PROCESS OPTIMIZATION OF DUPIC FUEL PELLET FABRICATION

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ABSTRACT

DUPIC pellets are remotely fabricated by using DUPIC powder prepared by the OREOX treatment of spent fuel pellets. DUPIC pellets were successfully fabricated using spent PWR fuel material with an average discharge burn-up of 27,300 MWd/tU. Sintered density, grain size and surface roughness of the DUPIC pellets were investigated on the basis of CANDU fuel criteria.

In order to optimize the DUPIC pellet manufacturing processes, 3 series of experiments for the pre-qualification and 3 series for the qualification were performed. In these experiments, the sintered densities of the pellets ranged from 10.35 g/cm^3(95.7 % of T.D.) to 10.43 g/cm^3(96.4 % of T.D.) and the average grain size ranged from 14.6 to 14.9 μm.

Based on these results, the optimum manufacturing processes of DUPIC pellets have been established. Then, under the control of the QA program developed with the assistance of AECL, 8 series of production runs have been performed to make DUPIC pellets in a batch size of 1 kg. The sintered densities of the fabricated pellets ranged from 10.26 g/cm^3 to 10.43 g/cm^3. The surface roughness of the ground pellets was less than Ra 0.8 μm by the dry grinding process. As the results of the production runs, DUPIC fuel pellets meeting the standard CANDU fuel specifications were successfully fabricated by the established processes.

1. INTRODUCTION

Since 1999, DUPIC fuel has been fabricated at KAERI using the spent PWR fuel discharged from the nuclear power plant Gori #1 in 1986 to develop DUPIC(Direct Use of spent PWR fuel In CANDU reactor) fuel cycle technology. DUPIC pellets are remotely fabricated by using DUPIC powder which was prepared by the OREOX(Oxidation and REduction of OXide fuel) treatment of spent fuel pellets extracted from spent PWR fuel rods[1-5]. At first, the DUPIC pellets could be remotely fabricated with the spent PWR fuel with a burn-up of 35,000 MWd/tU in a hot-cell. The sintered density of the fabricated DUPIC pellets ranged from 10.01 g/cm^3 to 10.43 g/cm^3 [6]. The grain size of the sintered DUPIC pellets ranged from 7.26 μm to 9.48 μm. The DUPIC pellets with a surface roughness of less than Ra 0.8 μm could be fabricated by the dry grinding process. Based on the analyses of the experimental results, the DUPIC fuel fabrication has been commenced.

In this study, DUPIC pellets were fabricated using spent PWR fuel rods with an average discharge burn-up of 27,300 MWd/tU. Sintered density, grain size and surface roughness of the DUPIC pellets were investigated on the basis of the criteria of CANDU fuel.

In order to optimize the DUPIC pellet manufacturing processes, 3 series of experiments for the

pre-qualification were performed. The sintered density, dimensions, surface roughness, surface defects and the grain size of DUPIC pellets have been measured by using the remote inspection systems developed to inspect highly radioactive DUPIC pellets in a hot-cell[7]. DUPIC pellets with 12.19 mm of diameter, 10.37~10.45 g/cm^3 of sintered density have been successfully fabricated in the pre-qualification test. The optimum process parameters for DUPIC pellet fabrication have been established based on the test. Then, the qualification test was performed for 3 batches of experiments to qualify the fabrication processes developed by the pre-qualification test[8]. DUPIC pellets with 12.189~12.200 mm of diameter, 10.35~10.43 g/cm^3(95.7 ~ 96.4 % of T.D.) of sintered density have been successfully fabricated in the qualification test. And, the average grain size ranged from 14.6 to 14.9 μm. As the results of the tests, DUPIC pellets satisfying the requirements of standard CANDU fuel were fabricated under process conditions of 130 MPa of compaction pressure, 1800 °C of sintering temperature and a sintering time of 10 hours.

Based on the results, the optimum manufacturing processes of DUPIC pellets have been established. Then, under the control of the QA program developed with the assistance of AECL, 8 series of production runs have been performed to make DUPIC pellets in a batch scale of 1 kg[9].

