Experiences in MOX fuel fabrication at the PFPF for the fast reactor

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Abstract

Since 1988, the Japan Atomic Energy Agency has been developing mixed plutonium-uranium oxide (MOX) fuel fabrication technologies at the Plutonium Fuel Production Facility (PFPF) to provide fuel on a large-scale for the experimental fast reactor JOYO and the prototype fast reactor MONJU and it has been fabricating MOX fuel assemblies for these reactors, too. Low-density pellets were adopted as the MONJU fuel. For the low-density pellet fabrication on a large-scale, various challenges were encountered such as thermal degradation of additives and inhomogeneous dispersion of pore-former in MOX granules. In order to resolve these challenges, countermeasures were considered such as using a new pore-former with a high softening temperature and using an improved granulation method for MOX powder. Also, test using the new pelletizing method (die wall lubrication pelletizing) for low-decontaminated TRU fuel fabrication was carried out at the PFPF. No large pores and no cracks in the obtained sintered pellets were observed. Finally, the standard deviation of pellet density in the test was comparable with that in past fabrications of JOYO pellets.

Key Words: Large-scale MOX fuel fabrication, low-density pellet, decay heat, particle size controlled recycled powder, die wall lubrication pelletizing

1. Introduction

The Japan Atomic Energy Agency (JAEA) has been working to advance fabrication technology of mixed plutonium-uranium oxide (MOX) fuel pellets for fast reactors (FRs). The total amount of MOX fuel fabricated for the experimental fast reactor JOYO reached 301 fuel assemblies and that for the prototype fast breeder reactor MONJU reached 366 fuel assemblies at the Plutonium Fuel Production Facility (PFPF) as of the end of 2016. The PFPF was constructed in 1988 under the concepts of automation and mass production, and its annual fabrication capacity was 5 tons of MOX per year. Various experiences have been gained and improvements in the fabrication process have been conducted to achieve efficient MOX fuel fabrication on a large-scale.

A significant extension of MOX fuel pin life is critically important for FRs to economically compete with light water reactors. When MOX fuel is irradiated to a high burnup beyond 100 GWd/t, the fission product gas release fraction and swelling rate become large, affecting the integrity and consequently, the fuel pin life. The use of low-density pellets, which was adopted for the fuel of MONJU, offered a powerful solution to these problems. These pellets can increase the free volume and improve the storage capacity of fission gas and mitigate the tendency toward swelling in the fuel matrix.

However, for the low-density pellet fabrication on a large-scale, various challenges were encountered such as thermal degradation by decay heat of organic compounds used as pore-formers and the large standard deviation of pellet density due to inhomogeneous dispersion of pore-formers in the granulated MOX powder.

In this paper, accumulated MOX fuel fabrication technologies such as countermeasures for the challenges mentioned above and recent R&D activity for low-decontaminated TRU fuel fabrication such as use of a new pelletizing method, or use of die wall lubrication pelletizing, are presented.

2. Experiences in FR MOX fuel pellet fabrication at the PFPF

2.1 Fabrication process of MOX fuel pellets for FRs

MOX fuel fabrication for JOYO at the PFPF was started in 1988 and that for MONJU was started in 1989. JOYO fuel consists of solid type high-density pellets (94%TD, **T**heoretical **D**ensity). On the other hand, MONJU fuel consists of solid type low-density pellets (85%TD).

