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An Alternative Method to Produce U_3O_8 Powder for MTR Fuel Elements

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Abstract. This work describes an alternative method of obtaining U_3O_8 powder for MTR type fuel elements by AUC calcinations and sintering at specific conditions. The obtained U_3O_8 powder is suitable to be used as a nuclear fuel. It could be checked by analyzing its physical and chemical properties and those ones resulting from the obtained fuel plates. Metallographic tests were carried out in those fuel plates in order to determine the U_3O_8 particle size distribution by image analysis. The main conclusions show the applicability of the method, owing to the similar powder and fuel plates properties, as compared to the conventional method.

Introduction

Among the objectives of IPEN-CNEN/SP that have been carried out since its foundation in 1959, we can find the research and development concerning the production of many radioisotopes employed by industry and nuclear medicine. The need to supply the IEA-R1m with fuel elements and the difficulty of acquiring the enriched U_3O_8 powder to produce those fuel elements forced IPEN to concentrate efforts to develop methods to produce the enriched U_3O_8 powder starting from ADU, instead of importing it.

The first method for U_3O_8 powder production adopted by IPEN-CNEN (employed until 1989) had many difficulties, mainly the time consuming manual grinding of the sintered pellets. This way, a great quantity of U_3O_8 sintered fines ($< 44\ \mu m$) was produced (the maximum content of fines allowed by specification requirements is 20%) [1] and had to be reintegrated to the process as scrap (with complex dissolution and precipitation operations).

After 1989, a new method for producing U_3O_8 powder was developed and introduced at IPEN-CNEN [2]. This method, also starting from ADU, was simplified and based on grinding of calcined pellets (current method, #1). All the exceeding content of as calcined U_3O_8 fines produced (related to the maximum content specified) could easily be reintegrated to the process by repressing.

In spite of the better operational characteristics of the current method of production, the development of an alternative method (proposed method, #2) without any kind of granulation step was started [3]. As a premise, this method should eliminate the difficult

pressing and granulation steps and besides, should also use AUC (Ammonium Uranyl Carbonate) as the raw material. As AUC [4] is used to produce UO_2 powder for PWR type fuels, the use of the same material – AUC – as U_3O_8 powder precursor is important in order to standardize the UF_6 conversion process. Fig. 1 presents the current and proposed methods to produce U_3O_8 powder.

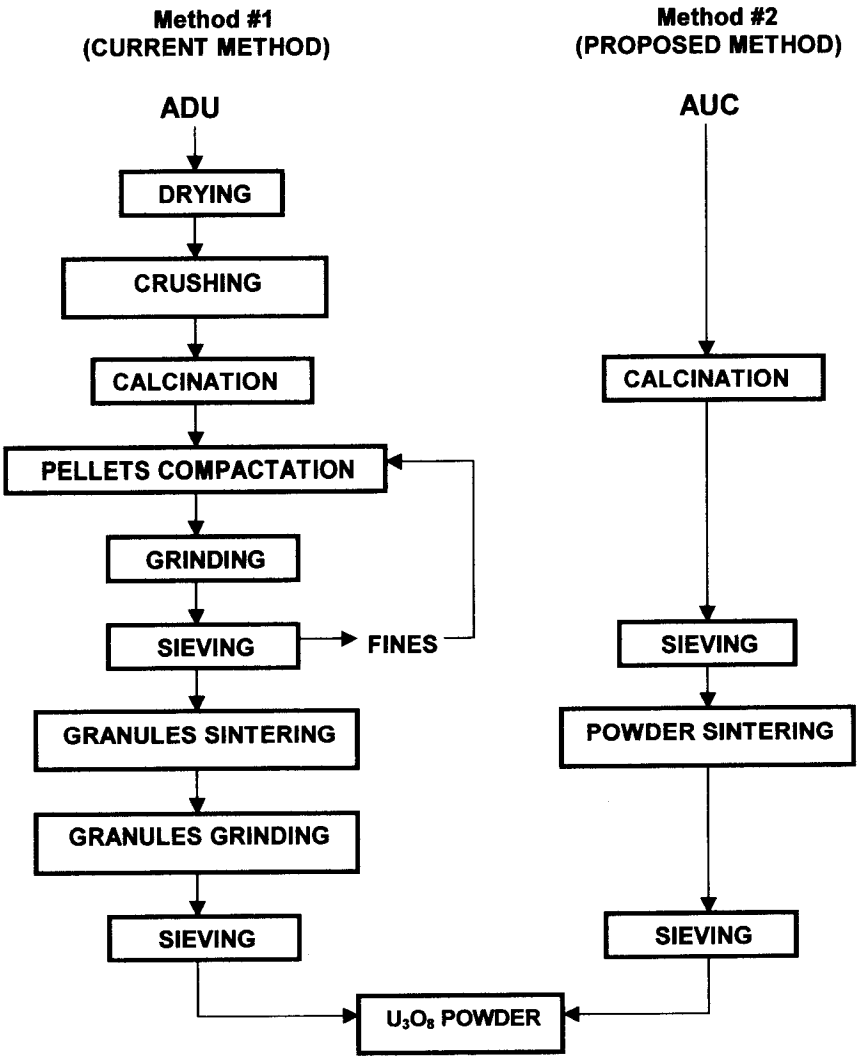


Fig. 1 – Methods of obtaining U_3O_8 powder as specified.

