Spray Coatings of Al_2O_3 Layer for Passivation of Al_2O_3-SiO_2-based Refractory

Joo-Young Yoon*, Jae-Won Lee, Sang-Chae Jeon, Yung-Zun Cho, Ju-Ho Lee, Kweon-Ho Kang, Do-Youn Lee, Jang-Jin Park, and Kwang-Hoon Cho

Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Republic of Korea

*yjy2746@kaeri.re.kr

1. Introduction

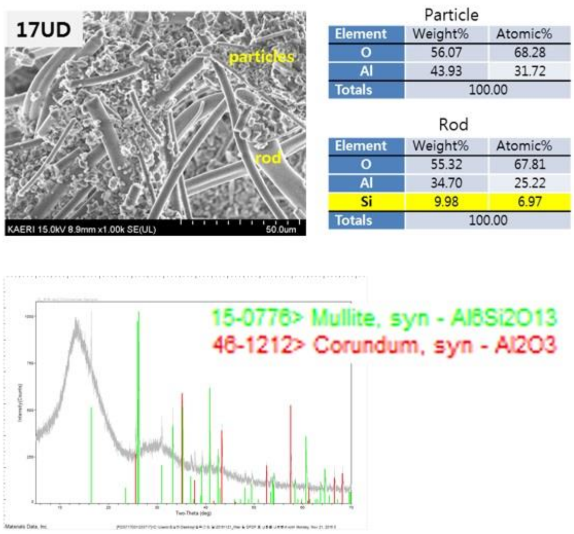
From few decades ago, pyroprocessing has been recognized as a promising dry processing technology to manage the accumulating spent fuel with less proliferation risk than wet processing. It starts with head-end process for the conversion of spent fuel into a feed material for following processes. The spent fuel generally contains various fission products as a result of nuclear reactions during operation. The preparation of feed material in the head-end process may include heat treatment at high temperatures, which leads to the release of volatile fission products from the spent fuel, so called off-gas. The emission of the off-gas from should be minimized due to its toxicity to human body.

Considering great importance of the off-gas treatment in terms of safety, many studies have focused on the capture of the off-gas by filters to reduce its emission rate [1-2]. The off-gas capture system in the head-end process should be connected to a sintering furnace where heat treatment of the spent fuel takes place. For high temperature operation, the sintering furnace is generally equipped with refractories, which contain Al_2O_3, SiO_2, etc. Those can react and form mullite ($Al_6Si_2O_{13}$) phase during heating as shown in Fig. 1. Paradoxically, the mullite phase is a major component of the filters for capturing of Cs. Cs is known to be stabilized by the formation of pollucite ($CsAlSi_2O_6$) phase with the mullite phase. It indicates that the Cs emitted from the spent fuel can be captured at the surface of refractories. This is contrary to the aim of the off-gas system and the contact between the Cs vapor and refractories should be restricted. The development of alternative composition for the refractories would be tremendous time- and effort-consuming work. In view of this, the passivation treatment on the surface of refractories by coating with inert material can be a route. From literature survey, Al_2O_3 and Y_2O_3 with high purity were chosen as possible coating materials that hardly react with Cs.

The present study aims at evaluating the possibility of the passivation by spray coating method. Cube-shaped commercial refractories containing SiO_2 were spray coated by high purity Al_2O_3 and the coated layer was examined before and after heat treatment. To this end, the applicability of this spray coating method was discussed for sintering furnace equipped with off-gas treatment system.

2. Experimental

The commercial refractory (designated as 17UD) in the form of cube (1 cm × 1 cm × 1 cm) and commercial Al_2O_3 spray (Al_2O_3, purity: 96%) were prepared. The Al_2O_3 layer was coated by spraying in air for 3 sec by 3, 4 and 5 shots for the formation of coating layer with different thickness. The sprayed cubic samples were dried in air for 10 min. The samples were then sintered under the following condition: at 1,500°C for 150 min in 4% H_2-Ar atmosphere, ramp/cooling rate was 5°C/min).



Particle

Element	Weight%	Atomic%
O	56.07	68.28
Al	43.93	31.72
Totals	100.00	

Rod

Element	Weight%	Atomic%
O	55.32	67.81
Al	34.70	25.22
Si	9.98	6.97
Totals	100.00	

Fig. 1. Physical properties of 17 UD.

