The Effect of $\text{U}_3\text{O}_8$ Powder Re-Sintering on the Meat of Dispersion Fuel Elements MTR Fuel Plate Type

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Received: 1/6/2014 Accepted: 1/1/2015

ABSTRACT

The quality of nuclear fuel depends on the control of many parameters to satisfy the design requirements and specifications. The fuel plate is the main component of the MTR fuel element, used in research reactors. Controlling the fuel particle size during fuel plate manufacture is one of the important parameters affecting the quality of the produced fuel element. Fuel particle size has a direct impact on the fuel element performance during its irradiation in the reactor. This includes the degree of irradiation damage of the fuel element and its capacity to attenuate fission gases to avoid swelling. Therefore, the design specifies the limitations on the particles size of the $\text{U}_3\text{O}_8$ powder. In general, the particle size should be $< 90$ µm. Up to 50% of the fuel powder can have particle size 45 µm. The powder with particle size 45-90 µm is termed “specified powder” while that with particle size $< 45$ µm is termed “fine powder”. Fuel powder used in the plate manufacture is produced through a series of steps. However, it may happen that the fine powder produced in a batch is more than 50%. If this takes place during the production of many batches, there may be an accumulation of fine powder which is not used in the fuel plates production, i.e., rejected powder. The present work explores the possibility of reprocessing the fine powder through heat treatment (re-sintering). Re-sintering was carried out at 1400°C for 12 hr. The specified powder produced from this process was used in the manufacture of fuel plates. The produced plates were examined using non-destructive and destructive tests. Examinations showed that the produced plates suffer from the “dogboning” phenomenon which would lead to the rejection of these plates. This was related to the morphology of the produced powder. Examination showed that the shape of the powder produced after re-sintering is much distorted compared to the normally produced powder.

Key Words: MTR-Type / Uranium Oxide ($\text{U}_3\text{O}_8$) Powder / Nuclear Fuel/Dogboning, Dispersion Fuel Plate/ Fishtail Defect.

INTRODUCTION

The MTR type of nuclear fuel is classified as a dispersion type nuclear fuel. It is used in research and test reactors. It consists of fuel plates assembled in a fuel assembly or fuel element. A dispersion fuel plate is one in which a powdered fuel is dispersed within an aluminum powder matrix with the core being completely enclosed or clad with aluminum. The powdered fuel itself can be $\text{U}-\text{Al}$ alloy, $\text{U}_3\text{O}_8$, $\text{U}_3\text{Si}_2$ and $\text{U}_3\text{Si}$.

The fuel plate type production raw materials are uranium hexafluoride ($\text{UF}_6$, 19.7±0.2 % $^{235}\text{U}$), pure aluminum powder, and nuclear grade 6061 aluminum alloy in sheets, bars, and rods. All these

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components are processed through series of the manufacturing, inspection, and quality control plan to produce the final specified MTR-type fuel elements (1).

The processes used for fabricating all these compounds into fuel elements are essentially the same (2). They consist of the following major operations: core preparation, plate fabrication and cladding, plate inspection, element assembly, attachment of hardware components, and final inspection. All the plate fabrication procedures make use of the picture-frame, hot-rolling technique, Fig. 1. The major components used in such an operation are two flat covers, a frame of the same width and length, with one or two cavities, and the fuel fillers which are the same size as the frame cavity. Hot rolling such a billet bonds all contacting surfaces and sizes the plate as shown in Fig. (2).

![Fig. (1): Picture-Frame technique: A) Assembling B) After welding](image1)

![Fig. (2): Schematic illustration of the rolled fuel plate.](image2)
In the present work we are concerned with the effect of particle size and morphology, so the discussion will be almost limited to the core preparation step.

1.1. CORE PREPARATION

The U$_3$O$_8$ has been made for nuclear reactors using different techniques. In the work described in this paper UF$_6$ is dissolved in water and then ammonia is added to precipitate ammonium-diuranate. The precipitate is calcinated at 800°C to convert it to U$_3$O$_8$, which is sized and calcinated again at 1400°C. This process increases the density through a sintering process.