Figure 1 shows the flow sheet of the MOX fuel fabrication process at the PFPF; this is called the JAEA process and it provides fuel for both JOYO and MONJU. Two kinds of plutonium materials, either PuO_2 powder prepared by oxalate precipitation or MH-MOX powder [1], can be used in the process. Three kinds of feed powders, UO_2 prepared by the ADU process, PuO_2 or MH-MOX powder, and dry recycled powder, are weighed to get the plutonium content specified by the fuel specifications in the mixed raw powder. Typical Pu content in the MONJU fuel pellets is about 30%. The feed powders are ball milled to get a homogeneous distribution of plutonium in the sintered MOX pellets. Figure 2(a) shows the ball mill used at the PFPF. Around 40 kg-MOX powder can be charged in this ball mill. Two distinct features of the ball milling step are to use a steel mill pot with silicone rubber inner lining and to use alumina (Al_2O_3) balls. Three different size balls are used to improve the crushing efficiency as shown in Figure 2(b). When a steel mill pot without the inner lining and steel balls were used in the past, some problems occurred such as that the MOX powder was excessively crushed, causing a large amount of the powder to adhere to the pot, and capping and cracking of the pellets to occur. In order to solve these problems, relatively small specific gravity alumina balls were adopted to facilitate the control of powder characteristics, and the silicone rubber lining was adopted to suppress powder adhesion. After blending additives such as zinc stearate as binder and pore-former with the milled powder, the blended powder is pressed into tablets at around 200 MPa. Next, the tablets are crushed into granules and sieved. Sizes of the obtained granules range from 250 to 900 μm. This granulation is needed because the milled MOX powder is not easily filled into the small die (die diameter size: about 5 – 6 mm) needed for fabrication of FR fuel pellets. Obtained granules are mixed with zinc stearate as lubricant, then pressed into green pellets at around 500MPa. These green pellets are sintered at around 1700 °C for 4 h in an atmosphere of Ar-5%H_2 mixed gas after dewaxing at around 800 °C for 4 h also in an atmosphere of Ar-5%H_2 mixed gas to remove additives from pellets. Typical ceramographs of a transverse section of two sintered pellets are shown in Figure 3. Large pores (around 100 μm in diameter) made by the pore-former are observed in the ceramographs and the number of the pores made by the pore-former in the MONJU pellet is larger than that in the JOYO pellet. Then, sintered pellets are transferred to the finishing and inspection equipment. In the equipment, the sintered pellets are surface-ground and their dimensions and density are measured and visually inspected. The outer diameter of the pellets becomes uniform as they pass through the grinding stones. Also, laser beams measure the overall length and outer dimensions of the pellets, and an electric balance weighs them, followed by an automatic calculation of their density. All the fabrication steps are dry processes to allow an increased mass for a batch production because wet plutonium has a smaller mass for criticality at than dry plutonium has.

Normally, the yield of MOX pellet fabrication for JOYO is from 0.8 to 0.9. However, the yield for the first loaded MOX fuel fabrication for MONJU was around 0.55. This low pellet fabrication yield was mainly due to thermal degradation of pore-former and inhomogeneous dispersion of pore-former in granulated MOX powder. Countermeasures for these challenges are described in the next three sub-sections.

