higher than that of usual (Pt-YSZ) electrode.

Keywords: tritium, highly tritiated water, water detritiation system

I. Introduction

Tritium and deuterium used as fuel in a fusion reactor present in its vacuum chamber and breeding blanket during operation. Tritium ratio to other hydrogen isotopes in the vacuum chamber or breeding blanket is expected to be from 1% to 50%. Therefore, water presented in the vacuum chamber and bleeding blanket is highly tritiated and its concentration will reach 3.0×10^{18}Bq·m^{-3} when tritium ratio is 50%. Therefore it is one of the key issues to recovery tritium from such highly tritiated water from the viewpoint of tritium inventory control in the vacuum vessel as well as tritium recover as a fuel. To reuse tritiated water as fuel, it should be reduced to hydrogen molecule which will be sent to the isotopes separation system in fuel cycle system. In ITER, electrochemical method using solid polymer electrolysis is selected for Water Detritiation System (WDS). In this system, tritium concentration of processing water is limited under 1.1×10^{16}Bq·m^{-3} due to the radiation damage of solid polymer. On the other hand, a ceramic electrolysis can be used even in high beta-ray radiation doze because it is not damaged by beta-ray from tritium. Therefore, the ceramic electrolysis would be only a method to recover tritium from the highly tritiated water. In the development of Fuel Cleanup system for ITER, ceramic electrolysis systems for processing both hydrocarbon and water had been developed [1]-[3]. For increase of efficiency on ceramic electrolysis, we had developed platinum and yttria stabilized zirconia (Pt-YSZ) compound electrode. For applying these systems to process tritiated water, characteristic of components should be improved and the efficiency of water reduction should be increased. We have developed new two types of electrode containing cerium oxide (ceria) to increase the efficiency of water reduction [4]. In this study, we focused on the electrode containing ceria and the effect of ceria concentration in the electrode on water decomposition efficiency was examined.

II. Principle of Ceramic Electrolysis Method

Figure 1 shows the schematic diagram of principle of the ceramic electrolysis method which is basically the same as the electrolysis of water using a polymer electrolyte. In both cases, the reduction of water take place on the electrode supplied with an electric potential. YSZ, which is well used as an oxide ion conductor, is selected as a solid ceramic electrolyte because of its stable performance. Electric charges needed for the reduction is supplied to the electrode on the YSZ at a high temperature (>873K) and water molecules reaching to electrode are reduced on the electrode. Hydrogen molecules produced are released from the electrode to the environment. Oxygen ions produced are dissolved into YSZ and permeated though it. On the anode, oxygen ions recombine with each other to form oxygen molecules which are released to environment. Thus, Hydrogen and oxygen produced can be separated into the different sides of electrolyte.

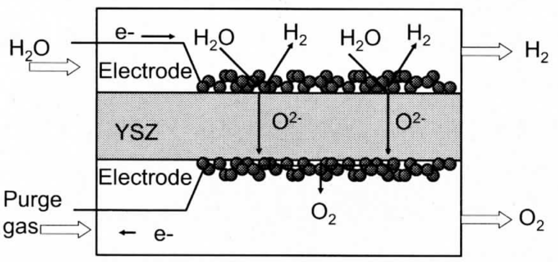

Fig.1 Schematic diagram of principle of ceramic electrolysis method

To improve the efficiency of the reduction reaction, many studies were carried out in the field of Solid Oxide Fuel Cell (SOFC). For example, the development of electrode has been performed to decrease over-voltage needed for reaction and to increase reaction point densities. Manufacturing electrodes with porous structure improves apparent mobility of water vapor and so on. In this study, we focused on the increase of reaction point density on the electrode. Figure 2(a) shows schematic diagram around the reaction point. It is considered that electrode reaction takes place on the three-phase boundary consisting of electrolyte, electrode and gasous phase. When electrode has both high oxygen ion conductivity and electron conductivity, reaction can occur on not only the three-phase boundary but also the surface of electrode, and the density of reaction points will increase as shown in Fig.2 (b). Ceria has not only high oxygen ion conductivity like YSZ but also electron conductivity. Therefore electrode containing Ceria has been developed and shows high performance in the field of SOFC.[4,5] From the knowledge of SOFC, we tried to develop new electrodes containing Ceria to increase the efficiency of water vapor reduction.

