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Effect of densification additive (Al (OH) $_3$) and U_3O_8 recycle in sintering UO_2–7wt% Gd_2O_3 fuel pellets

[1,2] Santos, L. R., [3]Riella, H. G.

[1]Centro Tecnológico da Marinha em São Paulo – CTMSP
Av. Professor Lineu Prestes, 2268
05508-900 São Paulo, SP
lauro@ctmsp.mar.mil.br

[2]Instituto de Pesquisas Energéticas e Nucleares, IPEN – CNEN/SP
Av. Professor Lineu Prestes, 2242
05508-000 São Paulo, SP
lrsantos@ipen.br

[3]Departamento de Engenharia Química e Alimentos – UFSC
Campus Universitário – Trindade
88040-900 Florianópolis, SC
riella@enq.ufsc.br

ABSTRACT

The nuclear fuels are the consumable parts of nuclear reactors, and this has several consequences. From an economic point of view, it is important to keep the fuel into reactor for long time. In this context the use of burnable poison, as advanced fuel based in gadolinium oxide dispersed in a uranium oxide matrix, is a technological solution adopted worldwide. The function of the burnable poison fuels is to control the neutrons population in the nuclear reactors cores during its start up and the beginning of the fuel burning cycle to extending their use. In consequence of the use of this advanced fuel, the nuclear reactors can operate with higher rate of power, optimizing the use of the nuclear fuels.

The objective of the present work is to show the development of UO_2-7wt% Gd_2O_3 burnable absorver containing pellets by using mechanical blending of ($Al(OH)_3$) densification additive and U_3O_8 of the recycling of nuclear fuel scrap. In the procedures, the gadolinium content of 7 wt% was established as a consequence of the P & D Cooperation Programmer firmed by the CTMSP and the INB, looking for the nationalization of this type of nuclear fuel used in the Nuclear Facility of Angra 2.

The experimental results permit to observe the effectiveness action of the compound $Al(OH)_3$ as a additive to promote the increasing in the densification of the $(U\text{-}Gd)O_2$ pellets during its sintering, when amounts of recycle are recycled to the production processing up to 10 wt%, and when 0,20 wt% of $Al(OH)_3$ is used as additive.

1. INTRODUCTION

In the nuclear fuel cycle stages of the reprocessing and storage of fuel "burned" usually require a high cost in addition to environmental problems. A strategy to reduce these problems is the adoption of measures to reduce the amount of wastes. The use of burnable poison, the basis of gadolinium dioxide, is a measure that contributes to this goal. The role of burnable poison is to control the population of neutrons in the early life of the reactor or the beginning of the cycle of burning of each reloading of fuel. In this way, the burnable poison may promote the recharge increasing time with higher rates of burning, consequently maximizing the fuel efficiency.

Commercially there are only three types of integrated burnable poisons: ZrB_2, Erbium and UO_2-Gd_2O_3. The first is obtained by coating a thin layer (20 μm) of zirconium boride (ZrB_2) on the surface of the UO_2 pellets. The Er_2O_3 is mixed and homogenized to UO_2 in proportions between 1 and 2 % by weight to produce the burnable poison pellets. The burnable poison most widely used is the UO_2-Gd_2O_3. The processes of manufacture of fuel UO_2-Gd_2O_3 can be divided mainly into two different lines: mixed oxides of U and Gd at molecular level (sol-gel, co-precipitation) or mixtures of oxides of U and Gd as fine powders [1]. The development of the process by mechanical blending was used in this work. This process would be implemented by using $Al(OH)_3$ additive that permits to achieve physical characteristics as high density and homogeneous porosity.

This paper deals with the problem of the obtaining of UO_2-7wt%Gd_2O_3 burnable absorver-containing fuel pellets that was added also the ($Al(OH)_3$) densification additive and the U_3O_8 of recycling of fuel scrap. Were studied two types of recycle: U_3O_8 sp which was obtained from calcination of sintered pellets and U_3O_8 gp which was obtained from calcination green pellets. This paper also describes the properties of UO_2-7wt%Gd_2O_3 fuel pellets fabricated by including the mentioned additive and recycling of fuel scrap. Both additive and recycling of fuel scrap have shown promise. In particular, it was found that the ($Al(OH)_3$) utilization is essential to reach the desired density and open porosity.

2. EXPERIMENTAL PROCEDURE AND RESULTS

The process proposed in this work is to obtain (U-Gd)O_2 pellets using UO_2 and Gd_2O_3 powders by mechanical blending, with densification additive (Al $(OH)_3$) and fuel scrap in the U_3O_8 form. The concentration of 7 wt% of Gd_2O_3 is an Angra 2 specification. The concentrations of $Al(OH)_3$ used were 0.10, 0.15, 0.20, 0.25 and 0.30 wt% and the fuel scrap recycle were 3, 7 and 10 wt%. From all the mixtures produced (U-Gd)O_2 sintered pellets utilizing the following conditions:

-homogenization time - 1 h
-matrix diameter - 11.15 mm
-compaction pressure - 4.0 tf/cm^2
-temperature / sintering time - l750 ° C / 4.0 h
-sintering atmosphere - industrial hydrogen (99.95%)
-green pellets or UO_2 powder calcination temperature / time- 500 ° C / 1.0 h
-sintered pellets calcination temperature / time - 500 ° C / 1.0 h

The physical characteristics of the (U-Gd)O_2 pellets used as a burnable poison in PWR, like Angra 2, should have the following specification [2]:

-hydrostatic density: 9.98 to 10.28 g/cm^3
-open porosity: ≤ 3.0 %
-average grain-size: 7 to 35 μm in at least 90 % of the cut surface
-average size of pores: the majority of the pores must have an average diameter ≤ 100 μm in at least 90% of the cut surface.

