# Development of density control technologies for MOX pellet using dry recycled powders

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**Abstract**

Using dry recycled MOX powder, this study aims to develop a technology for controlling the density of MOX pellets. A roll crusher and a jet pulverizer were employed to prepare recycled MOX powder, which had three types of particle sizes (coarse, medium, and fine). The sintering tests of the MOX pellets were performed using particle size and dry recycled powder addition rate as parameters, and the effect of dry recycled powder addition rate on pellet quality was evaluated. For the coarse and medium dry recycled powders, a decrease in density due to addition was confirmed, but for the fine dry recycled powders, almost no decrease in density due to addition was confirmed. From this, it is considered that the fine dry recycled powder can be used in the same manner as the raw material powder, such as the raw MOX powder provided the addition rate is up to approximately 40 wt%.

## INTRODUCTION

Various developments have been made to effectively use dry recycled powders obtained from crushing rejected pellets generated in the fast reactor fuel fabrication process. One development is the fabrication of low-density pellets using dry recycled powder. Low-density pellets in fast reactor fuels have been fabricated using organic additives as pore-formers and used to mitigate pellet-cladding mechanical interaction at high burn-ups [1, 2]. However, there is a problem; increasing the pore-former addition rate increases the dispersion in the density and decreases the product yield. Therefore, a method for decreasing density by increasing the addition rate of coarse dry recycled powder (hereinafter called "coarse powder") instead of adding the pore-former has been reported [3]. According to the report, a coarse powder addition of up to 25 wt% is effective in decreasing the pore-former addition rate and dispersion in density.

In this study, for further developing pellet density control, pellets were prepared by adding a combination of coarse powder and dry recycled powder having small particle size, which was adjusted using a jet pulverizer (medium and fine). The effect of dry recycled powder and pore-former addition rate on pellet quality was evaluated.

## experimental

### Pellet fabrication test

The pellet fabrication flow is shown in FIG.1. In the step involving the weighing of raw materials, UO2-PuO2 mixed oxide (MOX) powder prepared by the microwave heating denitration method (MH-MOX), UO2 powder prepared by the ADU method (ADU-UO2), and dry recycled powder were weighed; the total was 300 gMOX with approximately 30% plutonium content and 0–45 wt% concentration of dry recycled powder. In this test, three kinds of dry recycled powders with different particle sizes (coarse, medium, and fine) were prepared using a roll crusher and a jet pulverizer and were used. To prepare the coarse dry recycled powder, scrap pellets from the pellet fabrication process were crushed using a roll crusher in two steps and then classified by passing through a mesh. To prepare the medium dry recycled powder, crashed pellets using a roll crusher were ground using a jet pulverizer in one step. To prepare the fine dry recycled powder, crashed pellets using a roll crusher were ground using a jet pulverizer in two steps. The feed powder was loaded into a ball mill pot (volume 1 L) with alumina balls, pulverized, and then mixed for 5 h. The rotation speed was 100 rpm, and the ball load had a J value of 0.5. In the additive blending (I) step, milled powder and the organic additives, such as the pore-former (crystalline cellulose, particle size: approximately 75 to 100 µm) and binder (0.5 wt% zinc stearate) were mixed for 10 min by hand. In granulation step, milled powder was first pressed into tablets at approximately 200 MPa. The tablets were then crushed and sieved with mesh having 212 to 850 μm opening. In the additive blending (II) step, granulated powder and the lubricant (0.2 wt% zinc stearate) were mixed for 10 min by hand. Then, granulated powder was pressed into green pellets at approximately 400 MPa. Green pellets were dewaxed for 2 h at 800 °C and sintered for 4 h at 1,700 °C in an atmosphere of Ar + 5% H2 mixed gas. The sintering furnace is shown in FIG.2. A total of 21 types of pellets were sintered (TABLE 1). The density of the sintered pellets was determined by measuring the sample mass and dimensions.