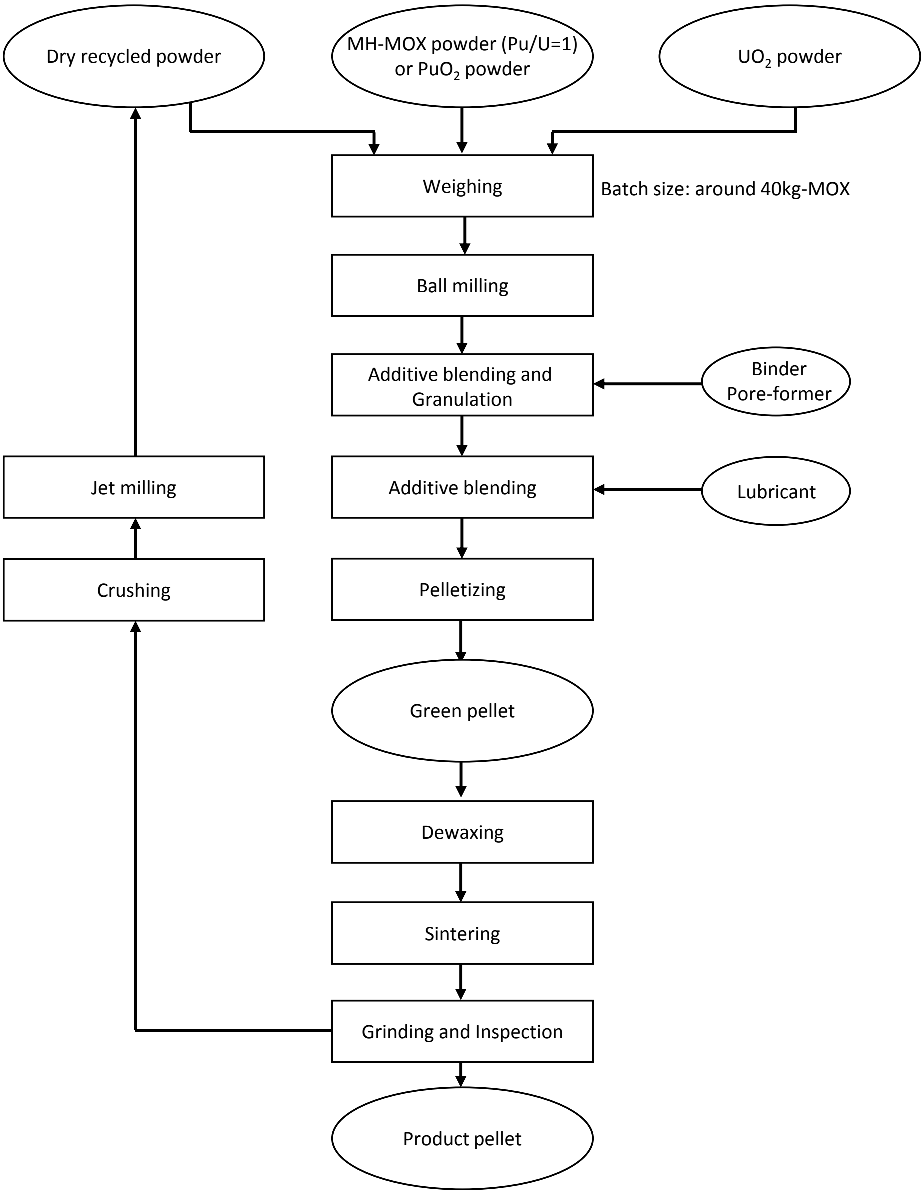
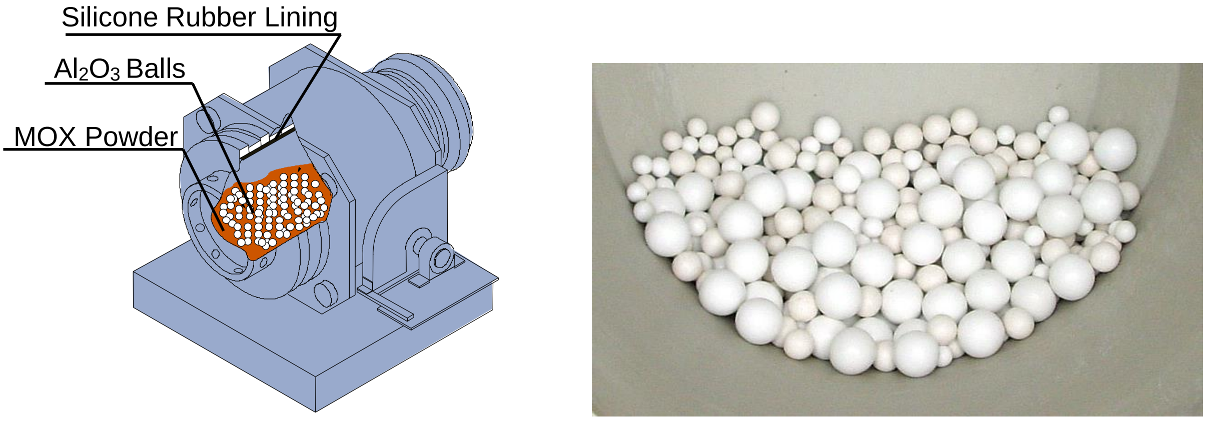
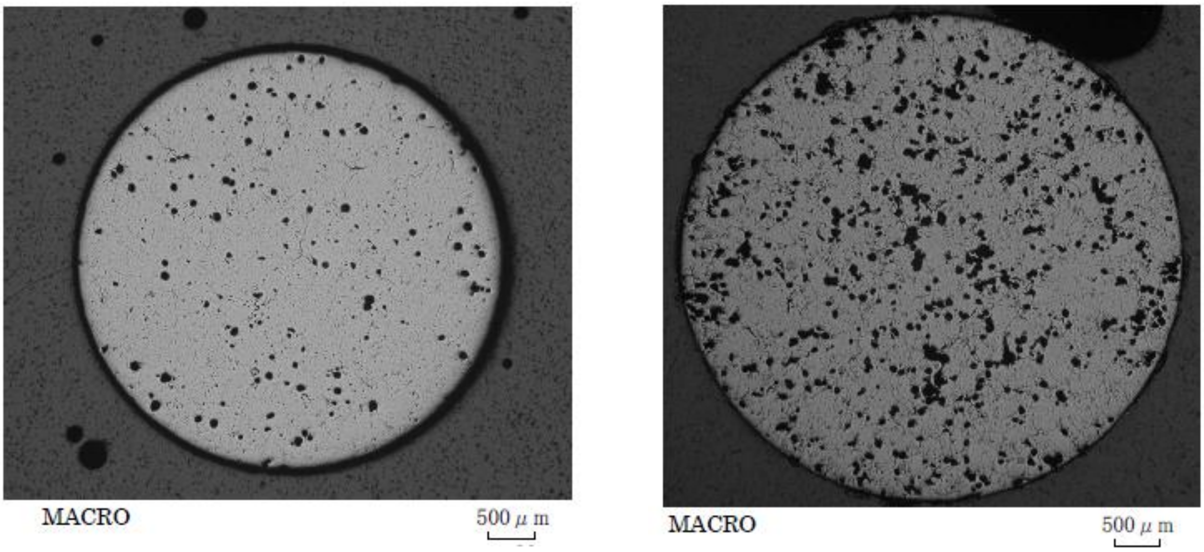