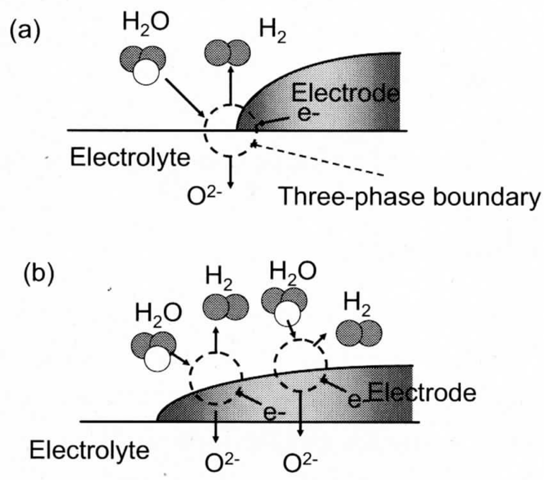

Fig.2 Schematic diagram of reaction point
(a) Three-phase boundary model
(b) Mixed conductivity electrode

III. Experimental

Electrodes containing different concentration of Ceria were prepared for this experiment. The Pt-Ceria paste with different mixing ratio (10%, 20%, 30%) was coated on both sides of YSZ disc (22mm in diameter, 1.5mm^t). To collect electric charge, Pt mesh was coated on all electrodes. To compare the electrodes containing Ceria with usual (Pt-YSZ) one, the Pt-YSZ electrode in the same size was also prepared. Composition of this electrode is 90% of Pt, 10% of YSZ. Water reduction experiment was carried out at 1073K in

Argon atmosphere under controlled water vapor concentration.

IV. Results and Discussion

Figure 3 shows a plot of current density of four types of electrodes against applied voltage at 2400ppm water vapor concentration. The efficiency of decomposition can be expressed as current, since only oxygen ions can diffuse though the YSZ electrolyte.

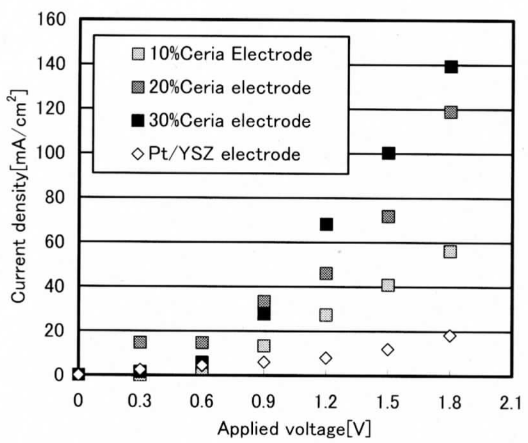

Fig.3 Current density of electrodes against applied voltage

Current density of all electrodes containing ceria is higher than that of Pt-YSZ. This result shows Ceria in electrode enhances strongly water decomposition. In the ceria electrodes, the current density increases with ceria concentration in the electrode and reaches high density (100mA/cm^2) in 30% ceria electrode at 1.5V. This current density is about one order magnitude higher than that of Pt-YSZ electrode. These results proved that ceria concentration in electrode strongly affects the water decomposition efficiency and the usage of such oxide containing can improve the decomposition efficiency dramatically. This improvement of efficiency is considered to be due to not only the increase of reaction points but also the change of microstructure of the electrode. In the previous paper [4], it was found that ceria in electrode changed the surface microstructure and modified the electrode resistance. To clarify the effects of ceria concentration, further investigations, such as the observation of electrode and the measurement of electrode resistance, would be required.

V. Conclusion

Electrodes containing different concentration of Ceria were prepared and water decomposition efficiency of these electrodes was studied. The current density increased with ceria concentration in the electrode and reached the value of 100mA/cm^2 for in 30% ceria-containing electrode at 1.5V. This current density is about one order of magnitude higher than that of usual (Pt-YSZ) electrode. These results proved that ceria concentration in electrode strongly affects the water decomposition efficiency. To clarify the effects of ceria concentration, further investigations, such as the observation of electrode and the measurement of electrode resistance, would be required.

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