*FIG. 1. Pellet fabrication flow*



*FIG. 2. Sintering furnace*

TABLE 1. DRY RECYCLED POWDER AND

PORE-FORMER ADDITION RATES FOR PELLETS

|  |  |  |
| --- | --- | --- |
| Sample No. | Dry recycled powder addition rate (wt%) | Pore-former addition rate (wt%) |
| Coarse | Medium | Fine |
| SH-0 | 0 | 0 | 0 | 0 |
| SL-01 | 1 |
| SL-02 | 2 |
| MH-1 | 5 | 0 | 25 | 0 |
| ML-11 | 1 |
| ML-12 | 2 |
| MH-2 | 10 | 0 | 25 | 0 |
| ML-21 | 1 |
| ML-22 | 2 |
| MH-3 | 10 | 0 | 35 | 0 |
| ML-31 | 1 |
| ML-32 | 2 |
| MH-4 | 0 | 10 | 20 | 0 |
| ML-41 | 1 |
| ML-42 | 2 |
| MH-5 | 0 | 15 | 20 | 0 |
| ML-51 | 1 |
| ML-52 | 2 |
| MH-6 | 0 | 15 | 30 | 0 |
| ML-61 | 1 |
| ML-62 | 2 |

### Densification test

Using the pellets obtained in the pellet fabrication test, a cumulative 12 h (4 h × 3 times) densification test was performed at 1700 °C in an atmosphere of Ar + 5% H2 mixed gas, which is the same as the sintering conditions.

## Results

### Pellet fabrication test

FIG. 3. shows particle size distributions and specific surface areas of the three kinds of dry recycled powders: coarse, medium, and fine. Both the modal diameter and specific surface area tended to increase in the following order: coarse, medium, and fine.



Specific surface area: 0.12 m2/g (coarse), 0.50 m2/g (medium), 1.24 m2/g (fine)

*FIG. 3. Particle size distribution of dry recycle powder*

TABLE 2 shows the sintered pellet densities. FIG. 4. shows the sintered pellet densities of sample No. MH-1 and MH-2 as functions of the coarse powder addition rate, which differ only in the addition rate of the coarse powder. The density tended to decrease as the coarse powder addition rate increased from 5 to 10 wt%. In addition, the rate of decrease in density was not constant; it was largest when 2 wt% of pore-former was added. A similar tendency was observed when medium dry recycled powder (hereinafter called "medium powder") was added instead of coarse powder.



*FIG. 4. Dependence of the addition rate of coarse powder on sintered pellet densities*

FIG. 5. shows the sintered pellet densities of sample No. MH-2, MH-3, MH-5, and MH-6 as functions of the fine dry recycled powder (hereinafter called "fine powder") addition rate. For fine powder, unlike the results of coarse and medium dry powder, the sintered pellet density was almost constant regardless of the dry recycled powder addition rate when 0–1 wt% of pore-former was added. Further, when the pore-former addition rate was 2 wt%, the sintered pellet density tended to increase slightly as the dry recycled powder addition rate increased.

TABLE 2. THE SINTERED PELLET DENSITIES

|  |  |  |
| --- | --- | --- |
| Sample No. | Number of pellets | Density (%T.D.) |
| Mean | Standard deviation |
| SH-0 | 17 | 93.80 | 0.30 |
| SL-01 | 20 | 90.70 | 0.34 |
| SL-02 | 17 | 85.50 | 0.11 |
| MH-1 | 17 | 94.34 | 0.51 |
| ML-11 | 20 | 91.06 | 0.16 |
| ML-12 | 17 | 85.75 | 0.38 |
| MH-2 | 19 | 93.07 | 0.22 |
| ML-21 | 20 | 89.29 | 0.32 |
| ML-22 | 19 | 79.34 | 0.36 |
| MH-3 | 17 | 93.15 | 0.23 |
| ML-31 | 20 | 88.61 | 0.35 |
| ML-32 | 17 | 80.47 | 0.53 |
| MH-4 | 17 | 95.72 | 0.15 |
| ML-41 | 20 | 92.12 | 0.10 |
| ML-42 | 17 | 87.44 | 0.18 |
| MH-5 | 19 | 94.02 | 0.16 |
| ML-51 | 20 | 89.85 | 0.36 |
| ML-52 | 19 | 80.21 | 0.25 |
| MH-6 | 17 | 94.14 | 0.22 |
| ML-61 | 20 | 90.22 | 0.12 |
| ML-62 | 17 | 81.34 | 0.44 |



*(a) coarse and fine (b) medium and fine*

*FIG. 5. Dependence of the addition rate of fine powder on sintered pellet densities*

FIG. 6. shows the sintered pellet density of the SH-0 series without the addition of dry recycled powders. The sintered pellet density tended to decrease as the pore-former addition rate increased. However, the rate of decrease was not constant; it was greater when increased from 1 to 2 wt% than when increased from 0 to 1 wt%.