2. FABRICATION OF DUPIC FUEL PELLETS

The OREOX process was selected to enhance the sinterability of the spent fuel powder based on the previous experimental results. DUPIC pellets were fabricated in accordance with the DUPIC fuel fabrication process flow shown in figure 1. Based on both the pre-qualification test and qualification test, the process parameters have been established as shown in table 1[5,6].

2.1 Raw material

DUPIC fuel pellets were fabricated with spent PWR fuel discharged from the nuclear power plant Gori #1 in 1986. The average burnup of the spent PWR rods used for this experiment is 27,300 MWd/tU.

2.2 Powder process

A spent PWR fuel rod was cut to make rodcuts with a length of 20~25 cm. Then, the rodcut was longitudinally slit by a slitting machine. The decladded pellets were oxidized to make a powder. All the oxidized powder was blended together to make a homogeneous powder feed. The homogeneity of the blended powder was measured by chemical analysis. U-235 difference from a mean value ranged from 0.62 to 1.7 %. Pu-239 difference from a mean value ranged from 0.46 to 1.27 %. These results met the criteria that the difference should be less than 5 %. The homogenized powder was treated through the three cyclic OREOX process, where the oxidation temperature was 450 °C, and the reduction temperature was 700 °C. The OREOX-treated powder was milled by a attritor at the speed of 450 rpm for 10 minutes and 600 rpm for 10 minutes. The milled powder was pre-compacted at the pressure of 62 MPa, then the pre-compacted pellets were granulated by using a sieve of mesh numbered #18. The granulated powder was mixed with zinc stearate to be suitable for final pressing.

2.3 Fabrication of DUPIC pellets

Green pellets were fabricated by the final compaction process with a pressure ranging from 96 MPa to 124 MPa. Zinc stearate was removed during heating of the green pellets for 3 hours at 800 °C in an Ar-4%H_2 atmosphere. The sintering process was performed at 1800 °C for 10 hours in an Ar-4%H_2 atmosphere.

The sintered density of a few of the pellets for 4 batches of 8 runs did not meet the criterion of 10.25 g/cm^3. The pellets were sintered again at 1850 °C for 5 hours in an Ar-4%H_2 atmosphere to increase the sintered density.

The sintered pellets were ground by a dry grinder to adjust the diameter and surface roughness. The pellets were characterized to evaluate their quality on the basis of the standard CANDU fuel specifications.

3. FABRICATION DATA

3.1 Characteristics of DUPIC powder

Both the apparent density and the tap density of DUPIC powders were measured after the OREOX process and the milling process. The apparent density of the OREOX-treated powder ranged from 0.63 to 0.84 g/cm^3. The tap density of the OREOX-treated powder ranged from 1.04 to 1.27 g/cm^3. After the milling process, the densities increased to become about 3 tim es larger than those of the OREOX-treated powders.

3.2 Characteristics of DUPIC pellets

The variations of the immersion density, the grain size, and the surface roughness of the DUPIC pellets were investigated with the variations of process parameters such as compaction pressure, sintering temperature and sintering time.

3.2.1 Density of slug

Slugs were prepared by the pre-compacting process for free-flowing granules. Geometric density of the slug was calculated by measuring the weight, length and diameter of the slug. The density ranged from 5.99 to 6.20 g/cm^3.

3.2.2 Density of green pellet

Geometric density of the green pellet was calculated by measuring the weight, length and diameter of the pellet. The density ranged from 6.52 to 6.57 g/cm^3. In this experiment, green density was controlled to produce defect-free DUPIC pellets by adjusting the final compaction pressure ranging from 96 MPa to 124 MPa.

3.2.3 Density of sintered pellet

Green pellets were sintered at 1800 . for 10 hours in an Ar-4%H_2 atmosphere. In case of the production runs in the batches numbered B02-04, B02-05, B02-06 and B02-07, the pellets were sintered again at 1850 . for 5 hours to increase the sintered density. In these cases, nonconformance reports were issued in accordance with the QA program of DUPIC fuel. Immersion density of the sintered pellet was calculated by measuring the pellet weight both in air and in water as shown in figure 2. The mean value of the sintered density for each production

run ranged from 10.278 to 10.379 g/cm^3. The minimum density of a sintered pellet was 10.258 g/cm^3. The maximum density of a sintered pellet was 10.433 g/cm^3. The shrinkage rate for the diameter ranged from 13.88 to 14.38 %.