**Grinding:**

The U$_3$O$_8$ powder is ground, using roll grinding, to produce an acceptable particle size distribution for the powder metallurgy process. Powders resulting from the grinding process normally contain about 40% fine particles (< 45 microns in diameter).

**Blending:**

Ground U$_3$O$_8$ and Al powder is blended to get a homogeneous mixture of the oxide fuel in Al. The speed (rpm) of the bending machine and the blending time are adjusted to achieve the required homogeneity.

**Compaction:**

After blending, the powder is placed in a special die for compaction. Compaction is carried out at room temperature and the pressure as well as the compaction duration is adjusted to get the required specifications. The product of this process is called billet or compact.

1.2. EFFECT OF FUEL POWDER MORPHOLOGY

Controlling the fuel particle size during fuel plate manufacture is one of the important parameters affecting the quality of the produced fuel element. Fuel particle size has a direct impact on the fuel element performance during its irradiation in the reactor. This includes hot spots within the plates, the degree of irradiation damage of the fuel element and its capacity to attenuate fission gases to avoid swelling.

Avoiding hot spots within the plates during operation is of a major concern$^{(3)}$. Hot spots can result in plate melting. Factors, in plate fabrication process, affecting hot spots, and which must therefore be controlled, are uranium loading, uranium distribution, core thickness, and quality of heat-transfer bonds. The most serious of these is the control of core thickness. If the properties of the core and frame material are not closely matched, an end phenomenon known as dogboning results (see Fig. 3). Dogboning is the core thickening that occurs at the ends of the core during roll bonding. This core thickening is of concern for two reasons. First is the thinning of the cladding over the dogbone. Secondly, the higher fuel concentration could result in excessive temperature (hot spots) during irradiation. The problem is circumvented by better matching of the properties of the frame and cores by varying the alloy or the heat treatments or by using small tapers at the core ends so less starting material is available in these areas$^{(3)}$. 

96
During fuel irradiation in the reactor many fission products are produced. Therefore, a certain volume of voids should exist inside the fuel plate to maintain these products inside the plates to avoid plate swelling. These voids can result through controlling the $\text{U}_3\text{O}_8$ powder size and its morphology. In MTR fuel plate type the voids should be about 12%\(^4\).

The degree of irradiation damage can be explained with the help of Fig. (4)\(^5\). In the fission process two new nuclides are formed (fission products). More than 80% of the total energy released by fission is imparted to the two fission fragments as kinetic energy. These large, extremely energetic, particles travel only a few microns, but in so doing, seriously disrupt the crystal lattice while dissipating their energy as heat. The Figure shows an ideal dispersion of uniform, spherical, fuel particles, sized to permit a reasonable amount of matrix metal between particles to remain undisturbed by fission fragments as shown in Fig. (4-A)

\textbf{Fig. (3)}: Examples of dogboning in dispersion fuel\(^3\).

\textbf{Ideal microstructure}

\textbf{Fig. (4)}: Two Causes of Overlapping Fission Damage Zones: (B) smaller fuel particle size and (C) higher fuel volume fraction\(^5\).
For a given volume fraction of fuel particles, the smaller the particle size, (Figure 4-b), the more likely will overlapping of the damaged fission recoil regions occur. On the other hand, for a given fuel particle size, the greater the total volume fraction of fuel particles in dispersion, the more likely will overlapping of the damaged fission recoil regions occur, Figure (4-c).