Figure 1. FR pellet fabrication process used at the PFPF



(a) Cut-away view of the ball milling machine *(b) Three different size balls*

Figure 2. Ball milling machine used at the PFPF



(a) JOYO pellet

(Pore-former content: 0.45%)

(b) MONJU pellet

(Pore-former content: 1.8%)

Figure 3. Ceramographs of a transverse section of two sintered pellets

2.2. Thermal degradation of organic compounds

MOX powder generates heat by decay of Pu and Am. Although the heating value of MOX powder is not so high, around 15 W/kg-Pu, the powder temperature at the center of the powder transfer container or the powder hopper of the granulation equipment is around 90 °C in a large-scale fuel production line such as at the PFPF because MOX powder has low thermal conductivity [2]. Figure 4 shows the powder transfer container used in the PFPF and its capacity is around 20 kg-MOX. Pellet production batch size is usually around 40 kg-MOX, so one production batch will be contained in two powder transfer containers.

Organic additives, especially pore-formers, added to MOX powder in the fuel fabrication process easily degrade under such a high temperature condition. The degradation of pore-formers results in quality loss of sintered pellets such as a large standard deviation of density. This problem is more serious for low-density pellet fabrication than for high-density pellet fabrication because a larger amount of pore-former is added to MOX powder for the low-density pellet fabrication compared to the amount for the high-density one. Typical pore-former contents are below 0.5% for JOYO and around 2.0% for MONJU fuel pellet fabrications.

The heat degradation problem of pore-former was first encountered at the PFPF during MOX fuel fabrication for the first fuel loading of MONJU; this production was done from 1989 to 1993. Initially, *K3* (glycerine trihydroxystearate) was used as a pore-former because *K3* had shown good pore-forming performance, had made good spherical pore shapes and had given a homogeneous pore distribution in sintered pellets in a small-scale pre-fabrication test (7.5 kg-UO_2 test batch). However, for about a 40 kg-MOX production batch, the MONJU fuel pellets fabricated using *K3* had a large standard deviation and inhomogeneously dispersed pores. Figure 5 shows a ceramograph of a transverse section of a sintered pellet fabricated using *K3*. Inhomogeneously dispersed pores and lack of good spherical shapes of the pores are observed in the ceramograph. These behaviors are due to the low softening point of *K3*, which is around 85 °C. So, other organic compounds were surveyed as pore-formers which have high heat resistance and MOX pellet fabrication tests using these organic compounds were carried out. Details of the tests were reported by Asakura et al. [3]. From the tests results, *Avicel* (crystal cellulose) was selected as the pore-former with the best performance. *Avicel* maintained the spherical shape of the pores formed at temperatures up to 150 °C. Also, the timing for the pore-former addition was switched from after granulation to before it to mitigate the problem of inhomogeneous dispersion of pores. In this way, the pore-former is strongly trapped in the MOX granules. Figure 3 (b) shows the ceramograph of a transverse section of a sintered pellet fabricated for MONJU using *Avicel* addition before granulation. Homogeneously dispersed pores and spherical shape pores are observed in the ceramograph.

Figure 6 shows the standard deviation of density of sintered pellets fabricated using *K3* and *Avicel* for the first loading of the MONJU inner core fuel pellets. The standard deviation within a production batch of sintered pellets fabricated using *Avicel* addition before granulation is smaller than that of sintered pellets fabricated using *K3* addition after granulation. Yield of MOX pellet fabrication is improved from about 0.3 to 0.6 by switching the organic additive pore-former from *K3* to *Avicel* and changing the timing of the pore-former addition.