3.2.4 Inspection of ground pellet

The sintered pellets were ground by the dry grinding process using a dry centerless grinder to adjust the diameter and surface roughness. After grinding, the shoulder width, the surface roughness, and the diameter of the sampled pellets were measured. The shoulder width ranged from 0.69 to 0.79 mm. The surface roughness of the ground pellets ranged from Ra 0.43 μm to Ra 0.79 μm. The DUPIC pellets with the surface roughness of less than Ra 0.8 μm were fabricated under the optimum process condition. The diameter of the ground pellets met the standard CANDU fuel specification. Then, visual inspection was conducted to check for any surface defect for all the fabricated pellets. Most of the pellets were defect-free on the surface. Some defects such as micro-crack and end-capping were found in a few pellets. 414 pellets were fabricated in this experiment. The yield rate reached to 91 %.

3.2.5 Microstructure of sintered pellets

Microstructure was investigated for two sintered pellets in the qualification test. The sampled pellet was divided into two parts by cutting it longitudinally. The cut surface was ground and etched. The microstructure of the pellet was observed by an optical microscope with a magnification of 200. Figure 3 shows the microstructure of an observed DUPIC pellet. The averaged grain size ranged from 14.6 to 14.9 μm.

3.2.6. Chemical analysis of DUPIC pellets

The composition of DUPIC powder and a pellet was chemically analyzed. The Cs-134/Cs-137 content of a sintered pellet was less than 1 % of that of the powder. 99 % of Cs was vaporized during the sintering process. It was confirmed that the vaporized cesium was trapped by the fission gas trapping filter of the off-gas treatment system.

4. CONCLUSIONS

10 kg of spent PWR fuel was transported to the DFDF(DUPIC Fuel Development Facility) for fabricating DUPIC fuel pellets. A pre-qualification experiment was performed to establish the process conditions of DUPIC pellets. Then, a qualification test was performed on the basis of the results of the pre-qualification experiment. And, the optimum manufacturing processes of DUPIC pellets were established. Then, under the control of the QA program developed with the assistance of AECL, 8 series of production runs were performed to make DUPIC pellets. The results were as follows.

- DUPIC pellets were fabricated in a batch scale of 1 kg under the QA program.
- Fabrication data were documented and analyzed in accordance with the QA program.
- Defect-free pellets with sintered densities ranging from 10.26 to 10.43 g/cm^3 were successfully produced.
- DUPIC pellets satisfying the standard CANDU fuel criteria were successfully fabricated by the optimized processes.

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TABLES

TABLE 1. FABRICATION PROCESS PARAMETERS OF DUPIC PELLETS

No.	Process	Operating Conditions
1	OREOX	•Oxidation : 450 ℃, 2hrs, Air, 5.5 L/min •Reduction : 700 ℃, 7hrs, Ar/4%H_2, 13L/min •Passivation : 80 ℃, 4hrs, Ar/2%O_2, 2L/min •Heating rate : 4 ℃/min
2	Milling	•Milling Time : 20 minutes - Zirconia ball - 5 mm(ϕ) - 450 rpm for 10 mins + 600 rpm for 10 mins
3	Mixing 1	•0.2 wt% zinc stearate after milling •Mix for 20 minutes
4	Pre-compaction	•62 Mpa •Die diameter : 14.34 mm
5	Granulation	•Sieve #18(1 mm opening)
6	Mixing 2	•0.2 wt% zinc stearate after granulation •Mix for 20 minutes
7	Compaction	•Final compaction pressure : < 130 MPa •Die diameter : 14.34 mm
8	Dewaxing	•Dewaxing temperature / time : 800 ℃ / 3 hrs •Gas : Ar-4%H_2 flow rate 4 L/min •Heating rate : 4 ℃ /min
9	Sintering	•Sintering temp./time : 1800 ℃ / 10hrs •Ar/4% H_2, 8.0 L/min •Heating rate : 5 ℃ /min

