Study on Manufacturing Technology of MnO-Al_2O_3 Doped UO_2 Pellets for Remedy of PCI Failure

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ABSTRACT

For the last 30 years, there have been research projects to develop additives in UO_2 for improvement of reliability and efficiency in operation. As a result, fuel vendors have introduced their own fuel additives and therefore, reached licensing or commercialization stage. Particularly, further research relevant to additives in UO_2 as a candidate of accident tolerant fuels has been carried out. Likewise, from the early 2000s in South Korea, research about doped fuel was performed and MnO-Al_2O_3 system was chosen as one of PCI failure retention additives. Conclusively, as a fuel vendor in Korea, KEPCO Nuclear Fuel (KNF) has conducted pre-industrialization manufacturing runs by using factory equipment in order to confirm the compatibility with commercial application after mockup lab test. Material properties have been measured, which are grain size, sintered, and resintered density to assess compatibility with commercialization and predict the ability of the PCI performance improvement with MnO-Al_2O_3. Additionally, the fundamental mechanism governing grain growth in UO_2 doping with MnO-Al_2O_3 has been analyzed with SEM image.

KEYWORDS*: MnO-Al_2O_3 system, PCI remedy, Commercial compatibility, Liquid phase*

I. INTRODUCTION

For the last 30 years, many research based on small quantity doped UO_2 has been conducted to lead to higher operation margin, especially, in pellet-cladding interaction (PCI) and as a result, several achievements have been introduced, which, generally, enhance microstructure evolution contributing to enlarged grains. AREVA, GNF and WEC atom have developed chromia[1, 2], aluminosilicate[3] and aluminochromia[4] system with this concept, respectively, and AREVA continues to work on its additive through enhanced accident tolerant fuel (EATF) program[5] by choosing it as a candidate.

Typically, the benefit regarding PCI performance behaviors with doped fuel is measured by ramp testing after steady state operation and its specific improvement mainly features enhanced creep resistance and plasticity. The two main improvements are, after a certain amount of fuel burnup, to suppress the applied stress between the fuel and the cladding by induced transient swelling and to retard transient fission gas release to open voidage [1, 5]. Increased fuel safety margin by doping in UO_2 pellet for retention of PCI failures during power ramping, has been achieved by accelerating grain growth rate.

Likewise, in keeping abreast of the world from the early 2000s in South Korea, many efforts have been exerted to develop variable ceramic doping system and sintering process. Consequently, not only MnO-Al_2O_3 system[6] but also stepwise sintering process[7] have been adopted as method for improved prohibition against PCI failure. Through the previous studies, it was introduced that an increased MnO-Al_2O_3 doped fuel pellet deformation in comparison to standard fuel has been measured and the oxygen potential change during sintering process has improved additive-effectiveness by lowing dopant addition and simultaneously, maximizing microstructure evolution. Between both developments, the new ceramic additive system which demonstrated compliance with ex-DC UO_2 in KNF, was implemented in the pre-industrialization manufacturing run and material properties were measured. Due to lack of knowledge about nature in the new additive system, before applying to the run, mockup lab test was performed to assure validity of homogeneous distributed dopant in ex-DC UO_2 powder of which low ability mixing with dopant not triggering grain enlargement.

Conclusively, this study is about a procedure to identify compatibility of MnO-Al_2O_3 system with fuel manufacturing process in KNF before commercial manufacturing. After mockup lab test using dozens gram of UO_2 powder, pre-run test using 5 kilogram was initiated. In order to assess impact of the system, grain size, sintered, and resintered density were measured and furthermore, using SEM, the grain growth mechanism was examined.

II. EXPERIMENTAL

Prior to demo run in process of industrialization manufacturing, MnO-Al_2O_3 doped UO_2 has been evaluated in laboratory to establish its optimum concentration which is applied to demo. Material properties of doped pellets were assessed, which are grain size, sintered, and resintered density estimated by the linear intercept and the water immersion method, respectively. Additionally, grain growth mechanism has been roughly dealt with from SEM image of 10 wt. % the additive incorporated pellet.

II.A. Mockup lab test

To establish the optimum concentration, each 40 gram of ex-DC UO_2 in KNF has been mixed with the additive in the range of 1000 to 3000 ppm using Turbula mixer^{TM} for 4 hours and then, compacted and sintered. There are 3 batches and 3 lots assigned of mixing and sintering-wise, respectively. Every sintering has been carried out for 14 hours around 1730 ~ 1770 ^oC using a commercial production furnace filled with hydrogen gas in factory. The each mixture of lot and sintering of batch are listed in TABLE I. A green body incorporating 10 wt. % of it was sintered for 2.5 hours in a furnace in laboratory.