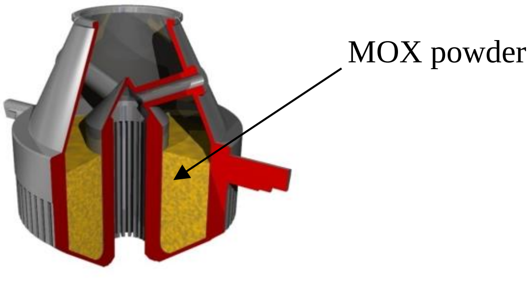


Figure 4. Powder transfer container (Capacity: about 20 kg-MOX)

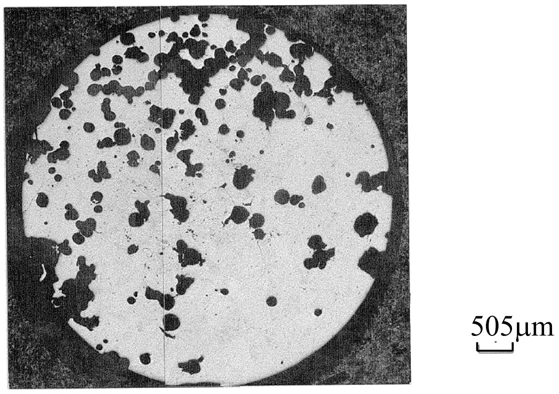


Figure 5. Ceramograph of a transverse section of a sintered pellet fabricated using K3 as pore-former

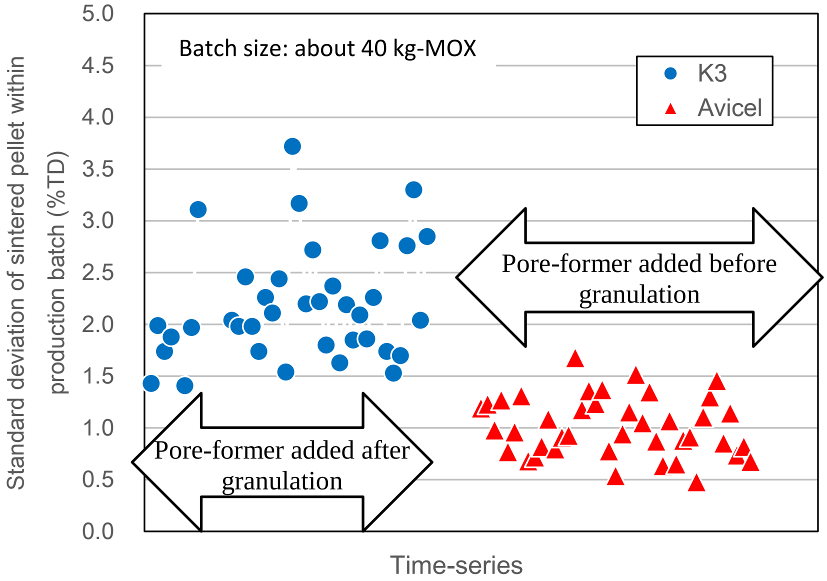


Figure 6. Standard deviation of sintered pellets density within the production batch for fabrication of the first loading of MONJU inner core fuel pellets

2.3. Improvement of additive blending and granulation step

As mentioned in sub-section 2.2, organic additive performance is strongly affected by decay heat of MOX powder in large-scale production. Binder (zinc stearate) and pore-former (*Avicel*) are added to MOX powder in the additive blending and granulation step and this step is time consuming; it takes four work shifts to process one batch of 40 kg-MOX powder. Therefore it can be assumed that organic additive performance is easily degraded by decay heat in this step. In order to reduce the thermal degradation of additives, the production batch size is halved (20kg-MOX) in this step. Figure 7 shows the standard deviation of sintered pellet density. Half-size batch handling in the step is clearly an effective way to reduce the standard deviation of the sintered pellet density. This effect on the standard deviation might be due to mitigation of the decay heat effect on organic additives. The half-size batch for MOX powder production is returned to its full size in the next step, lubricant blending, to avoid loss of production efficiency.