FIGURES

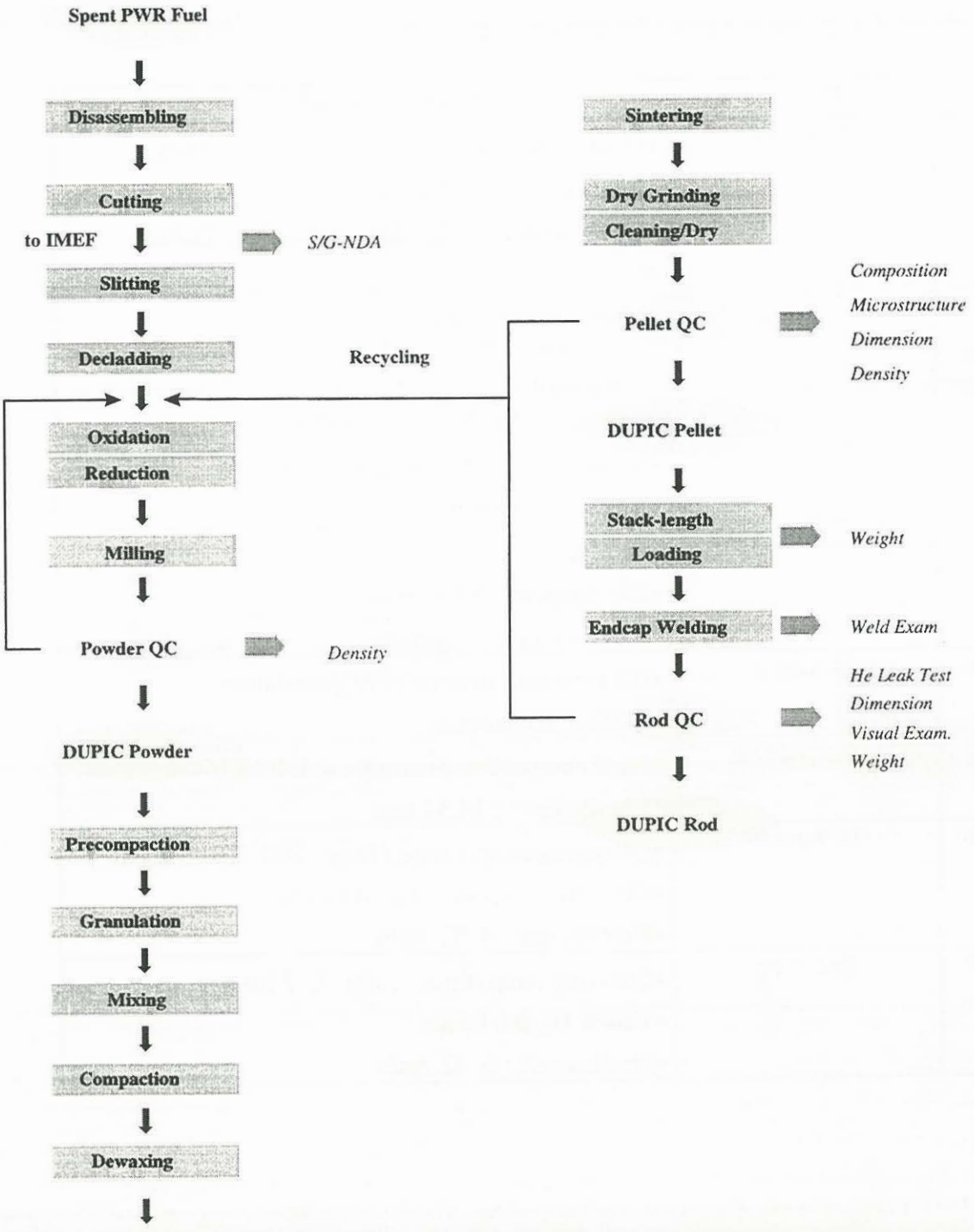

FIGURE 1. DUPIC FUEL FABRICATION PROCESS FLOW

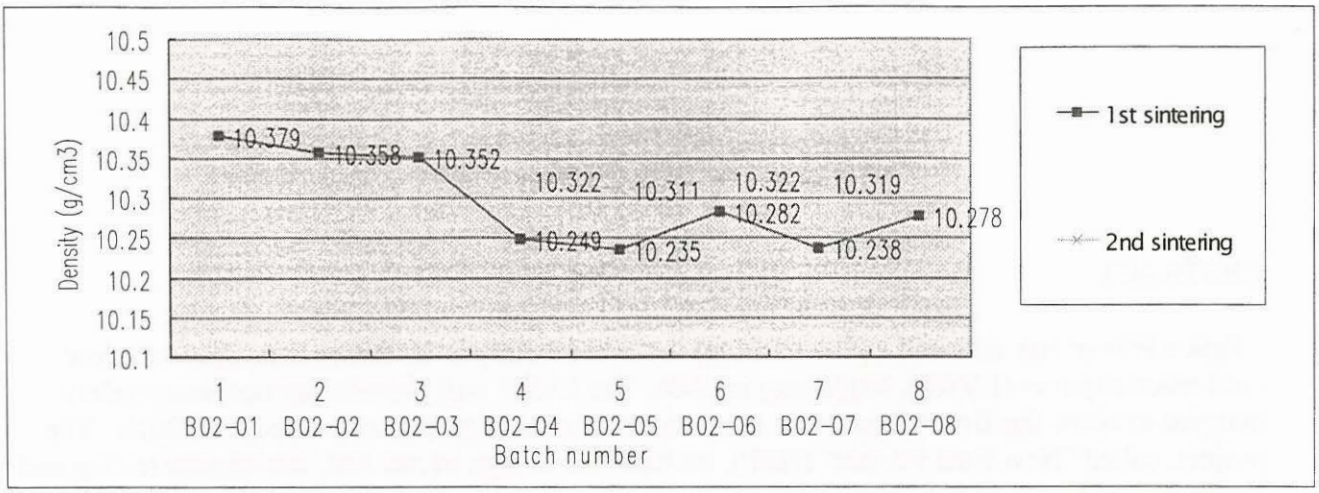

