Fabrication of metallic channel-containing UO2 fuels

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1. Introduction

The uranium dioxide is widely used as a fuel material in the nuclear industry, owing to many advantages. But it has a disadvantage of having the lowest thermal conductivity of all kinds of nuclear fuels; metal, carbide, nitride [1].

It is well known that the thermal conductivity of UO₂ fuel is enhanced by making, so called, the CERMET (ceramic-metal) composite which consists of both continuous body of highly thermal-conducting metal and UO₂ islands [2,3]. The CERMET fuel fabrication technique needs metal phase of at least 30%, mostly more than 50%, of the volume of the pellet in order to keep the metal phase interconnected. This high volume fraction of metal requires such a high enrichment of U that the parasitic effect of metal should be compensated. Therefore, it is attractive to develop an innovative composite fuel that can form continuous metal phase with a small amount of metal.

In this investigation, a feasibility study was made on how to make such an innovative fuel. Candidate metals (W, Mo, Cr) were selected, and fabrication process was conceptually designed from thermodynamic calculations. We have experimentally found that a metal phase envelops perfectly UO_2 grains, forming continuous channel throughout the pellet, and improving the thermal conductivity of pellet.

2. Experiments

2.1. Sample preparation

The key process of our CERMET fuel fabrication is the oxidation of metal to oxide, melting and then reduction of it. W or Mo powder containing UO $_2$ green pellets were sintered in $\rm H_2$ at $1700\, \text{for}\ 4\ h$. The sintered pellets were annealed at appropriate temperature in CO $_2$ /CO mixing gas atmosphere and then subsequently heated to $1650\, \text{Gn}\ H_2$ and maintained for $2\ h$. The annealing temperature and CO $_2$ /CO ratio were determined so that the WO $_3$ and MoO $_3$ phases were stable and melted.

For the Cr-containing CERMET, Cr_2O_3 -containing UO_2 green pellet was heated to $1700\,^{\circ}\text{Cn}$ H_2/H_2O mixed gas and then held for 4h. The sample was subsequently annealed in H_2 for 2 h at the same temperature.

2. 2. Characterization of the CERMET fuels

The microstructures of polished pellets were observed with an optical microscope. The detailed morphology of grain structure and cation distribution were investigated by SEM and electron probe microanalysis.

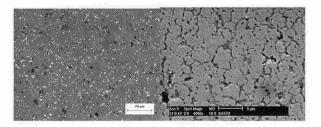
The thermal diffusivity was measured by the laser flash method from room temperature to 1400°C in vacuum using a laser flash apparatus (Netzsch LFA 427). The measured thermal diffusivities were tentatively compared with that of pure UO₂ sample. The relative difference in thermal conductivity between metal and UO₂ was measured with scanning thermal microprobe.

3. Results

3.1. W. Mo-CERMET fuels

Figs. 1a, 1b and 1c show the microstructures of the metallic particle-dispersed UO₂ pellet (fig. 1a), annealed pellet of Mo dispersed UO₂ in CO₂/CO atmosphere (fig. 1b), and the UO₂-W CERMET pellet obtained by reduction of annealed pellet (fig. 1c), respectively.

The microstructure characteristics and EPMA results indicated that the W or Mo were oxidized and melted during the heat treatment step. The melted oxide penetrated along the grain boundary of UO₂ and is interconnected with each other. During the reduction step the liquid metals were transformed to solid metals, which were precipitated along the grain boundary.



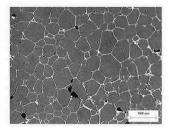


Fig. 1. The pellet microstructure transition; (a) as sintered, (b) annealed in CO_2/CO atmosphere, (c) reduced in H_2 gas.

Fig. 2 shows the ratio of thermal diffusivity of 6vol% W containing UO₂-W CERMET to pure UO₂ pellet. The

thermal diffusivity of UO₂-W CERMET is increased by about 40~80% than that of pure UO₂ sample.

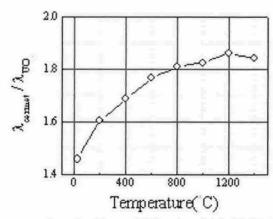


Fig. 2. The ratio of thermal diffusivity of UO_2 -W CERMET to pure UO_2 pellet

3. 2. Cr-CERMET fuel

Fig. 3a shows the optical microstructure of sintered pellet obtained by sintering in H_2/H_2O mixing gas atmosphere. The liquid phase sintering was occurred during the sintering step. The EPMA result shows that the oxygen-rich and oxygen-poor chromium oxides were precipitated along the grain boundary. The UO_2 –Cr CERMET (fig. 3b) was obtained by reduction of liquid sintered pellet of Fig. 3a. The continuous metal Cr connection was shown in this figure.

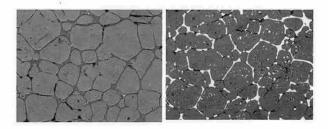


Fig. 3. The microstructure of Cr-oxides containing liquid phase sintered pellet (a) and UO₂-Cr CERMET (b).

4. Conclusion

An innovative fuel consisting of UO₂ grains enveloped with continuous metal channel has been successfully fabricated even with about 6 vol% of W, Mo and Cr.

Acknowledgements

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