Steam Oxidation Behavior of Cr-coated U_3Si_2 Atomized Particle

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1. Introduction

U_3Si_2, originally developed as a fuel material for research reactors [1], is recently getting attention as an Accident Tolerant Fuel candidate for LWR [2] due to its good thermal conductivity and high uranium density, compared to UO_2.

In the case of U_3Si_2 fuel application for research reactors, manufacturing process was well established and a lot of fuel performance data has been accumulated. However, to utilize the U_3Si_2 for LWR, many technical issues should be addressed because the LWR fuel assembly has quite different structure and the operating environment is much more severe.

The violent reaction with water or high temperature steam in the event of a breach of the cladding is one of the most critical issues for the application of this fuel in LWRs. Volume expansion and pulverization of U_3Si_2 pellet due to the oxidation reaction in the event of leaker scenario leads to cladding bulging, fuel rod failure and escalating accident progress.

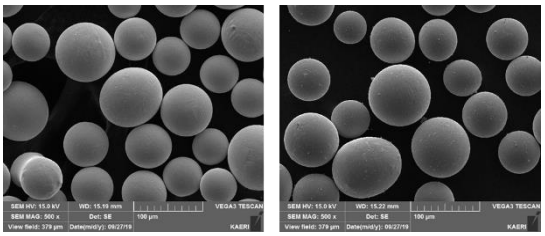
In order to mitigate or minimize the oxidation of U_3Si_2, several approaches have been proposed. Alloying with corrosion-protective metallic elements or surface coating on the U_3Si_2 fuel pellets would be typical examples. Although these efforts have in some cases shown enhancements like increasing the onset oxidation temperature or slowing the oxidation rate, these methods have also found to be insufficient to prevent the massive fragmentation of fuels by oxidation.

One of the mitigation approaches being studied in KAERI is to coat thin Cr layer on the surface of U_3Si_2 spherical particles, which were fabricated through centrifugal atomization process [3]. It is expected that thin Cr layer on the U_3Si_2 particle acts as a barrier for the direct contact between steam and U_3Si_2 thereby prevents the oxidation of U_3Si_2 core. In addition, when the U_3Si_2 pellet is fabricated with coated particles, a honeycomb-shaped Cr-rich wall is located inside the pellet and this multiple Cr-rich layer is expected to efficiently delay oxidation of pellet by steam ingress.

In this study, we are introducing preliminary experimental results on the effect of thin Cr coating layer on the steam oxidation behavior of U_3Si_2 spherical particles. The weight gain under the high temperature steam for coated- and uncoated- particles was measured and compared. The phase change after the oxidation was also investigated by using SEM and XRD analysis.

2. Experimentals

Fig. 1 shows the SEM images of the as-fabricated and Cr-coated U_3Si_2 particles. Spherical U_3Si_2 particles were fabricated by centrifugal atomizing process. The diameters of fuel particles are ranged from 45 μm to 150 μm and the average diameter is about 90 μm. Cr-coated U_3Si_2 particles were prepared though physical vapor deposition. Fig. 2 shows the cross sectional image of coated particles and EDS line scan profile. The thickness of Cr layer was estimated to be about 350 nm. This thickness is equivalent to 2 wt% of Cr addition in U_3Si_2.



(a) (b)

Fig. 1. U_3Si_2 atomized particles. (a) As-fabricated particles (b) Cr-coated particles

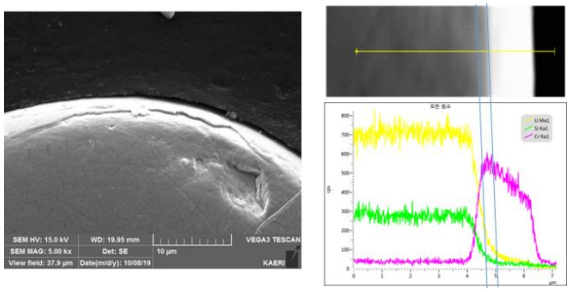


Fig. 2. (a) Cross-sectional image of Cr coated U_3Si_2 (b) EDS line-scan profile across the coating layer

The steam oxidation behavior was investigated at 450, 550°C in the Ar and steam mixed gas atmosphere. The weight gain was measured as a function of time and the steam fraction in the mixed gas was about 50%. After the steam oxidation test at 450°C, the particle shape and phase changes were examined using SEM and XRD.

3. Results

3.1. Comparison of Steam Oxidation

Fig. 3(a) shows the comparison of weight gains as a function of time under steam environment for two sample particles. The test result clearly shows that the oxidation was meaningfully mitigated in the coated U_3Si_2 particles. The weight gain for uncoated sample was ranged from 15% to 18% depending on annealing

temperature and time. However, the weight gain in coated sample was not more than 5 % under the same test conditions.