Therefore, specifications for fuel particle size and morphology in general are set to avoid the above mentioned concerns. In general, $U_3O_8$ the particle size should be $<$ 90 µm. Up to 50% of the fuel powder can have particle size $<$ 45 µm. The powder with particle size 45-90 µm is termed “specified powder” while that with particle size $<$ 45 µm is termed “fine powder”. Fuel powder used in the plate manufacture is produced through a series of steps and the fine powder is normally about 40% as mentioned above. However, it may happen that the fine powder produced in a batch is more than 50%. If this takes place during the production of many batches, there may be an accumulation of fine powder which is not used in fuel plate production, i.e., rejected powder.

Many parameters can lead to increasing the fine powder ratio, such as milling time after calcinations processes and fluorine concentration during the calcinations due to mal functioning of the gasses-extraction system. The furnace conditions, particularly the temperature, are important both for the fluorine content and the specific surface of the powder produced.

Since the $U_3O_8$ powder is a valuable material, attempts to use the rejected powder should be done. The present work explores the possibility of reprocessing the fine powder through heat treatment (re-sintering) to get specified powder.

**EXPERIMENTAL PROCEDURES**

2.1. Production of $U_3O_8$ Powder:

The starting material for the present work was produced during the normal production of $U_3O_8$. The $U_3O_8$ production starts by reacting UF$_6$ gas with water to form uranyl fluoride (F$_2$UO$_4$). Then ammonia solution is added in a process which leads to the precipitation of ammonium-diuranate (ADU). The ADU obtained is calcinated at about 800°C which is converted to $U_3O_8$ in an oxidizing atmosphere according to the next reaction:

$$3[(NH_4)_2U_2O_7(solid)] \rightarrow 2(U_3O_8) + 4(NH_3) + 5(H_2O) + N_2(gas)$$

The product ($U_3O_8$) is then milled, and the smaller particles are agglomerated. Afterwards it is sized to less than 150 microns using sieve shaker. Heat-treatment (sintering) at 1400°C is used to obtain the high density $U_3O_8$ required. Subsequently the material is treated in a mortar, milled and sieved to specified ($U_3O_8$) particle size, Fig. (5), which in between 90 to 45 microns, and fine $U_3O_8$ particles which are less than 45 microns.

The fine powder used in the present work was accumulated from some batches in which the fine powder was more than 50% of the match.
2.2. Resintering of Fine Powder:

The starting material in this study is the fine U₃O₈ powder, Fig. 6. This fine powder was subjected to re-sintering process at 1400°C for 12h. The fine powder was agglomerated after re-sintering at 1400°C to form specified powder (more than 45µ). The agglomeration process is due to the increase in surface energy. After milling and sieving the re-sintered powder was classified into specified and fine powders according to the particle size as above.
The specified powder, Figure (7), was then used to produce fuel plates; it was mixed with about 50% fine powder. Pure Al powder was then added and the mixture went through the blending process. The blended powder was pressed to form compacts. Each compact was enclosed in frame and covered with two cover plate as shown in Figure (1). The last product (sandwich) is subjected to hot rolling to obtain fuel plate as shown in Figure (2).

The produced plates went through the normal quality control inspections until it was cut to the final dimensions. To assure that the plates are conforming to the specified dimensions, especially the thickness of the cladding material, destructive testing is carried out. In this test plates were cut as shown in Figure (8). Four specimens, 1.5 cm x 3 cm each, are cut along the plate length. Specimens 1&2 are used to measure the meat and clad thickness at the ends of the plate, while specimens 3&4 are used for measurements at the plate middle. After mounting, polishing and etching, the sections were inspected with the optical microscope to measure the fuel core and the cladding thicknesses at the corresponding locations.

Fig. (8): Sections for the cladding and core thickness measurements.

Working with radioactive materials involves always health hazards. Although U is only an alpha emitter, but its presence in the form of powder, e.g., $\text{U}_3\text{O}_8$, represents the danger of human intake of airborne activity by ingestion or inhalation. The present study was done in a careful way in controlled area and inside sealed glove boxes, under slightly negative pressure, to avoid these problems.