The characterization of samples were carried out by X-ray diffraction (XRD, Bruker, D8 Advanced A25) and scanning electron microscope (SEM, Hitachi, SU-8010) and energy dispersive X-ray spectroscope (EDS, Horiba, X-MAX).

3. Results and Discussion

Fig. 2 shows 17UD samples spray coated with different thickness of Al_2O_3 layers were shown before heat-treatment. After sintering at 1,500^{o}C for 150 min in 4% H_2-Ar, regardless of the thickness of coated layers, all the samples were integrity and maintained homogeneously covered Al_2O_3 layer.

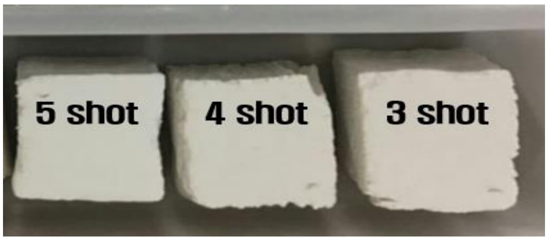
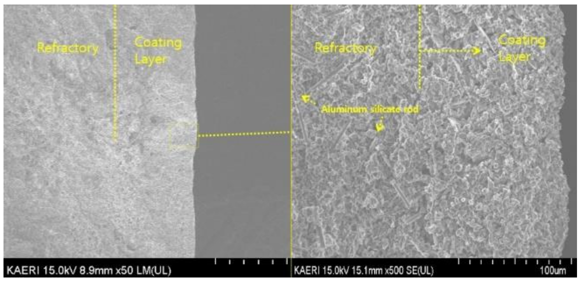


Fig. 2. Spray coated 17UD samples Al_2O_3 after drying.

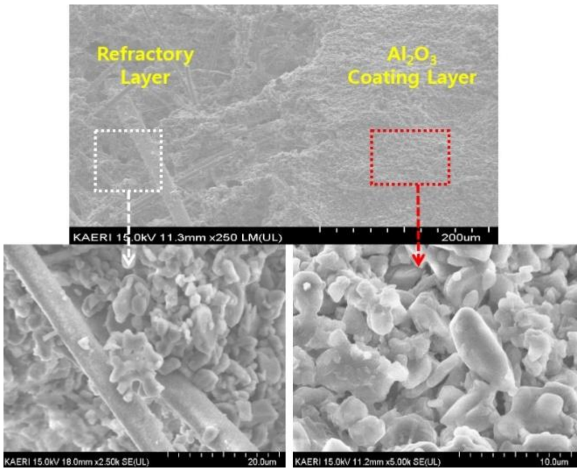
Fig. 3 shows the microstructure of interface between the 17UD substrate and coated Al_2O_3 layer. The newly formed layer was deposited uniformly and made a good adhesion possibly due to sufficient surface roughness at the surface of 17UD before coating. In addition, without polishing and other surface treatment, the interface region can be recognized by the presence of rod shape phases in the 17UD substrate but without it in the coated Al_2O_3 layer. As shown in Fig. 1, the rod shape phase was observed as aluminum silicate by SEM/EDS. The thickness of coated Al_2O_3 that contains only particle shape grains was measured as 100 μm. Considering the refractories are not contact anything during operation, although there is no evidence on the durability of coated layers, this spray coating can be an easy and effective way to deposit the protective layer on the 17UD in terms of physical stability.

4. Conclusions

The spray coating behavior of high purity Al_2O_3 on refractory material containing SiO_2 were investigated. For the formation of protective layer, the adhesion and connectivity of the coated layer were satisfactory. In a physical viewpoint, its durability also should be confirmed. Also, the reactivity of coated layers with



(a) Image of coating area



(b) Image of Al_2O_3 coating and refractory layer

Fig. 3. SEM image of Al_2O_3 coated layer and 17UD.

Cs vapor needs to be examined for the application to in the sintering furnace equipped with the off-gas treatment system. The chemical reaction of the coated samples with Cs vapor will be discussed in future study.

REFERENCES

[1] Park, J.J., Shin, J.M., Yang, J.H., et al., 2015. Fabrication and performance of fly ash granule filter for trapping gaseous cesium. Nukleonika 60, 565-570.

[2] Yang, J.H., Yoon, J.Y., Lee, J.H., Cho, Y.Z., 2017. A kaolinite-based filter to capture gaseous cesium compounds in off-gas released during the pyroprocessing head-end process. Annals of Nuclear Energy 103, 29-35.