

EFFECT OF IMPURITIES ON THE MICROSTRUCTURE OF DUPIC FUEL PELLETS USING THE SIMFUEL TECHNIQUE

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The influence of fission products' contents on the DUPIC fuel powder and pellet properties was experimentally evaluated using SIMFUEL as a surrogate for actual spent PWR fuel due to the high radioactivity of spent fuel. Pure UO_2 and SIMFUEL pellets with fission products equivalent to a burn-up of 35,000 MWd/tU and 60,000 MWd/tU were used as impurities in this study. The specific surface area of the powder milled after the OREOX treatment increased and resulted in sintered pellets with a theoretical density (TD) higher than 95%, regardless of the impurity contents. However, the grain size of the sintered pellets decreased with the increasing impurity contents. As a result of the dissolved oxides in UO_2 from the impurity groups, the specific surface area of the OREOX powder increased with an increase of the impurities. The grain size of the sintered pellets was significantly decreased by the metallic and oxide precipitates.

KEYWORDS : DUPIC, SIMFUEL, Fission Products, Sintered Density, Grain Size

1. INTRODUCTION

The fuel burn-up in Pressurized Water Reactor (PWR) is increasing to improve fuel's economy and to reduce the spent fuel per unit energy production. When considering a strategy to extend the burn-up of PWR fuel, it is necessary to analyze the effect of spent fuel burn-up on DUPIC (Direct Use of spent PWR fuel In CANDU reactor) fuel fabrication from various viewpoints as a key parameter that influences the fuel fabrication characteristics. A high burn-up spent fuel near 60,000 MWd/tU has a higher amount of fission products compared with a typical spent fuel irradiated at a normal burn-up of 35,000 MWd/tU.

DUPIC is a fuel recycling technology that directly fabricates CANDU fuel from spent PWR fuel material using a dry process [1]. Fission gases such as Xe and Kr are removed from the fuel powder converted from a spent fuel pellet during the OREOX process (oxidation and reduction of oxide fuels), and volatile nuclides such as Cs are removed from the green pellet during the sintering process [2, 3]. However, other fission products remain in the fuel powder and green pellet, and thus, these fission products could significantly affect the DUPIC fuel powder and pellet properties.

The influence of the fission product contents and groups on the DUPIC fuel powder and pellet properties were experimentally evaluated using SIMFUEL (SIMulated FUEL) as a surrogate for actual spent PWR fuel due to the

high radioactivity of spent fuel [4-6]. SIMFUEL replicates the chemical state and microstructure of the three groups of fission products: the dissolved elements in the UO_2 matrix, the metallic precipitates, and the oxide precipitates, excepting the fission gas and volatile elements [7-9]. The SIMFUEL was prepared using a mixture of UO_2 and surrogated metallic oxides for the fission products.

In the present work, the influence of the fission products' contents and groups as an impurity in the simulated DUPIC fuel powder and pellet properties was investigated using pure UO_2 and SIMFUEL pellets with equivalent burn-ups of 35,000 MWd/tU and 60,000 MWd/tU, respectively.

2. EXPERIMENTAL

2.1 Pellet Preparation

The fission product composition of irradiated UO_2 fuel depends on its initial enrichment and irradiation history. The ORIGEN (Oak Ridge Isotope Generation and Depletion) code was used to calculate the fission product inventories to be added to the UO_2 powder to prepare the SIMFUEL pellets [10]. The fifteen elements listed in Table 1 represent the major fission products excepting the volatile elements. The additives of these stable oxides were milled using a mortar and mixed with the UO_2 powder in a turbular type mixer. Wet attrition

Table 1. Chemical Composition and Surrogated Oxides Added to UO₂ as Fission Products

Impurity groups	Uranium and fission products	Surrogate oxides	Element composition (wt.%)	
			35,000 MWd/tU ^{a)}	60,000 MWd/tU ^{b)}
	U	UO ₂	96.768	94.505
Dissolved oxides (SS)	Y	Y ₂ O ₃	0.048	0.086
	La	La ₂ O ₃	0.172	0.227
	Ce (Pu, Np) ^{c)}	CeO ₂	0.814	1.405
	Nd (Pr, Sm) ^{c)}	Nd ₂ O ₃	0.630	1.054
		SUM	1.664	2.772
Dissolved oxides/oxide precipitates (OP)	Sr	SrO	0.078	0.164
	Zr	ZrO ₂	0.388	0.659
	Ba	BaCO ₃	0.193	0.278
		SUM	0.659	1.101
Metallic precipitates (MP)	Mo	MoO ₃	0.350	0.613
	Ru(Tc) ^{c)}	RuO ₂	0.317	0.595
	Rh	Rh ₂ O ₃	0.047	0.058
	Pd	PdO	0.145	0.266
		SUM	0.859	1.532
Oxide/metallic precipitates	Te	TeO ₂	0.049	0.09

^{a)} Cooling time: 15 years

^{b)} Cooling time: discharge

^{c)} The fission product in parenthesis was replaced by the fission product in front of parenthesis.

milling for 4 hours was used to obtain a homogeneous powder mixture. The powder mixture was pressed into green pellets at 300 MPa and sintered at 1700 °C for 6 hours in an atmosphere of Ar-4%H₂. Pure UO₂ pellets were prepared using the same milling, compaction, and sintering processes.

2.2 Simulated DUPIC Powder Preparation

For the preparation of the powder from the pure UO₂ and SIMFUEL pellet, the oxidation and reduction step, called the OREOX process, was applied repeatedly. The oxidation was performed at 500 °C in air and the reduction at 700 °C in Ar-4%H₂. The passivation was conducted at 70 °C in Ar-2%O₂. The OREOX powder was prepared through three cycles of oxidation and reduction. An attrition mill with a jar volume of 1.5 L was used to grind the OREOX powder into a finer powder. The milling medium was zirconia balls with a 5 mm diameter. The OREOX powder was poured into the milling jar at a constant ball-to-powder weight ratio (40:1) and milled at 150 rpm; the millings were performed for 15 minutes, which has been determined as optimal by a previous study [11].