FIGURE 2. DENDITY OF SINTERED DUPIC PELLETS

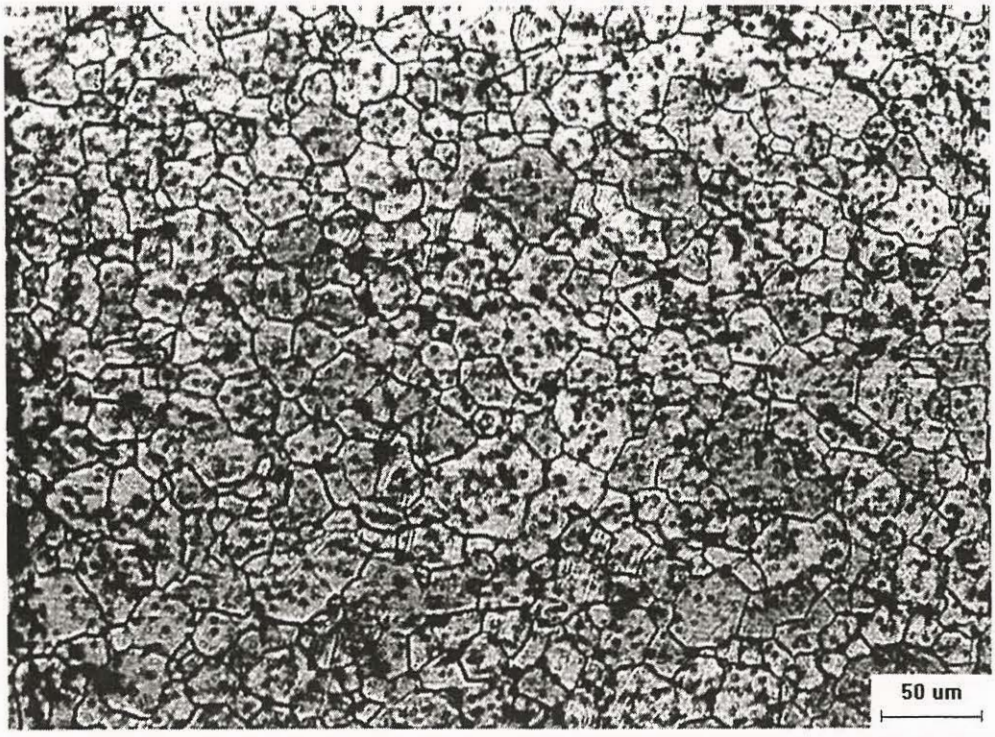

FIGURE 3. MICROSTRUCTURE OF SINTERED DUPIC PELLET