Experimental

Initially, experimental lots of U_3O_8 powder (300 g) were processed to study the production process parameters in order to determine the condition suitable to process a full-size lot (6

Kg AUC). The main process parameter studied was the calcination temperature, as follow:

- Calcination: 600ºC-3h, 700ºC-3h, 800ºC-3h.
- Sieving: 44 μm < D < 177 μm (A) and D < 44 μm (B) – 100g-15min.
- Sintering (A) and (B) separately: 1400ºC/6h and 1400ºC/24h.
- Sieving: 44 μm < D < 88 μm (A) and D < 44 μm (B) - 100g-15min.
- Analysis in terms of Specific Surface Area, Density and Impurity Content.

After the definition of the best calcination condition, a full-size lot of AUC (6Kg) was processed, as follow:

- Calcination: 600ºC-3h.
- Sieving: the same way that experimental lot.
- Sintering (A) and (B) separately: 1400ºC/6h.
- Sieving: 44 μm < D < 88 μm (A) and D < 44 μm (B) - 250g-60min.

The behavior of the U_3O_8 particles in the subsequent fuel plate fabrication process was verified by rolling the fuel plates # 05/99 and #07/99, with 29 wt% (total of fines generated) and 0 wt% fines, respectively.

The influence of the U_3O_8 fines content on the microstructure of the fabricated fuel plates was analyzed by means of image analysis. The obtained results were compared to the ones obtained from a standard fuel plate (#432), which was fabricated with U_3O_8 powder produced according to the traditional method (current method, #1).

Results and Discussion

The characteristics of the produced U_3O_8 powder and fuel plates are shown in Tab. 1. According to this table, the results of specific surface area, density, and impurities content for the U_3O_8 powder fabricated according to the proposed method (method #2) satisfy the specifications [1]. As can be seen from these results, the only feature that does not comply with the specification is the total fines content. The cladding thickness, fuel core length and its residual porosity were measured and also agree with the specified values. The results indicated that the fuel plates fabrication behavior was very similar to the one observed in fabricating the standard fuel plate #432.

Fig. 2 shows the particle size distributions of the meats into the two fuel plates obtained by the proposed method (#05 and #07/99). These distributions were compared to the distribution of the standard fuel plate (#432). Each curve was constructed based on the results acquired from the Quantikov System [5] and represents the measuring results of approximately 12.000 fissile particles dispersed in the aluminum matrix. According to Fig. 2, the U_3O_8 particle mean diameters of the fuel plates #05/99 and #07/99 (proposed method) are slightly smaller than the #432 fuel plate mean diameter. Fig. 3 also confirm that. Besides, the initial content of fines of the fuel plates obtained from the proposed method does not affect the disposition of the distributions. This fact can also be checked by observing the micrographs presented in Fig. 3 (a) and (b). The U_3O_8 particle size distribution after the fuel plate rolling just appears to be dependent on the original morphology of the U_3O_8 particles or, in other words, on the adopted U_3O_8 powder obtaining method, as shown by the SEM micrographs presented in Fig. 4 (a) and (b). The morphologies of the U_3O_8 particles of the powders obtained by both methods (method #1 and #2) were quite different. The U_3O_8 particles originated by the proposed method #2

seem to be constituted by a number of smaller primary particles that are aggregated in the powder sintering step of powder production. Probably these U_3O_8 particles are crushed during the fuel plates rolling, which causes the size distribution mean to be smaller.

Table 1 - Physical and chemical properties of U_3O_8 sintered powder and fuel plates