2.3 Simulated DUPIC Pellet Fabrication

Acrawax powder of 0.2 wt.% was added to the OREOX powder as a lubricant and mixed with the milled powder in a turbular type mixer for 25 minutes at 25 rpm. Then, the mixed powder was pressed into green pellets using a pressure of 300 MPa via a single acting hydraulic press. The green pellets were sintered at 1700, 1750, and 1800 °C for 6 h in an atmosphere of Ar-4%H₂ with a flow rate of 3 L/min. The heating and cooling rates were controlled at 5 °C /min.

2.4 Characterizations of the Powder and Pellet

The mean particle size of the powder was measured using a laser particle size analyzer (Malvern, UK), and the specific surface area of the powder was determined via the BET method. The bulk density and tap density were measured using ASTM B329 and B527, respectively. The phase change of the powders after oxidation was analyzed using X-ray diffraction (XRD), and a scanning electron microscope (SEM) was applied to observe the powder morphology. The density of the sintered pellet was determined using a water immersion method

(Archimedes principle), and the grain size of the sintered pellet was determined via a linear intercept method using a sample thermally etched at 1550 °C for 2 h in Ar-4% H_2 .

3. RESULTS AND DISCUSSION

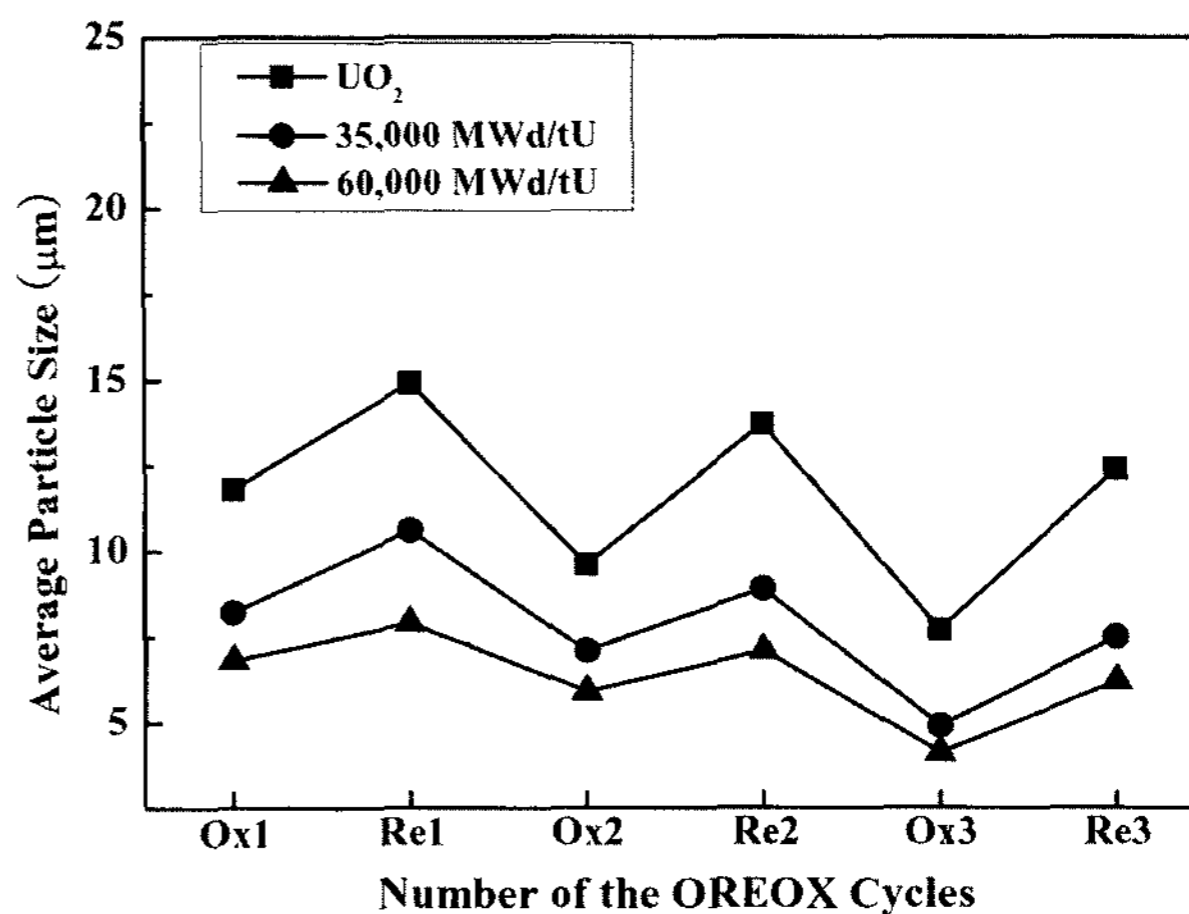
3.1 Effect of Fuel Burn-Up on Powder Characteristics

The densities of the SIMFUEL pellets prepared via the method described in Section 2.1, measured using an immersion method, were 10.37 g/cm³ (96% TD) for the SIMFUEL with a 35,000 MWd/tU burn-up, and 10.21 g/cm³ (95.9% TD) for 60,000 MWd/tU burn-up, respectively. The average grain size of the SIMFUEL pellets, determined using a linear intercept method, respectively, were 7 μ m for the SIMFUEL with a 35,000 MWd/tU burn-up, and 4 μ m for 60,000 MWd/tU burn-up, respectively. For the pure UO_2 pellets manufactured using the same preparation method, the density and average grain size were 10.64 g/cm³ (97.1% TD) and 8 μ m, respectively. In contrast to the sintered density, the grain size of the sintered pellets was significantly affected by an increasing amount of impurities.