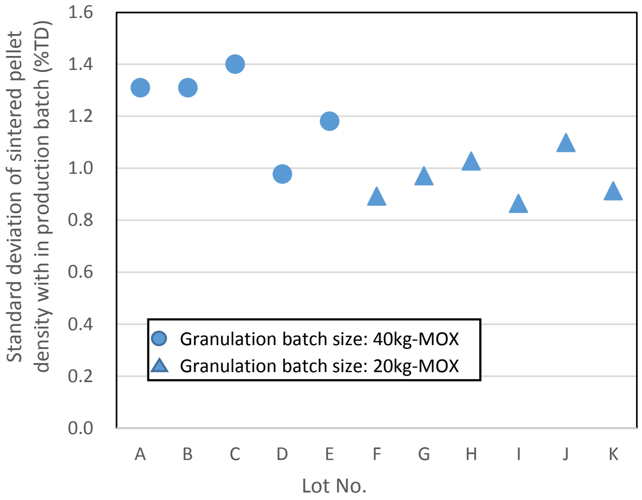


Figure 7. Standard deviation of sintered pellet density within production batches

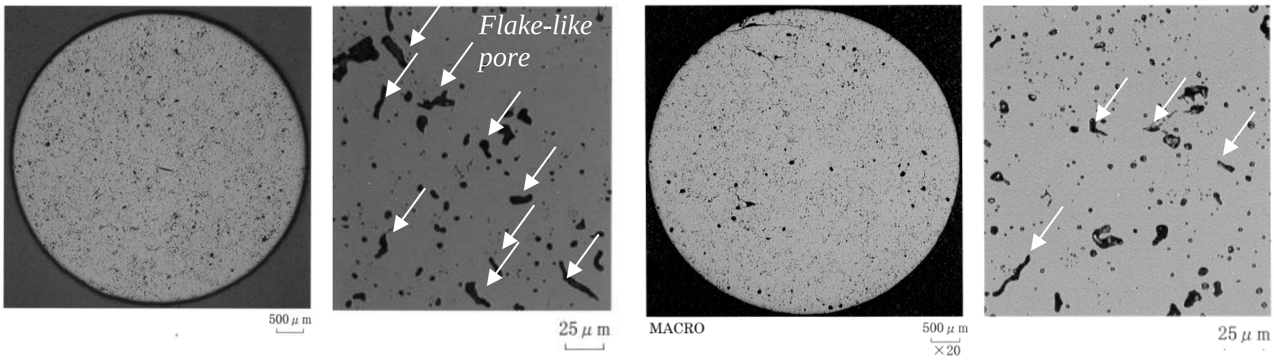
2.4. Further improvement of standard deviation of pellet density

As mentioned in sub-section 2.2, the standard deviation of MONJU pellet density within a production batch was improved by introduction of *Avicel* as pore-former. However, the standard deviation was still higher than that of the JOYO pellet density because of the large difference in bulk density between pore-former (below 1 g/cm^3) and ball milled MOX powder (about 3 g/cm^3). In order to reduce the standard deviation of MONJU pellet density, some tests were conducted on a new pellet density control method using particle size tuned recycled powder obtained by pellet crushing and jet mill operation conditions, or on minimizing the pore-former addition content [4]. It was observed that sintered pellet density was decreased with increased particle size of recycled powder used as feed powders in the tests. Typical ceramographs of sintered pellets produced at the PFPF using different particle size recycled powders are shown in Figure 8. Larger size pores are observed in the ceramograph of Figure 8 (a) for which larger particle size recycled powder was added. Murakami et al. [4] reported on the pellet density reduction effect by recycled powder addition and noted that activity of the recycled powder was lower than the rest of the feed powders, or MH-MOX and UO_2 powder, because the recycled powder was obtained by crushing and jet milling once-sintered pellets. Hence, it is difficult to shrink the interstices in the first stage of sintering, and interstices above a certain size are retained as stable pores in the sintered pellet. Also, it seems that the number of pores produced by addition of the large particle size recycled powder is larger than that produced by addition of the small particle size recycled powder. As Murakami et al. [4] reported, the shape of the pores produced by addition of the recycled powder was not spherical but flake-like. The flake-like pores observed in Figure 8 would be formed by recycled powder addition. The fraction of the number of pores produced by the recycled powder addition was analysed using a shape factor to evaluate the pellet density decrease effect by recycled powder addition. The shape factor of a pore was determined by the following equation.

$$\text{Shape factor} = \frac{4\pi \times area}{(perimeter)^2} \qquad (1)$$

The shape factor of a perfectly spherical pore is 1. And the shape factor decreases as the pore shape deviates from perfectly spherical. It was reported that the flake-like pores had shape factors less than 0.5 [5]. So, the effect on pellet density reduction of recycled powder could be roughly evaluated from the estimated shape factor as being less than 0.5. Figure 9 shows the relationship between of number of shape factors of pores in a sintered pellet being less than