FIG. 7. shows vertical section images of the sintered pellets. The big pores were confirmed inside MH-1, and cracks were confirmed inside MH-1 and MH-4.



*FIG. 6. Dependence of the addition rate of pore-former on sintered pellet densities*



*(a) MH-1 (b) ML-12 (c) MH-4 (d) ML-42*

*FIG. 7. Vertical section images of the sintered pellets*

### Densification test

TABLE 3. shows the re-sintered pellet densities. FIG. 8. shows a large amount of densification after re-sintering three times (12 h in total). As shown in FIG. 8., densification (density reduction due to re-sintering) was large in the first (4 h) re-sintering and sharply decreases in the second (8 h) and third (12 h) re-sintering. The shrinkage of these pellets was not completed after sintering for 4 h, and it is supposed that the sintering was almost completed by the re-sintering process in 4 h. In addition, the MH-1 and MH-4 series tended to decrease in density after re-sintering.

TABLE 3. THE RE-SINTERED PELLET DENSITIES

|  |  |  |
| --- | --- | --- |
| Sample No. | Number of pellets | Density (%T.D.) |
| 4 h | 8 h | 12 h |
| Mean | Standard deviation | Mean | Standard deviation | Mean | Standard deviation |
| SH-0 | 5 | 94.41 | 0.19 | 94.42 | 0.19 | 94.38 | 0.17 |
| SL-01 | 5 | 91.54 | 0.09 | 91.57 | 0.08 | 91.46 | 0.06 |
| SL-02 | 5 | 86.75 | 0.11 | 86.93 | 0.12 | 86.98 | 0.17 |
| MH-1 | 8 | 94.98 | 0.09 | 94.79 | 0.08 | 94.70 | 0.13 |
| ML-11 | 5 | 91.34 | 0.08 | 91.22 | 0.08 | 90.95 | 0.10 |
| ML-12 | 5 | 86.75 | 0.30 | 86.61 | 0.29 | 86.47 | 0.19 |
| MH-2 | 5 | 93.88 | 0.13 | 94.05 | 0.10 | 94.09 | 0.11 |
| ML-21 | 5 | 90.09 | 0.19 | 90.27 | 0.06 | 90.31 | 0.08 |
| ML-22 | 5 | 81.28 | 0.36 | 81.60 | 0.14 | 81.98 | 0.31 |
| MH-3 | 5 | 94.04 | 0.13 | 93.85 | 0.14 | 93.96 | 0.16 |
| ML-31 | 5 | 89.50 | 0.20 | 89.46 | 0.12 | 89.54 | 0.22 |
| ML-32 | 5 | 82.28 | 0.25 | 82.38 | 0.26 | 82.59 | 0.32 |
| MH-4 | 8 | 95.94 | 0.16 | 95.70 | 0.13 | 95.64 | 0.19 |
| ML-41 | 5 | 92.39 | 0.07 | 92.18 | 0.08 | 91.83 | 0.10 |
| ML-42 | 5 | 88.23 | 0.09 | 88.08 | 0.08 | 87.92 | 0.06 |
| MH-5 | 5 | 94.81 | 0.06 | 94.74 | 0.10 | 94.74 | 0.07 |
| ML-51 | 5 | 90.65 | 0.22 | 90.54 | 0.14 | 90.56 | 0.17 |
| ML-52 | 5 | 82.82 | 0.23 | 83.25 | 0.18 | 83.57 | 0.12 |
| MH-6 | 5 | 94.90 | 0.15 | 94.75 | 0.07 | 94.73 | 0.09 |
| ML-61 | 5 | 90.81 | 0.14 | 90.74 | 0.13 | 90.86 | 0.09 |
| ML-62 | 5 | 83.19 | 0.22 | 83.68 | 0.16 | 83.81 | 0.28 |



*FIG. 8. Densification after re-sintering*

## DIscussion

For the test in which coarse + fine powder was added, the density decrease behavior of each powder due to the addition of coarse powder or pore-former are as follows:

* Density decrease due to the addition of coarse powder

5 wt% increase ⇒ approximately 1.5 %T.D. density decrease (from MH-1 and MH-2)