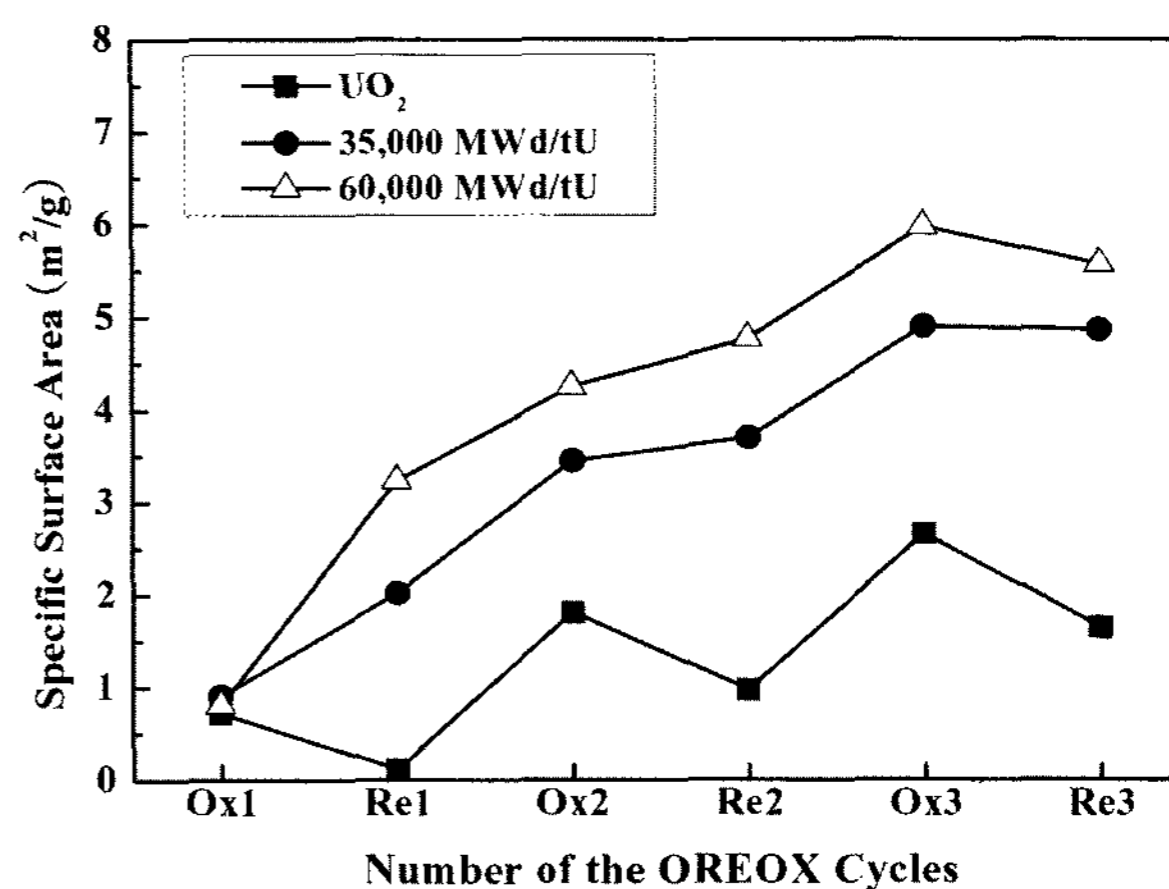
Figure 1 shows the average particle size and specific surface area of the pure UO_2 and the simulated DUPIC powder with the number of OREOX cycles. The average particle size of the fuel powders was determined via the grain size of the UO_2 and SIMFUEL pellets [12]. The average particle size of all fuel powders decreased due to the breakage of particles resulting from a volume expansion caused by a transformation of the UO_2 (TD: 10.96 g/cm³) phase into a U_3O_8 phase (TD: 8.4 g/cm³) during the oxidation steps, whereas it increased due to the bonding between the particles during the reduction steps. The specific surface area of the oxidized pure U_3O_8 powder in the reduction step decreased as the particle size increased. However, the specific surface area of the simulated DUPIC powder increased with an increasing particle size in the first and second reduction steps. In the third reduction step, the specific surface area of the simulated DUPIC powder decreased with an increasing particle size.

Figure 2 shows the SEM micrographs of the cyclic OREOX powders. The powders obtained after the first oxidation step (Figs. 2(a-1), (b-1), and (c-1)) show powders that are coarse and angular with large cracks. After the first reduction step, the surface of the UO_2 particles (Fig. 2(a-2)) remained coarse, whereas that of the simulated DUPIC particles (Figs. 2(b-2) and (c-2)) became rougher with an increasing amount of impurities, resulting in an increasing specific surface area. The three cycled OREOX powders for the SIMFUELS (Figs. 2(b-3) and (c-3)) showed sponge-like particle shapes with internal pores.

The physical properties of the pure UO_2 and simulated DUPIC powder produced via milling after three cycles of OREOX treatment of the pure UO_2 and SIMFUEL pellets are shown in Table 2. The specific surface area of the OREOX powders was significantly increased using dry



(a) Average particle size



(b) Specific surface area

Fig. 1. Characteristics of the SIMFUEL and UO_2 Powders with the Number of OREOX Cycles: (a) Average Particle Size and (b) Specific Surface Area

attrition milling due to the breakage of the particle size into the submicron range, thus resulting in an improvement of the sinterability. However, the order of the value for the specific surface areas according to the types of powder was not changed by the milling process. The bulk and tap densities of the OREOX powders also increased considerably after the milling process. Therefore, a higher packing density of the cyclic OREOX powder during compaction was obtained as shown in Table 2.