0.5 to the number of all pores and the added recycled powder content of the sintered pellet. The number of flake-like pores, or in other words, the effect of decrease of pellet density, is increased with increasing recycled powder addition and increasing recycled powder particle size. From this result, a larger added content of larger particle size recycled powder is better for fabricating low-density pellets because the amount of organic compound addition can be reduced. Murakami et al. [4] also reported that a high added content of large particle size recycled powder (<250μm), about 40%, led to large densification in a 24-h re-sintering test. In their report, the recommended low-density (85%TD) pellet fabrication condition was added content of 25% of the large particle size recycled powder and around 1.4% of pore-former, and these conditions could provide less than 1%TD of densification for the 24-h re-sintering. Average standard deviation of pellet density within a production batch was stabilized around 0.7 under the recommended fabrication conditions for MONJU fuel at the PFPF, this value is fairly small compared to that of pellets fabricated for the first loading of MONJU.



(a) Recycled powder particle size: <250 μm

Added recycled powder content: 20%

Pore-former content: 0%

(b) Recycled powder particle size: <100 μm

Added recycled powder content: 25%

Pore-former content: 0%

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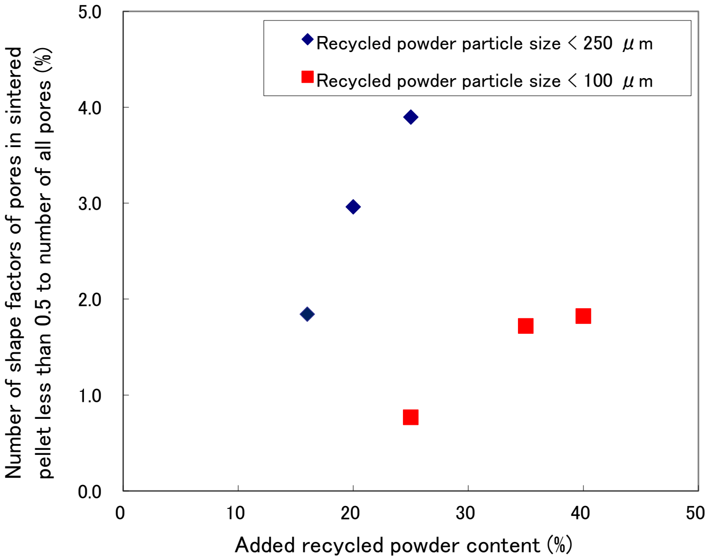

Figure 9. Relationship between the number of shape factors of pores in a sintered pellet being less than 0.5 to the number of all pores and added recycled powder content of the sintered pellet

3. Recent R&D activity at the PFPF: Large-scale die wall lubrication pelletizing test

Die wall lubrication pelletizing for MOX fuel fabrication has been studied in a cold test and a small-scale hot test for low-decontaminated TRU fuel fabrication [6]. By adopting the die wall lubrication pelletizing in the high-density MOX fuel pellet fabrication process, the lubricant mixing and de-waxing step could be removed from the fabrication process. The die wall lubrication method is explained in Figure 10 by comparison to the conventional method.

A large-scale MOX test was conducted to confirm productivity and pellet quality of a new route for pellet fabrication, in which die wall lubrication is applied instead of lubricant mixing as in the conventional powder mixing process. In this test, about 38 kg-MOX of annular type high-density pellets was fabricated using die wall lubrication pelletizing equipment. The pressing equipment can press MOX powder into two annular green pellets simultaneously. Figure 11 shows a photograph and a ceramograph of an obtained pellet. No large pores and no cracks are observed. Content of impurities in the obtained pellet meets specifications of JOYO and MONJU fuels. Standard deviation of pellet density in the test is comparable with that in past fabrication of JOYO pellets (solid type high-density pellets). Also, the pelletizing rate under automated operation is 7.5 cycle/min. This pelletizing rate is acceptable for a large-scale MOX pellet fabrication line. However, the large-scale annular pelleting test using the die wall lubrication method was conducted for just one batch. So, further large-scale tests will be needed to confirm product pellet quality and to find the best pelletizing conditions.