TABLE I. Explanation of batch for UO_2 powder with MnO-Al_2O_3 and lot for sintering pellets

*Batch / **Lot	1	2	3
1	O (1000, 1500, 2000 ppm)	X	X
2	O (1000, 1500, 2000 ppm)	O (1000, 1500, 2000, 3000 ppm)	X
3	X	X	O (1000, 1500, 2000, 2300, 3000 ppm)

*Batch: Independent sintering performed in the same furnace at times
**Lot: Independently mixed powders in each bottle

II.B. Demo run test

The optimum concentration, 2300 ppm, has been established and introduced to demo run test consisting of powder and sintering process as shown in Fig. 1. Equipment used in the powder process has high compliance with that of factory and particularly, the Nauta-typed mixer featuring a spiral screw in a silo chamber is precisely equivalent with practical one. About 5 kg powder has been mixed and 60 pellets have been sintered and then sintered density was measured. From 16 out of the pellets, grain size was measured. A 24 hours thermal stability test at 1700 ^oC has been also measured, which is for resintered density.

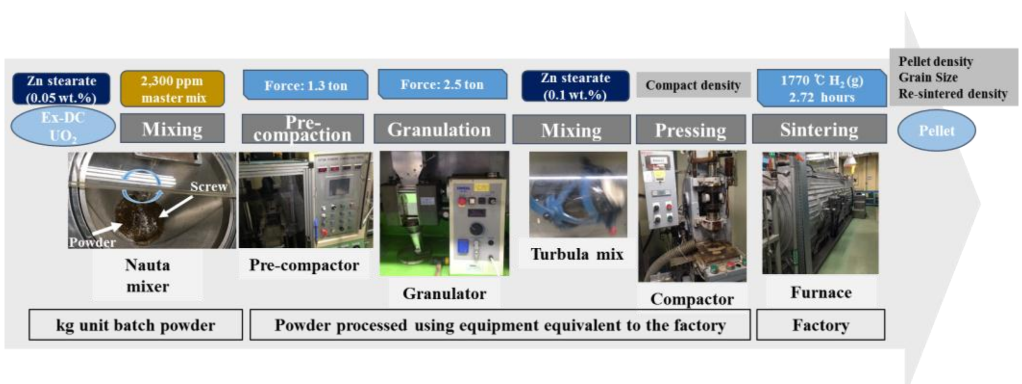

Fig. 1. Demo run in consequent commercial process for producing pellets

III. RESULTS AND DISCUSSION

III.A. Mockup lab test

Grain size increment with batches and lots as shown in Fig. 2 features that the property increases linearly with the concentration. Specifically, morphological changes of grains is shown in Fig. 2 with the increasing content, which gradually expand. Based on the result, an additive concentration over 2300 ppm leading to 40.1 µm in 2D grain size should be implemented to meet the qualifications of the proven UO_2 resistant to PCI failure by incorporation of additive, of which criteria in relevant to 2D grain size is usually over 40 µm. The density of sintered pellets has estimated to be over 98 % and this distribution is acceptable in production standard.

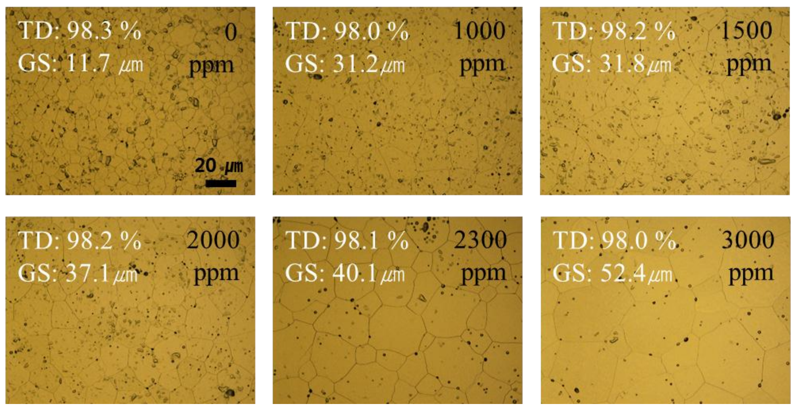

Fig. 2. Grain size increment with increasing the additive content in each batch and lot

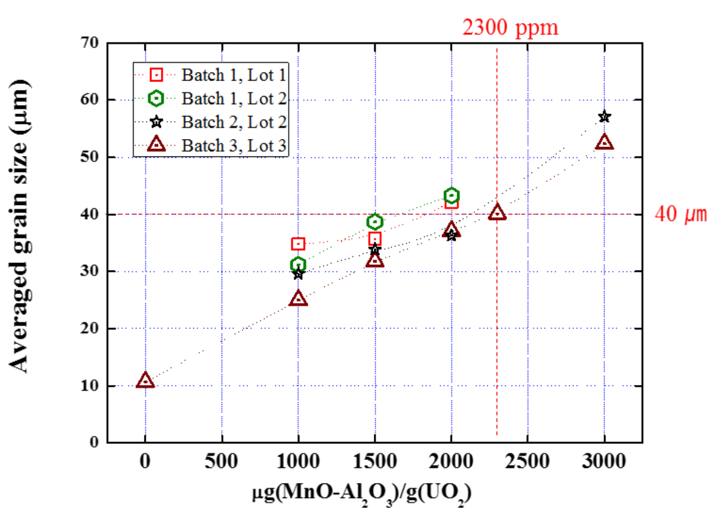

Fig. 3. Grain size in pellet with $MnO\text{-}Al_2O_3$ content

In order to analyze grain growth behavior, microstructure of the excessively incorporated pellet was observed with SEM as shown in Fig. 4.