3.2 Effect of Fuel Burn-Up on the Simulated DUPIC Pellet Characteristics

The variations in the sintered density and average grain size of the simulated DUPIC pellets as a function of the sintering temperature are shown in Fig. 3. The simulated DUPIC pellets were fabricated using the milled powder

Table 2. Characteristics of the Pure UO₂ and Simulated DUPIC Powders

Properties	Pure UO ₂		35,000 MWd/tU		60,000 MWd/tU	
	As produced	After milling	As produced	After milling	As produced	After milling
Average particle size (μm)	12.4	0.8	7.6	0.6	6.2	0.4
Specific surface area (m ² /g)	1.64	2.65	4.86	5.38	5.57	6.21
Bulk density (g/cm ³)	0.94	1.80	0.71	1.63	0.72	1.95
Tap density (g/cm ³)	1.53	3.12	1.87	3.05	1.83	3.12
Green density (g/cm ³)	5.51	6.05	5.57	6.27	5.41	6.02

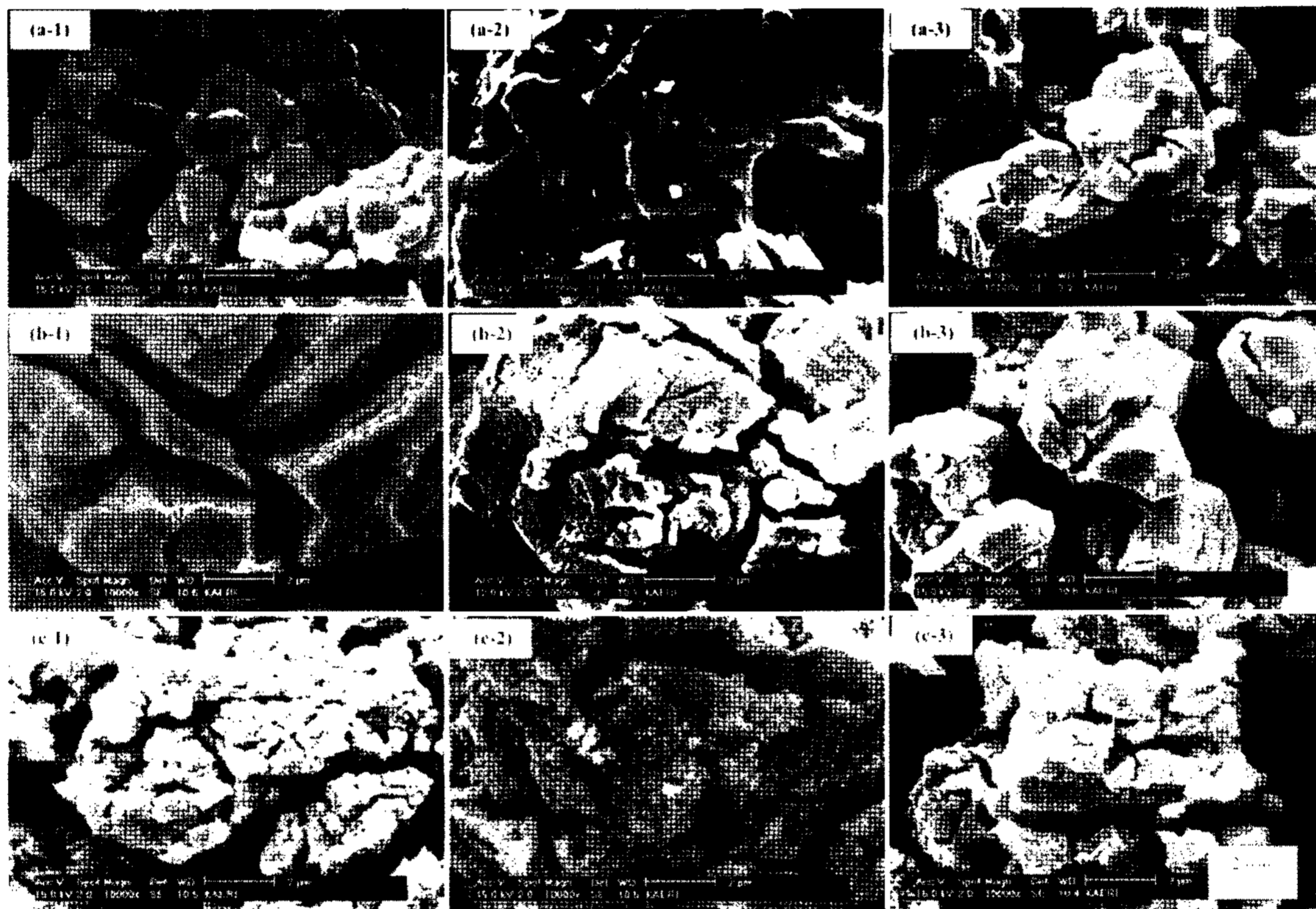


Fig. 2. Morphologies of the OREOX Powder with the Number of OREOX Cycles: (a-1) First Oxidation, (a-2) First Reduction, and (a-3) Third Reduction for UO₂; (b-1) First Oxidation, (b-2) Second Reduction, and (b-3) Third Reduction for 35,000 MWd/tU SIMFUEL; and (c-1) First Oxidation, (c-2) Second Reduction, and (c-3) Third Reduction for 60,000 MWd/tU SIMFUEL

with the characteristics shown in Table 2. The densities of the simulated DUPIC pellets increased linearly with an increasing sintering temperature and were in the range of 10.34 (95.9% TD) at 1,700 °C to 10.44 g/cm³ (96.8% TD) at 1,800 °C for 35,000 MWd/tU and from 10.32 (96.9% TD) to 10.39 g/cm³ (97.6% TD) for 60,000 MWd/tU, respectively. The sintered density of the pellets fabricated from the powder of 35,000 MWd/tU with a higher specific surface area was slightly higher than that of 35,000 MWd/tU. The sintered density of the pure UO₂ pellets, which were fabricated by pressing the powders at a pressure of 300

MPa followed by sintering at 1700 °C using the powder in Table 3, was measured as 10.52 g/cm³ (96.0% TD). Hence, the sintered density of the simulated DUPIC pellets was not significantly influenced by the amount of impurities.