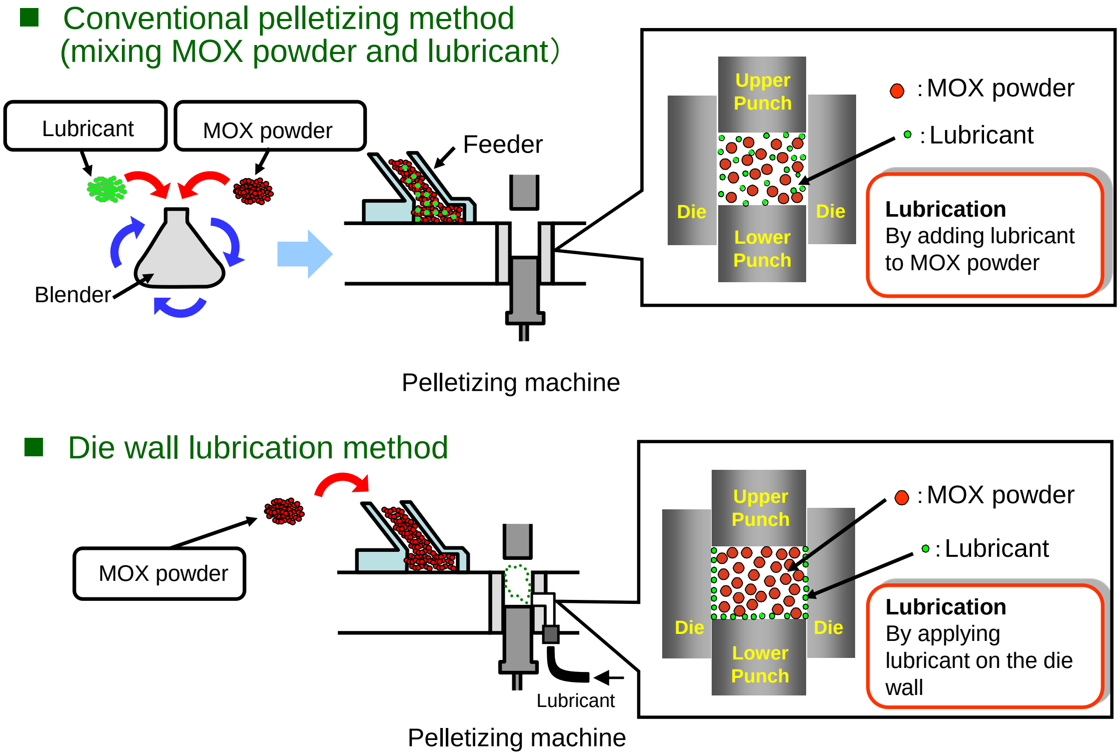

Figure 10. Comparison between the conventional and the new pelletizing method

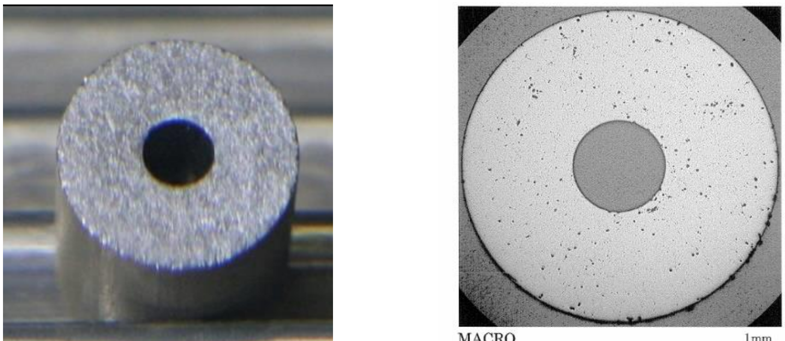

Figure 11. Photograph and ceramograph of an annular type high-density pellet obtained by the die wall lubrication pelletizing method

4. Conclusion

JAEA workers have developed large-scale MOX pellet fabrication technology for fuel of FRs and have fabricated 301 fuel assemblies for JOYO and 366 fuel assemblies for MONJU at the PFPF since 1988. During this time, many pellet fabrication difficulties were encountered, especially for MONJU fuel production due to the addition of large amounts of organic additives to the feed powders. In large-scale, low-density MOX fuel pellet production, organic additive performance is easily degraded by decay heat of the MOX powder. Countermeasures for the decay heat such as using high heat resistance organic additives and reducing content of added pore-former when using large particle size recycled powder have been developed. The die wall lubrication pelletizing method was tested for large-scale MOX pellet fabrication. No large pores and no cracks in the obtained sintered pellets were observed. Content of impurities in the obtained pellets met specifications of JOYO and MONJU fuels. Standard deviation of pellet density in the test was comparable with that in past JOYO pellet fabrications. However, the large-scale test using the die wall lubrication method was carried out for just one batch. Therefore, further large-scale tests will be needed to confirm product pellet quality and to find the best pelletizing conditions.

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