	U_3O_8 sintered- (ADU)- Current Method	U_3O_8 sintered – (AUC) – Proposed Method	Specified Value
Specific Surface Area (m^2/g)	< 0.1	< 0.1	< 0.1
Density (g/cm^3) Hg Porosimeter	8.2	8.2	> 8.0
Fines <44μm (%)	20	29	20
Impurity Content (ppm)			
Cd	0.3	0.3	Max. 0.1
B	0.1	0.1	Max. 2
P	<55	<55	Max. 250
Fe	100	120	Max. 250
Cr	30	14	Max. 200
Ni	2	6	Max. 200
Mo	<2	<2	Max. 250
Zn	<10	<10	Max. 250
Si	50	90	Max. 250
Al	60	60	Max. 250
Mg+Ca	30	20	Max. 200
Mn	2	5	Max. 250
Pb	1	1	Max. 250
Sn	5	10	Max. 250
V	<3	<3	Max. 250
Cu	5	5	Max. 250
Co	<10	<10	Max. 10
F+Cl	<30	<30	Max. 350
	Current Method – Fuel plates # 05/99 //// 07/99	Proposed Method Fuel plate #432	Specified Value
Central Coating thickness (mm)	0.35 //// 0.35	0.35	0.34±0.01
Terminal Coating thickness (mm)	0.29 //// 0.29	0.29	0.28±0.02
Core length (mm)	599.5 //// 597.5	590.5	600±10
Residual porosity (% Volume)	7.18 //// 7.69	7.41	7.50±0.50

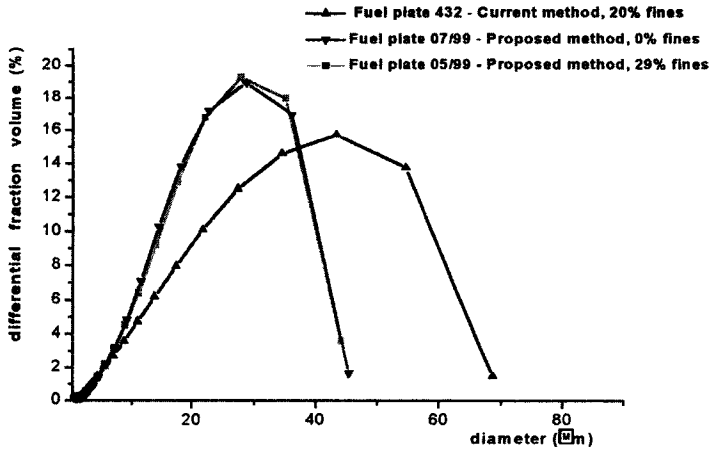
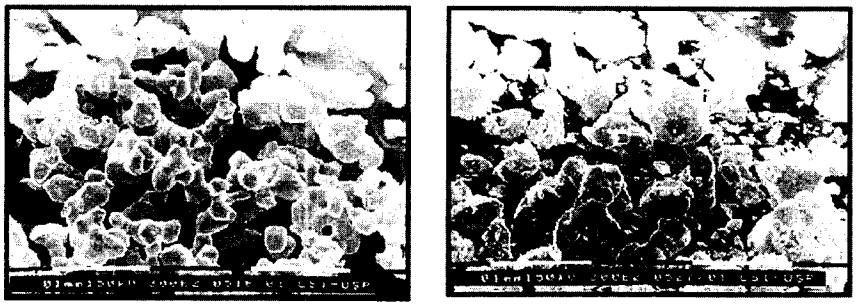


Fig. 2 – U_3O_8 particle size distributions of the considered rolled fuel plates.

(A) *(B)* *(C)*

Fig.3 – U_3O_8 particles after rolling. Fuel plates # : (A)-05/99 - 29% fines; (B)-07/99 - 0% fines (proposed method) and (C)-432 - 0% fines, current method. (550X).



(A) *(B)*

Fig. 4 – SEM micrographs of U_3O_8 particles before rolling from: (A) ADU and (B) AUC. (300X).

In spite of the bigger fines content (29 wt%) resulting from the proposed method, published data [6] enable the use of U_3O_8 powder with high fines content. Irradiation tests demonstrated a good irradiation performance of fuel plates containing up to 50 wt% U_3O_8 fines (< 44 μm). On the other hand, unexpectedly there are no references on the literature specifying the particle size distribution in the fabricated fuel plate meat. The results obtained in the present work demonstrated that the initial content of fines in the U_3O_8 powder do not affect the particle size distribution in the rolled fuel plate meat.

Conclusions

The U_3O_8 powder starting from AUC (proposed method) and the corresponding fuel plates have the physical and chemical characteristics in agreement with the demanded specifications to be employed as MTR type fuels. The chosen processing condition is: calcination at 600° C-3h and sintering at 1400°C-6h. The sintering for 24 hours did not modify the U_3O_8 powder morphology and size.

The number of processing steps of the proposed method is smaller than the conventional method. In spite of the higher U_3O_8 powder fines content produced by the proposed method (29 wt%), it can not be discarded, as irradiation tests have demonstrated.

The results show that the initial fines content before rolling does not modify the particle size distribution of the fabricated fuel plates. It is just dependent on the U_3O_8 powder obtaining method. However, an irradiation test of a fuel plate produced according to the proposed method (with 29% fines) is planned to be carried out at the IEA-R1m research reactor.

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