The grain sizes of the simulated DUPIC pellets above a sintering temperature of 1,750 °C were constant; however, the amount of impurities in the SIMFUEL significantly affected the average grain size of the simulated DUPIC pellets, decreasing it with an increasing SIMFUEL burn-up. The average grain size of the simulated DUPIC pellets

for the 35,000 MWd/tU and 60,000 MWd/tU SIMFUELS after sintering at 1,700 °C was 7 and 4 μm, respectively.

The microstructures of the pure UO₂ pellet with an average grain size of 8 μm and the simulated DUPIC pellets for the 35,000 MWd/tU and 60,000 MWd/tU SIMFUELS

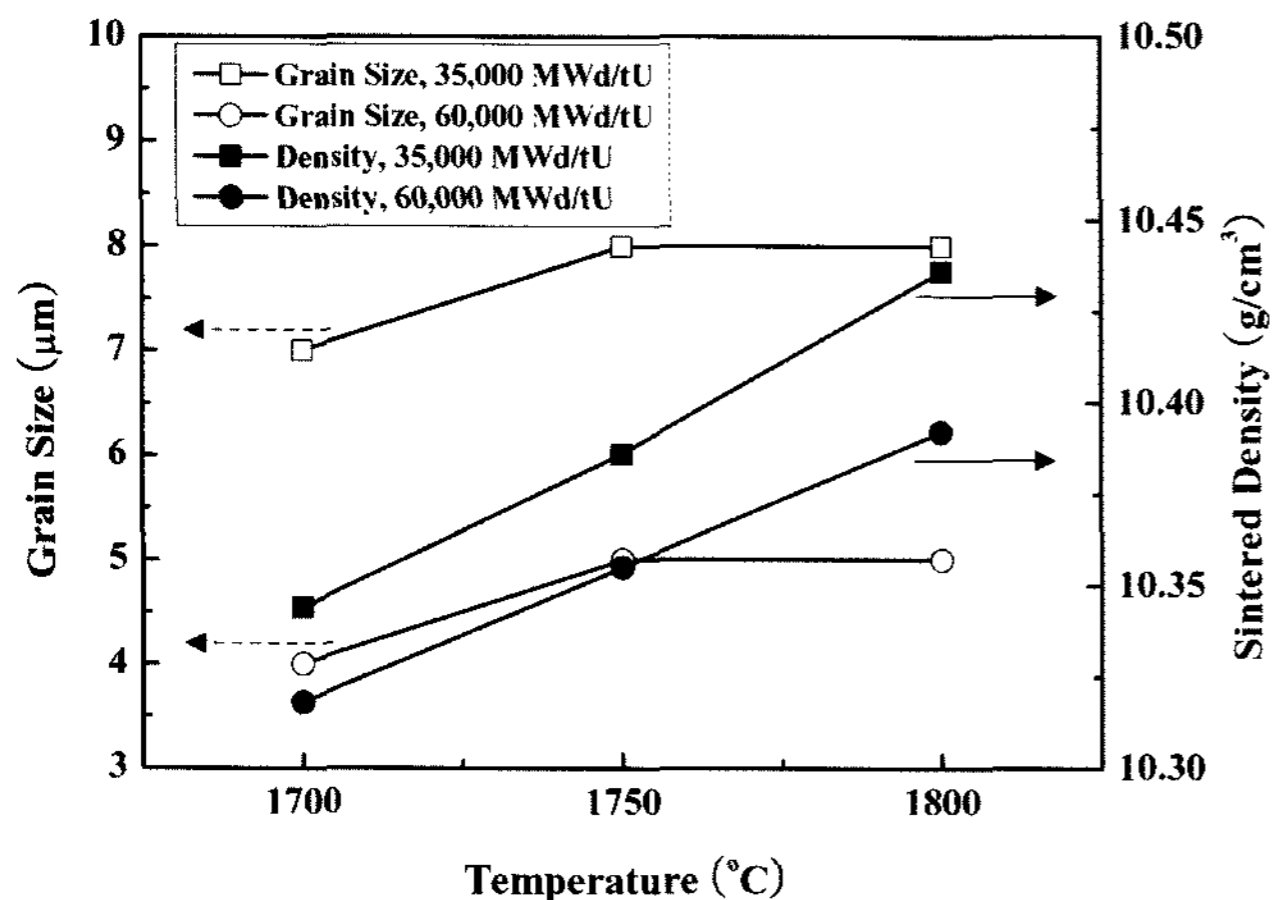


Fig. 3. Grain Size and Sintered Density of the Simulated DUPIC Pellets with the Sintering Temperature

after sintering at 1,700 °C are shown in Figs. 4(a), (b), and (c), respectively. The simulated DUPIC pellets show the same grain shape as the pure UO₂ pellet. However, for the simulated DUPIC pellets, metallic precipitates with a bright, spherical granular shape in the optical micrograph were primarily distributed at the grain boundary. With an increasing burn-up, more metallic precipitates were observed at the grain boundary, which indicates that the metallic precipitates would strongly inhibit the movement of a grain boundary and result in a reduced grain growth [13].

3.3 Effect of the Impurity (Fission Product) Groups

In order to investigate the influence of the impurity (fission product) groups on the grain growth, SIMFUEL and simulated DUPIC pellets with dissolved oxides (SS-UO₂), oxide precipitates (BaSrZrO₃, OP-UO₂), and metallic precipitates (MP-UO₂) with different burn-ups (Table 1) were fabricated. Table 3 shows that the grain size of the pellets of the three groups fabricated using the SIMFUEL pellet preparation process decreased in the order of SS-UO₂, OP-UO₂, and MP-UO₂ and also decreased with an increasing amount of the impurity group.