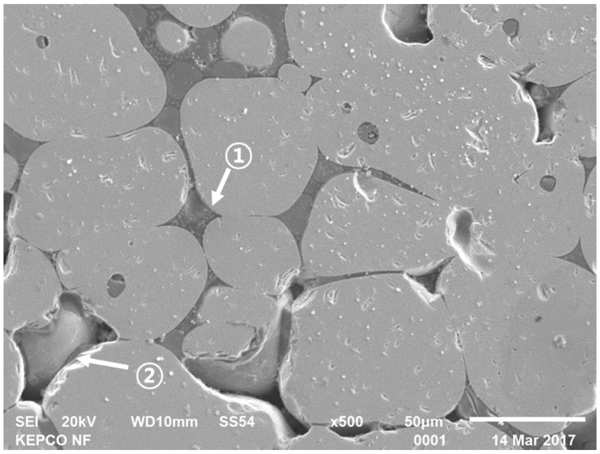

Fig. 4. Round-shaped grains covered by second phase (①) and volatilized area (②) in 10 wt. % MnO-Al_2O_3 added UO_2

It is shown in Fig. 4 that the UO_2 grains are round-shaped and are fully surrounded by second phase which forms a low dihedral angle. That is, MnO and Al_2O_3 formed a glass phase during its coexistence at high temperature and it is assumed that the phase promoted grain growth by wetting grains which resulted in rapid mass transport through the liquid phase. In addition to the existence of the second phase, there is another feature of volatilized mark in the microstructure, which seems to be filled with the additives and then it has diffused out during sintering due to the high vapor pressure.

III.B. Demo run test

Fig. 5 shows images of UO_2 microstructure incorporated with 2300 ppm of MnO-Al_2O_3 which was manufactured through the mockup and the demo run test. From this consequence, it is proven that manufacturing on the scale of kg batch through industrialized equipment is compatible to the result of laboratory work. Namely, not like the previous work done with Cr_2O_3[8, 9], the Nauta mix method seems to be available to mix the additive and ex-DC UO_2 homogenously, even in mass production. Because the additive ability to help the acceleration is not vulnerable to manufacturing process not as much as influencing microstructure, it is concluded that MnO-Al_2O_3 seems to be a prominent candidate system to be introduced to KNF fuel production system to suggest solution to the PCI failure. There is positive perspective about PCI retention ability from the microstructure having large grains averaged in over 40 μm of MnO-Al_2O_3 doped UO_2. Additional tests were performed which are sintered and resinterd density and both meet the standard UO_2's criteria.

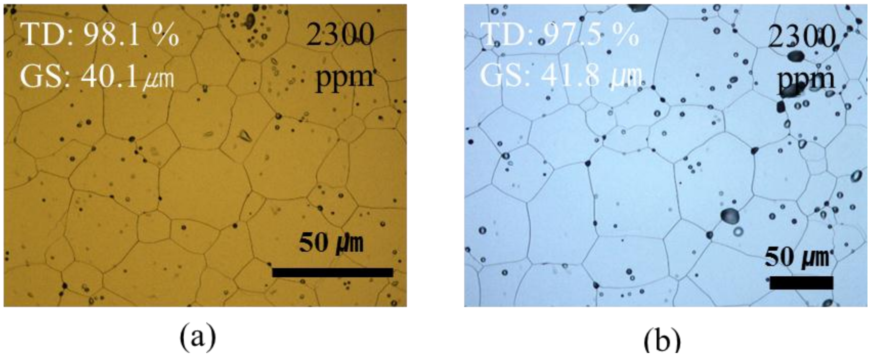

Fig. 5.Microstructure of 2300 ppm of MnO-Al_2O_3 added UO_2 manufacture through (a) Mockup and (b) Demo run test.

Currently, high temperature creep test is being conducted and the incorporated UO_2's creep rate is identified as superior to standard UO_2 several times.

IV. CONCLUSIONS

In KNF, evaluation of suitability of MnO-Al_2O_3 system in practical process has been assessed and tests in laboratory and factory were consecutively conducted. As a result of lab. test, the optimal concentration of MnO-Al_2O_3 was established to enable grains range over 40 μm in average, so did it in demo run, which fulfils requirement to be considered as UO_2 featuring PCI failure resistance. Other material properties as sintered and resintered density were measured also and all out of pile test meet the quality requirement in standard UO_2. Therefore, it brings to conclusion that MnO-Al_2O_3 system is eligible to be applied to industrialization manufacturing work in KNF. In order to clarify the fast grain growth, a proposal about its mechanism has been established through additionally performed analysis using SEM, which the presence of liquid phase consisting of MnO and Al_2O_3 at high temperature, promotes mass transport.

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