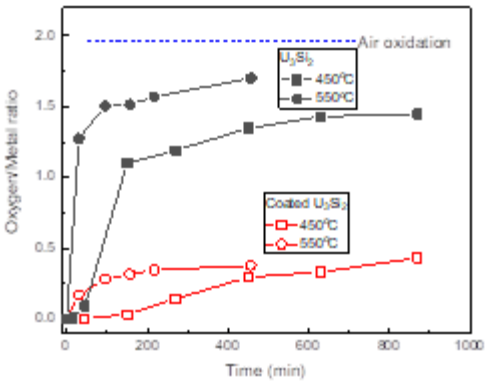
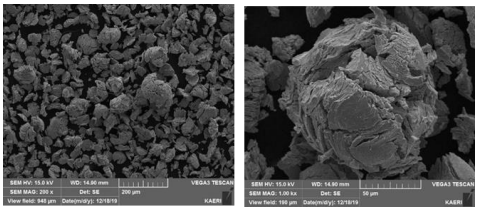


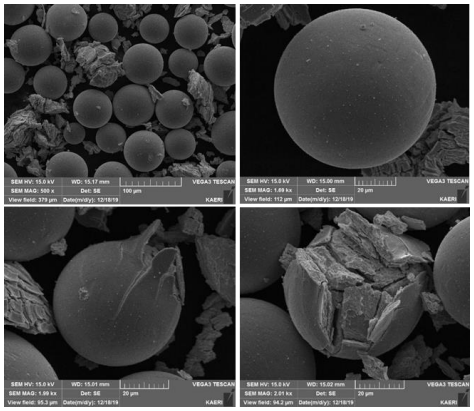
Fig. 3. Comparison of weight gain between sample particles during the annealing in the steam. The weight gain was converted to oxygen to metal ratio.

3.2. Comparison of Particle Shape and Phase Change

Fig. 4 shows the SEM images for the sample particles after the steam oxidation test. Due to the large volume expansion during the oxidation, the uncoated particles lost its spherical shape and fragmented into small powders. However, in coating sample, most of the particles remain intact after the test. In addition, even damaged particles were not fully pulverized and partly retained its spherical shape. This indicates that coating layer, which tightly wraps the particle, provides positive effect to minimize the fragmentation of oxidized particle.



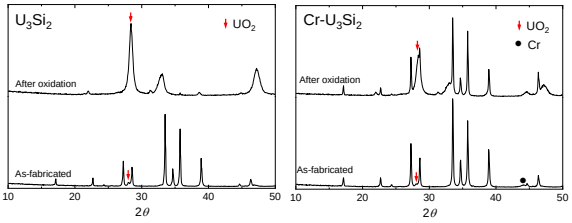
(a)



(b)

Fig. 3. Particle shape change after the oxidation test
(a) Uncoated U_3Si_2 (b) Coated U_3Si_2

Fig. 5 shows the XRD patterns for the sample particles before and after the oxidation test. XRD pattern for as-fabricated uncoated sample reveals typical U_3Si_2 phase diffraction pattern with small amount of UO_2 phase. UO_2 phase was formed due to the surface oxidation during the storage. The U_3Si_2 phase was transformed mostly to $(USi)O_2$ phase after the oxidation. Broad peak width indicated the lattice distortion. This lattice distortion is due to the valence and ionic size differences between U and Si ion. In the case of Cr-coated U_3Si_2, diffraction peak from Cr was additionally observed in the sample before oxidation. The diffraction pattern of coated sample after the oxidation is very different to that of uncoated sample. The sample is a mixture of U_3Si_2 and $(USi)O_2$ phases and there is no noticeable change in the XRD peaks for U_3Si_2 phase compared to as-fabricated coated sample.



(a) (b)

Fig. 5. XRD patterns of samples before and after the oxidation test. (a) Uncoated U_3Si_2 (b) Cr-coated U_3Si_2

4. Summary

The steam oxidation test showed that Cr coating on the surface of U_3Si_2 spherical particle significantly mitigate the weight gain due to the oxidation under the test conditions. The SEM images and XRD patterns for the sample after the oxidation revealed that uncoated U_3Si_2 particles are mostly oxidized to $(USi)O_2$ phase, while only a fraction of coated U_3Si_2 oxidized to $(USi)O_2$ and the main part remain intact.

ACKNOWLEDGEMENT

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REFERENCES

[1] Ann Leenaers, Jared Wight, Sven Van den Berghe, Ho J. Ryu, Jean-François Valery, U-Si based Fuel System, Comprehensive Nuclear Materials 2nd edition, Elsevier Inc., 2020.
[2] J. M. Harp, F. Cappia, L. Capriotti, Postirradiation Examination of the ATF-1 Experiements-2018 status, NTRD-FUEL-2018-000054, 2018.
[3] K.H. Kim, D.B. Lee, C.K. Kin, I.H. Kuk, K.W. PaiK, JNST, 34 (1997) 1127-1132.