The DUPIC pellets simulated according to the impurity groups were fabricated by pressing the powders at a pressure of 300 MPa followed by sintering at 1700 °C

Table 3. Density and Grain Size of the Sintered Pellets with the Impurity Groups and Burn-Ups

Burn-up (MWd/tU)	Impurity group	SIMFUEL		Simulated DUPIC	
		Density (g/cm ³)	Grain size (μm)	Density (g/cm ³)	Grain size (μm)
35,000	SS	10.55	7.4	10.52	6.1
	OP	10.56	6.5	10.26	5.6
	MP	10.75	4.0	10.65	3.8
60,000	SS	10.52	7.0	10.48	5.4
	OP	10.54	6.0	10.20	4.6
	MP	10.77	3.1	10.77	2.8

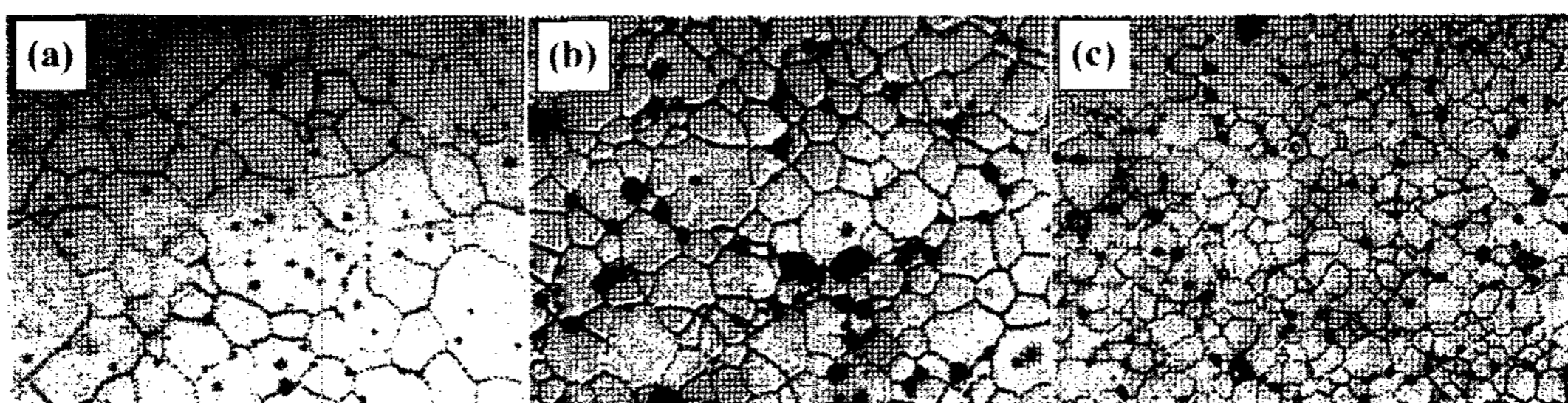


Fig. 4. Optical Micrographs of the Simulated DUPIC Pellets Sintered at 1700 °C: (a) 35,000 MWd/tU and (b) 60,000 MWd/tU

using the milled powder (Table 4) obtained from the simulated DUPIC powder preparation process. Also, the grain size of the simulated DUPIC pellets of the three groups decreased in the order of SS-UO₂, OP-UO₂, and MP-UO₂ as shown Table 3. For 35,000 MWd/tU, the grain size of the three groups' pellets was lower than that of the simulated DUPIC pellets. However, the grain size of the SS-UO₂ and OP-UO₂ pellets for 60,000 MWd/tU was higher than that of the simulated DUPIC pellets. The grain size of the SS-UO₂ pellets slightly decreased with an increasing amount of the dissolved oxide, whereas the grain sizes of OP-UO₂ and MP-UO₂ significantly decreased with an increasing amount of the oxide precipitates and the metallic precipitates, respectively. Therefore, the grain growth of the simulated DUPIC pellet was affected by the metallic and oxide precipitates from the impurity groups.

The metallic precipitates and oxide precipitates, as the secondary phase formed at the grain boundary during sintering of SIMFUEL green pellets, inhibited the movement of the grain boundary, resulting in a reduced grain growth. Also, stable metallic precipitates and oxide precipitates during the preparation of the simulated DUPIC fuel powder existed in a stable form and interfered with the grain growth of the simulated DUPIC fuel pellet. The limiting average grain size using the Zener pinning equation [13] was directly proportional to the diameter of the precipitates and inversely proportional to the precipitates volume fraction. It has been reported from the TEM observation by Lucuta et al. [14] that the diameter of the oxide precipitates was approximately 0.1 μm . In this study, the diameter of the metallic precipitates as seen from the observation of the SEM was approximately 1 μm . Although the diameter of the metallic precipitates was approximately 10 times larger than that of the oxide precipitates, the volume fraction of the metallic precipitates was approximately 15 times larger than that of the oxide precipitates [14]. Therefore, the grain growth in the simulated DUPIC pellet, using the Zener pinning equation, is expected to be governed

primarily by the metallic precipitates. The experimental results shown in Table 3 are in good agreement with those estimated by the Zener pinning equation.

An increase in the specific surface area of the simulated DUPIC powder was evaluated through the changes in the particle surface characteristics during the first and second reduction steps, as shown in Figs. 1 and 2, respectively. Thus, in order to investigate the influence of the impurity groups on the powder properties, simulated DUPIC powders using sintered pellets, as shown in Table 3, were prepared via the method described in Section 2.3.

Table 4 shows the average particle size and specific surface area of the powder prepared by three cycles of oxidation and reduction. The average particle size of the OREOX powders depended on the grain size of the raw pellets, excepting the OP-UO₂ powder, which showed the highest average particle size among the OREOX powders. Although the grain size of the OP-UO₂ pellet was lower than the MP-UO₂ pellet, the specific surface area of the OP-UO₂ powder for the same burn-up was higher than the MP-UO₂ powder. The high specific surface area of the OP-UO₂ powder appears to result from a separation of the oxide precipitate near the grain boundary from the UO₂ grain during the OREOX process. However, the specific surface area of the powders of the three groups was significantly lower than that of the powders from the 35,000 and 60,000 MWd/tU SIMFUELS (Table 2).

