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Inert Gas Fusion of a UO_2 pellet with metallic fluxes

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1. Introduction

To extract the retained fission gas, Kr and Xe, in an irradiated nuclear fuel, a fuel segment should be dissolved or annealed using appropriate techniques[1,2,3]. The chemical dissolution procedure is very lengthy, laborious and produces by-product gases, NOx, SOx which should be removed from the extracted gas[1,2]. Consequently, it is necessary to establish a simple, fast and clean dissolution method for UO_2 fuel. Important considerations in the selection of a fusion method are its completeness of decomposition and gas extraction. Inert gas fusion technique with a metallic flux has the decomposition capability of a UO_2 pellet[2]. The technique being presented in this paper for the melting of a UO_2 fuel is an inert gas fusion.

2. Experimental and discussions

2.1 Inert gas fusion

A LECO RH-600 Hydrogen determinator equipped with an EF-500 furnace, an impulse furnace, was employed for an inert gas fusion of a UO_2 segment. The impulse furnace is a resistive–type heating unit consisting of two large water-cooled copper electrodes, an upper electrode and a lower electrode, using a small graphite crucible. The inert gas fusion occurs inside a graphite crucible under a helium atmosphere at elevated temperatures provided by an electrode furnace. A segment of a UO_2 pellet, about 0.1 g, is fused with metal fluxes, tin, iron, nickel, copper, in a graphite crucible heated to a high temperature by an impulse furnace under a helium atmosphere. The fusion current and heating time of the EF-500 impulse furnace was reviewed to establish the optimum melting condition of a UO_2 segment. Two kinds of UO_2 pellets, an unirradiated pressurized water reactor(PWR) UO_2 pellet and a SIMFUEL which was a simulated irradiated UO_2 pellet with a burnup of 60,000MWd/tU, were supplied for a fusion experiment.

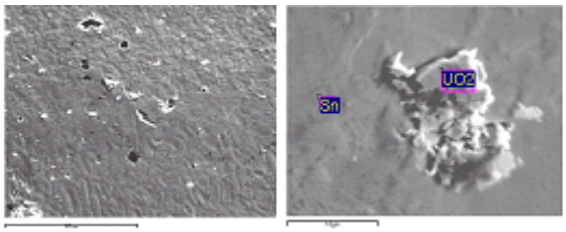
2.2 Analysis of the UO_2 melt

After an inert gas fusion of UO_2 segments with metallic fluxes, the resultant melts were carefully separated from the graphite crucible. Non destructive tests were conducted for the recovered melt with an electron probe micro analyzer(EPMA) and an x-ray diffraction(XRD) spectrometer. After finishing the non destructive tests, the melts were dissolved using acids, and the chemical compositions of them were determined with an inductively coupled plasma-atomic emission spectroscopy(ICP-AES).

2.3 Analysis results of the unirradiated PWR UO_2 melts

About 0.1 g of an unirradiated PWR UO_2 segment was fused with around 1.0 g of a metallic flux, tin, iron, nickel, copper at 750 A and 50 seconds. It was found that the obtained melts of the unirradiated PWR UO_2 segment with tin, iron and nickel fluxes showed a good melting appearance. However, the electrode furnace was contaminated with lots of red dust when the unirradiated PWR UO_2 segment was fused with a copper flux. The melt appearance of the unirradiated PWR UO_2 segment fused with a tin flux was a very shiny silver color and a bead shape and it was clearly separated from the graphite crucible.

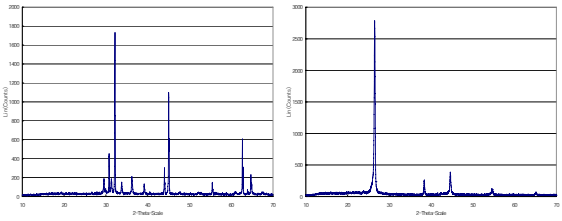
Fig. 1 is the secondary electron image(SEI) of the melt of the unirraidated PWR UO_2 segment fused with tin flux. At a 1000 magnification of an SEI, the melt shows a homogeneous alloy composition and it is difficult to find oxide phase. However, a small uranium oxide phase as melted form in a tin phase, at around 10 μ m in size, is found at a 4000 magnification of a SEI. The mechanisms by which the uranium oxide decomposes in a graphite crucible are not fully understood[4].



x1000 x4000

Fig. 1. Secondary electron images of the melt of the unirradiated PWR UO_2 segment fused with a tin flux at 750A, 50seconds.