RESULTS AND DISCUSSION

The particles in real dispersion-fuel systems are far from the ideal system shown in Fig.(4)\(^6\). Particles in real systems are quite irregular and their sizes will range from a few microns to several hundred microns. This is because the handling of powders invariably fines by attrition, especially in mixing or blending procedures. Also, the hot-working processes utilized causes clustering, fragmentation, and stringering of the particles. All that can be done with real dispersions is to process them in a way to make them as nearly ideal as possible by controlling the working parameters.

Figures (5) and (6) show the specified powder and the fine powder, respectively. Figure (7) shows the powder produced after re-sintering of the fine powder shown in Figure(6). Comparing figures (5) and (7), it is clear that resintering resulted in a much distorted powders shape.
Studies in the field of U$_3$O$_8$ are few while most of uranium-oxide-sintering studies are concerned with UO$_2$. Even in most of the cases where the sintering of U$_3$O$_8$ is studied, it is combined with UO$_2$. It is possible that the reason is that UO$_2$ is used in most of the working nuclear power reactors, while U$_3$O$_8$ is used only in some of the research reactors. Moreover in the most recent research reactor fuel, U$_3$O$_8$ is replaced by uranium silicides.

One of the important steps during the manufacture is the heat-treatment of the U$_3$O$_8$ powder. K.W. Song et al.(7) found that heat treatment at 1000-1500°C produce crystalline growth and enhance bonding among U$_3$O$_8$ particles. Also, it causes an increase in U$_3$O$_8$ powder density.

Previous studies(8, 9) showed that the characteristics of U$_3$O$_8$ particles formed by heat treatment at 1400°C of U$_3$O$_8$ produced from the calcinations of ADU were dependent on the chemical composition and chemical formula of ADU specially the content of fluorine ions as well as operation parameters during precipitation of ADU, the temperature of calcinations furnace, and the extraction system of decomposition gases during the calcinations process.

In powder metallurgy (PM), the powder size, shape and ductility dictate its ability to be fabricated into useful parts. The shape of the powder affects the strength of pressed compact and the degree of segregation of bended powders. The spherical powders, with little or no frictional interaction do not hold together in compact or resist segregation as well as other shapes. Ductile materials will readily deform giving good adhesion in pressed compact (10).

J. Williams et al.(11) studied the effect of particle size on the sintering behavior of UO$_2$. Compacts of these two powders pressed at 15000 psi and sintered in argon at 1450°C for two hrs gave densities of 8 g/cm$^3$ and 10.2 g/cm$^3$ respectively. They indicated that uranium dioxide of a sufficiently small particle size would be oxidized at room temperature, the rate of oxidation being greater the finer the particle size and thus the oxide of higher oxygen content would have the smaller particle size.

Kun Woo Song et al.(8, 9) studied the sintering process for U$_3$O$_8$ powder made by oxidation of defective UO$_2$ pellets was mixed with UO$_2$ powder, and Nb$_2$O$_5$ was then added. The results showed that the sinterabity of U$_3$O$_8$ powder, which is the oxidized product of defective UO$_2$ pellets, is enhanced by additional thermal treatments. The particle size of the U$_3$O$_8$ powder is shown to be dependent on the oxidation temperature. As this temperature increases, the stress involved in the oxidation is so relieved that the related pellet pulverization is significantly suppressed and the particle size of U$_3$O$_8$ becomes larger. They found that the powder morphology of the sintered UO$_2$ is different from that of the sintered U$_3$O$_8$. The UO$_2$ powder has a round shape and a smooth surface while the U$_3$O$_8$ particles have angular shapes, Fig.9. It is to be noted that this U$_3$O$_8$ was produced by the oxidation of sintered UO$_2$. So it may be appropriate to compare the product of this U$_3$O$_8$ with the resintering results in the present study, Figure (7).
I.V. Petrov and V.V. Basov (12) investigated the influence of the initial state of powder, pore-forming additions, and U$_3$O$_8$ on the microstructure and strength of fuel pellets. They found that the initial state of uranium dioxide powder has no effect on the density, microstructure, and strength of pellets. Pore-forming agents and the U$_3$O$_8$ used in fabrication lower the pellet strength because their particles are not spherical. To increase pellet strength, it was recommended that U$_3$O$_8$ be subjected to special processing to spheroidize the particles before mixing with uranium dioxide powder.