Figure 5 shows the results of the SEM observations for the OREOX powders obtained from the first and third reduction steps from the SS-UO₂, OP-UO₂, and MP-UO₂ pellets with a 60,000 MWd/tU burn-up. The particle surfaces in the three groups of the powders obtained by the first reduction step are very coarse, unlike the powders obtained by the 35,000 and 60,000 MWd/tU SIMFUELS (Figs. 2 (b-2) and (c-2)). Also, after the third reduction step, morphologies with sponge-like particle shapes with internal pores (Figs. 2(b-3) and (c-3)) were not observed in the powders of the three groups.

Table 4. Characteristics of the OREOX and Milled Powders with the Impurity Groups and Burn-Ups

Burn-up (MWd/tU)	Impurity group	Average particle size (μm)		Specific surface area (m^2/g)	
		As produced	After milling	As produced	After milling
35,000	SS	8.8	0.7	1.78	3.00
	OP	10.1	0.8	1.89	3.53
	MP	6.7	0.5	1.82	2.96
60,000	SS	8.3	0.6	1.89	3.31
	OP	8.7	0.8	2.88	4.22
	MP	6.3	0.4	2.78	3.64

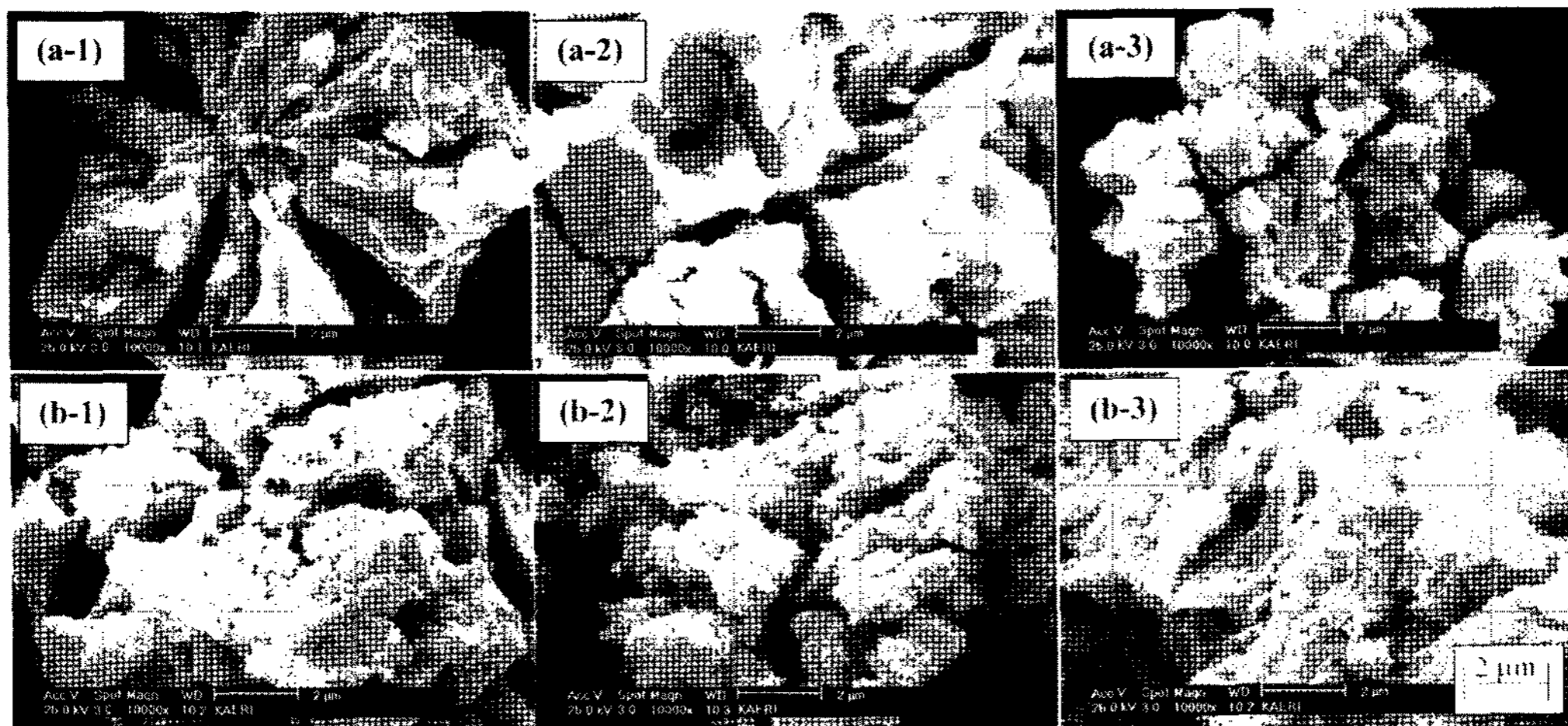


Fig. 5. SEM Morphologies of the Powders Obtained from the Reduction Steps in the OREOX Cycles: (a-1) First SS-UO₂, (a-2) OP-UO₂, (a-3) MP-UO₂ for the First Reduction; and (b-1) First SS-UO₂, (b-2) OP-UO₂, (b-3) MP-UO₂ for the Third Reduction

In order to interpret this phenomenon in terms of a phase change, the powders obtained after the oxidation step were analyzed by XRD. Figure 6 shows the XRD patterns of the powders produced from the first oxidation step. The oxidation of the SS-UO₂ and OP-UO₂ pellets with a small amount of dissolved oxides resulted in a single product of an (U,SS)₃O₈-type orthorhombic phase.