* Density reduction due to the addition of pore-former

0 → 1 wt% ⇒ approximately 3 %T.D. density decrease (from SH-0 and SL-01)

1 → 2 wt% ⇒ approximately 5 %T.D. density decrease (from SL-01 and SL-02)

As shown in FIG. 4., when the density of ML-22 decreased independently by the addition of coarse powder or pore-former, the sintered pellet density of ML-22 was approximately 85 %T.D. However, the sintered pellet density of ML-22, which increased the addition rate of both coarse powder and pore-former, deviated significantly from these tendencies, and it further decreased by approximately 5 %T.D. From this, it is considered that when the addition rates of the coarse dry powder and pore-former are increased to or above certain levels, a synergistic effect that decreases the sintered pellet density of both is generated. This tendency was identical for the test in which the medium + fine powder and pore-former were added. In this test, this effect was observed with 10 wt% coarse powder + 2 wt% pore-former or 15 wt% medium powder + 2 wt% pore-former.

As shown in FIG. 5., the sintered pellet density tended to increase when the fine powder was added to the condition where the density decreased due to the above synergistic effect. When dry recycled powder having a relatively large particle size is added, voids between the dry recycled powders are formed in a green pellet. Then, the ratio of the voids to the entire powder increased as the dry recycled powder addition rate increased. Most of the voids are filled with a powder having a smaller particle size. In this test, it is considered that the raw material powder (MH-MOX and UO2 powder) and fine powder correspond to the powder having a small particle size. However, when the powder filling the voids was the raw material powder, voids were generated due to the difference in the shrinkage ratio with the dry recycled powder during sintering, and the density decreased. Moreover, when it was filled with fine dry powder, it is considered that the decrease in density was alleviated because the proportion of voids generated was small because there was almost no difference in shrinkage.

Densification after re-sintering is shown in FIG. 9 for each sample No. The MH-1 series was excluded because the density decreased. From FIG. 9, in the result of the addition rate of 0–1 wt% pore-former, there was an insignificant difference in densification due to the difference in the addition rate of the dry recycled powder.



*FIG. 9. Densification for each sample No.*

However, when a 2 wt% pore-former was added, densification significantly increased by adding the 10 wt% coarse powder + 25 wt% fine powder (ML-22), and densification was slightly reduced by adding 10 wt% the fine grain (ML-32). From these results, it is considered that the sintering rate was lowered by adding the coarse powder and the pore-former in a certain amount or more, and the influence was alleviated by increasing the addition rate of the fine powder. In addition, the same tendency was observed in the test in which the medium + fine powder was added.

From the vertical cross-sectional images shown in FIG. 7, cracks were observed inside some sintered pellet to which the dry recycled powder was added. Details are shown below. This is considered to be due to the difference in the shrinkage rate during sintering with the raw material powder. Moreover, no similar cracks were observed inside the sintered pellets to which 2 wt% pore-former was added, although the addition rate of the dry recycled powder was the same, which was because the difference in volume changed due to the difference in shrinkage between the dry recycled powder and the raw material, the growth of microcracks, etc. were alleviated by the pores generated by the addition of the pore-former.

## conclusion

Sintering tests of MOX pellets conducted using particle size and dry recycled powder addition rate as parameters. The results are summarized as follows.

For the coarse and medium powders, a decrease in density due to addition was confirmed, but for the fine powders, almost no decrease in density due to addition was confirmed. From this, it is considered that the fine powder can be used in the same manner as the raw material powder, such as the raw MOX powder provided the addition rate is up to approximately 40 wt%.

When dry recycled powder (coarse or medium) and pore-former were added simultaneously, a synergistic effect was produced in addition to the density decrease effect of both, and the density was lower than expected. In addition, this synergistic effect occurred within the range of this test at 10 wt% coarse powder + 2 wt% pore-former or 15 wt% medium powder + 2 wt% pore-former. Further, it is considered that this synergistic effect can be alleviated by adding fine powder.

It is considered that the addition of coarse and medium powder can delay the progress of sintering by adding it together with the pore-former, and the influence can be suppressed by adding fine powder.

A large amount of dry recycled powder addition induced cracks in the pellets, but the addition of 2% by weight of pore-formers did not.

References

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