The XRD spectra of the melts of the unirradiated PWR UO_2 segment fused with a metallic flux, tin and iron are presented in Fig. 2. The XRD data indicates that the uranium oxide is entirely converted to a carbide(MC2) or oxy-carbide form as a result of the high temperature reactions between the metal fluxes, the UO_2 segment and the graphite crucible.



Sn Fe

Fig. 2. XRD patterns of the melts of the unirraidated PWR UO_2 segments fused with metallic flux at 750A, 50 seconds.

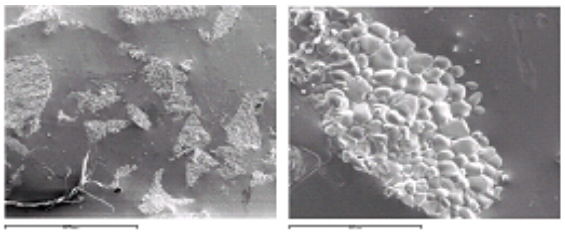
The analysis results of the ICP-AES for the melts of the unirradiated PWR UO_2 segments fused with a metallic flux are presented in Table 1. The weight ratios of U and a metal flux for the obtained melts are similar to those of the input compositions for the fusion.

Table 1. ICP-AES analysis results of the melts of the unirradiated PWR UO_2 segments fused with a metallic flux

Compositions of fusion material		Compositions of UO2 melt (wt%)	
UO2(g)	Fe(g)	U	Fe
0.152	0.6	14.6	68.4
UO2(g)	Ni	U	Ni
0.153	0.654	17.8	74.8
UO2(g)	Cu	U	Cu
0.144	1.4	8.3	86.7
UO2(g)	Sn	U	Sn
0.091	0.981	8.62	85.1

2.4 Analysis results of SIMFUEL UO_2 melts

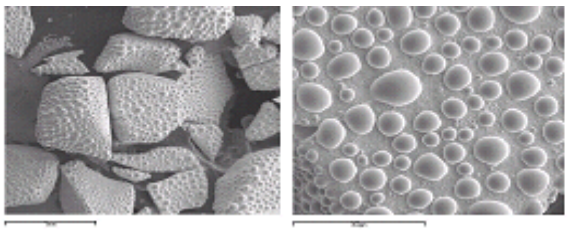
As shown in Fig. 3, many uranium oxide phases are found on the melt of the SIMFUEL UO_2 segment fused with a tin flux at 750A, 50 seconds fusion. It is the same for the melts of the SIMFUEL fused with other metallic fluxes, iron, nickel and copper at the same fusion conditions. Furthermore, some uranium oxide particles were found on the melt of the SIMFUEL segment fused simultaneous using of two kinds of metallic fluxes, nickel and tin, at 850A, 35 seconds. It is predicted that the SIMFUEL UO_2 is more resistant to an inert gas fusion when compared to the unirradiated PWR UO_2.



x100 x1000

Fig. 3. Secondary electron images of the melt of the SIMFUEL segment fused with a tin flux at 750A, 50 seconds.

As presented in Fig. 4., the melt of the SIMFUEL segment fused with a nickel flux at 900A and 85 seconds shows a good alloy appearance and many islands of UNi alloy are found on the surface of it due to a high fusion temperature. The analysis results of the EPMA reveals that its composition is relatively homogeneous and the calculated chemical formula is $M(UNi)C_{3..23}$. It is the same for the melts of the SIMFUEL segments fused with other metallic fluxes, tin, iron, nickel and tin, at the same fusion conditions.



x40 x200

Fig. 4. Secondary electron images of the melt of the SIMFUEL segment fused with nickel, at 900 A, 85 seconds.

However, the melts of the SIMFUEL segments fused with metallic fluxes at 900 A were solidified at an upper position of the graphite crucible due to an extremely high fusion temperature. Therefore, it is required to not use an excessive flux to prevent the danger of an electrode melting with a melt at a very high fusion current.

3. Conclusions

Around 0.1 g of an unirradiated PWR UO_2 segment was fused well with about 1.0 g of a metallic flux, tin, iron, nickel, at 750A and 50 seconds. But the melts of the SIMFUEL segments fused at the same conditions contained many uranium oxide particles. According to the current experiment results, it is required to use a higher current and a longer time, 900A and 85 seconds, for a good fusion of a SIMFUEL UO_2 pellet with a metallic flux. Due to using a very high current, however, the possibility of an electrode solidifying can be a problem.

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