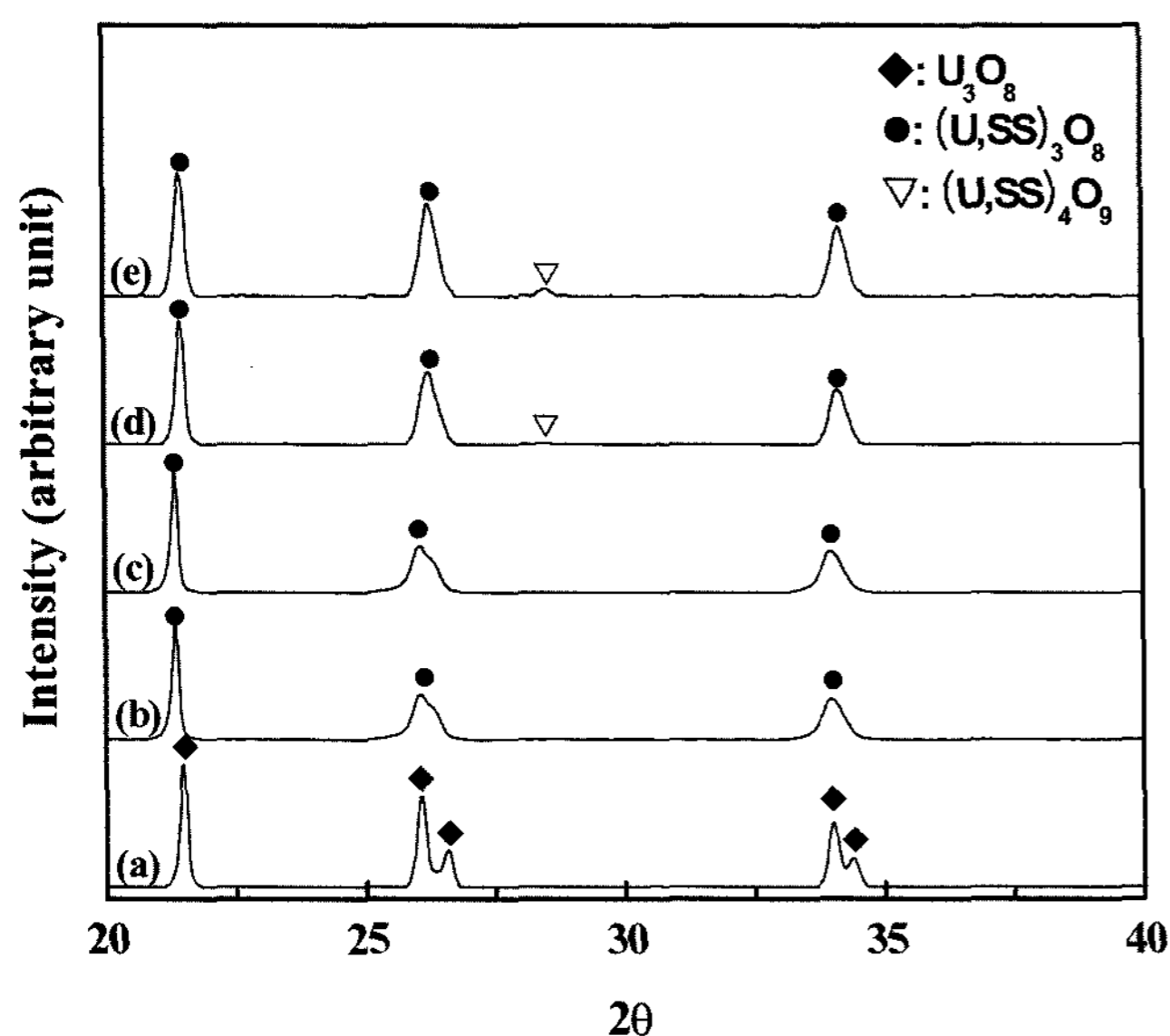


Fig. 6. XRD Patterns of the Powders Produced by the First Oxidation Step: (a) UO₂, (b) SS-UO₂, (c) OP-UO₂, (d) 35,000 MWd/tU SIMFUEL, and (e) 60,000 MWd/tU SIMFUEL

However, the oxidation product of the 35,000 and 60,000 MWd/tU SIMFUEL pellets was an (U,SS)₃O₈-type orthorhombic phase with a small (U,SS)₄O₉-type fluorite phase, which increased with an increasing burn-up. During the reduction step, a volume expansion of 2% for U₄O₉ and a volume reduction of 23% for U₃O₈ was caused by the transformation of the U₄O₉ (TD: 11.2 g/cm³) and U₃O₈ (TD: 8.4 g/cm³) phases, respectively, into a UO₂ phase (TD: 10.96 g/cm³). Therefore, the rougher particle surfaces appear to be caused by the difference in the volume change between the two phases formed on a single particle.

4. CONCLUSIONS

DUPIC is a fuel recycling technology that fabricates CANDU fuel directly from spent PWR fuel material using a dry process. Considering the trend for extended burn-up in a PWR fuel, it is necessary to analyze the effect of spent fuel burn-up on the fuel fabrication process. The fabrication characteristics that improve the density and grain size of the DUPIC fuel in relation to its fuel performance were evaluated experimentally using SIMFUEL as a surrogate for actual spent PWR fuel due to the high radioactivity of spent fuel.

In this experimental study, SIMFUELS with burn-ups of 35,000 MWd/tU and 60,000 MWd/tU were used to investigate the influence of fission products' contents as impurities on the fuel powder properties and on the density and grain size of the simulated DUPIC pellets. SIMFUELS were prepared using a mixture of UO₂ and surrogate metallic oxides for fission products. The specific surface area of the powders milled after an OREOX treatment

increased and resulted in sintered pellets with a higher than 95% TD regardless of the impurity contents. However, the grain size of the sintered pellets decreased with an increase of the amount of impurities. The specific surface area of the OREOX powders increased with an increase of the impurities due to the dissolved oxides in UO₂ from the impurity groups. The grain size of the sintered pellets significantly decreased due to the metallic and oxide precipitates groups.

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