**Core and Cladding Thickness:**

Figure (10) shows the results of metallographic examination of a plate produced using normal powder, while Fig.11 shows those of a plate produced using the re-sintered powder. The two plates are produced with the same procedure and using the same values for the parameters during all the production steps. Figure (10) shows that, in the case of the normal powder, the core and cladding material thickness were about 0.7 mm and 0.4 mm, respectively, along the whole plate length. These dimensions agree very well with the required specifications. On the other hand, Figure (11) shows that in the case of the re-sintered powder, while the core and clad thickness in the middle of the plate are also about 0.7 mm and 0.4 mm, respectively, they were about 0.9 and 0.3 at the plate end.

These results show that the plate produced using the re-sintered powder suffers from dogboning. The core thickness at the plate ends is more than that at the plate middle while the reverse is observed for the clad thickness at these locations. This is to be compared to the almost uniform core and clad thickness in the case of the plate produced the normal powder. This could be attributed to the difference in the morphology between the normally produced powder (Fig.5) and the resintered powder (Fig.7).

The cladding thickness can be affected by parameters, such as mechanical properties of aluminum alloys used as cladding for fuel core, characteristics of the U$_3$O$_8$ powder used as fuel core, aluminum powder used as dispersed matrix, roll mill diameter and speed. The irregular angular shape of the powder was shown to decrease the flowability of the powder compared to those of round shape (8-10). Therefore it
could be concluded that the characteristics of the re-sintered powder was behind the appearance of the dogboning in the produced plates. This results in the agglomeration of the fuel particles at the plate ends and thinning of the clad at these locations, which would lead to the rejection of the plates.

Fig. (10): The shape of normal fuel core (U\textsubscript{3}O\textsubscript{8} + Al) and AA6061 clad (using normal U\textsubscript{3}O\textsubscript{8} powder), A: plate end, B: plate middle.

Fig. (11): The shape of the fuel core (U\textsubscript{3}O\textsubscript{8} + Al) and AA6061 Clad. [(1, 2 at the plate ends), (3,4 at the plate meddle)]
CONCLUSION

The particle size of the U₃O₈ powder used in the fabrication of the research reactor fuel plates has a strong effect on the performance of the fuel during its usage in the reactor. Therefore, the design specifies the limitations on the particles size of the U₃O₈ powder. In general, the particle size should be < 90 µm. Up to 50% of the fuel powder can have particle size 45 µm. The powder with particle size 45-90 µm is termed “specified powder” while that with particle size < 45 µm is termed “fine powder”. Fuel powder used in the plate manufacture is produced through a series of steps. However, it may happen that the fine powder produced in a batch is more than 50%. If this takes place during the production of many batches, there may be an accumulation of fine powder which is not used in the fuel plates production, i.e., rejected powder.

In the present work the possibility of reprocessing U₃O₈ fine powder through heat treatment (re-sintering) was explored. Re-sintering was carried out at 1400°C for 12 hr. The specified powder produced from this process was used in the manufacture of fuel plates. The produced plates were examined using non-destructive and destructive tests. Examinations showed that the produced plates suffer from the “dogboning” phenomenon (increase the core thickness and decrease of the clad thickness at the plate end) which would lead to the rejection of these plates. At the plate end, the plate core thickness was about 0.9 mm instead of 0.7 mm and the clad thickness was 0.3 mm instead of 0.4 mm, due to using re-sintered U₃O₈ powder. It is concluded that these changes are due to the change in U₃O₈ powder morphology as a result of the re-sintering process.

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