-resintering behavior: a change in density before and after annealing at temperature ≥ 1700 °C with a residence time ≥ 24 h in a reducing atmosphere, shall not be greater than 0.142 g/cm^3.

hydrostatic density and open porosity

The hydrostatic density is obtained using the Archimedes Principle that says: the measure of the volume of a sample is calculated using the thrust experienced by this sample in a liquid appropriate. The value of density is obtained thorugh the equation:

d.h. = M / (Mu - Mi) x dl

and open porosity is determined by the equation:

p.a. = (Mu - M) / (Mu - Mi) where:

d.h. = hydrostatic density
p.a. = open porosity
M = mass of dry sample
Mu = mass of wet sample
Mi = mass of the sample immersed
dl = density of the liquid in the temperature of completion of test

For analyze the importance of densification additive, (U-Gd)O_2 pellets were produced using only fuel scrap recycle - U_3O_8 sp and U_3O_8 gp, without making use of the $Al(OH)_3$ additive. Also were produced pellets using only $Al(OH)_3$ additive, without fuel scrap recycle Analyzed the following parameters: density hydrostatic, open porosity, pore size average, grain size average and density hydrostatic variation.
The Table 1 shows the results, and their standard deviations, for (U-Gd)O_2 pellets with 7 wt% of Gd_2O_3 in which added 3, 7 and 10 wt% fuel scrap recycle, but not used the $Al(OH)_3$, and Table 2 illustrates the results obtained, with their standard deviations, for (U-Gd) O_2 pellets with 7wt% of Gd_2O_3, which were added only $Al(OH)_3$ at concentrations of 0,10, 0.15, 0.20, 0.25 and 0.30 wt%.

Table 1. - Physical characteristics of UO_2–7wt%Gd_2O_3 sintered pellets using only the recycle fuel scrap, without densification additive - Al (OH)$_3$

U_3O_8 sp (%)	U_3O_8 gp (%)	geometric density (g/cm^3)	hydrostatic density (g/cm^3)	open porosity (%)	pores - average size (µm)	grains - average size (µm)
0,0	0,0	9,83 ± 0,02	9,81 ± 0,02	4,02 ± 0,28	5,02 ± 4,14	4,38 ± 2,95
3,0	--	9,77 ± 0,02	9,71 ± 0,02	3,64 ± 0,04	4,58 ± 3,60	4,68 ± 2,81
7,0	--	9,77 ± 0,01	9,70 ± 0,01	3,74 ± 0,12	4,24 ± 3,49	3,91 ± 2,56
10,0	--	9,75 ± 0,02	9,69 ± 0,00	3,78 ± 0,15	4,21 ± 3,04	4,38 ± 2,91
--	3,0	9,76 ± 0,01	9,68 ± 0,01	3,89 ± 0,04	5,27 ± 4,15	3,99 ± 2,84
--	7,0	9,70 ± 0,02	9,64 ± 0,01	4,17 ± 0,05	4,22 ± 3,48	4,51 ± 2,86
--	10,0	9,66 ± 0,02	9,60 ± 0,01	4,55 ± 0,12	4,54 ± 3,72	4,21 ± 2,75

Where: **sp** = **s**intered **p**ellets and **gp** = **g**reen **p**ellets

Table 2. - Physical characteristics of UO_2–7wt%Gd_2O_3 sintered pellets using only densification additive - $Al(OH)_3$

$Al(OH)_3$ (%)	geometric density (g/cm^3)	hydrostatic density (g/cm^3)	open porosity (%)	pores - average size (µm)	grains - average size (µm)
0,10	10,18 ± 0,01	10,12 ± 0,02	0,20 ± 0,22	4,5 ± 3,7	10,0 ± 8,4
0,15	10,20 ± 0,02	10,13 ± 0,03	0,11 ± 0,06	5,2 ± 4,1	8,6 ± 6,4
0,20	10,26 ± 0,01	10,18 ± 0,03	0,15 ± 0,09	4,9 ± 4,4	10,4 ± 6,8
0,25	10,20 ± 0,00	10,17 ± 0,02	0,20 ± 0,16	5,9 ± 4,3	10,9 ± 8,0
0,30	10,28 ± 0,02	10,17 ± 0,02	0,20 ± 0,09	5,3 ± 4,9	9,9 ± 9,1

By Table 1, in any of the mixtures was obtained the desired value for hydrostatic density and open porosity. This test showed that there is need to use a densification additive, agreement with the literature that says that one way to improve the densification is using additives [4].

The Fig. 1 shows internal structure micrography of a UO_2-7wt%Gd_2O_3 pellet, which was added 10 wt% U_3O_8 sp recycle, and is not used densification additive - Al $(OH)_3$. There are a lot of internal pores, so that does not get the specified density.

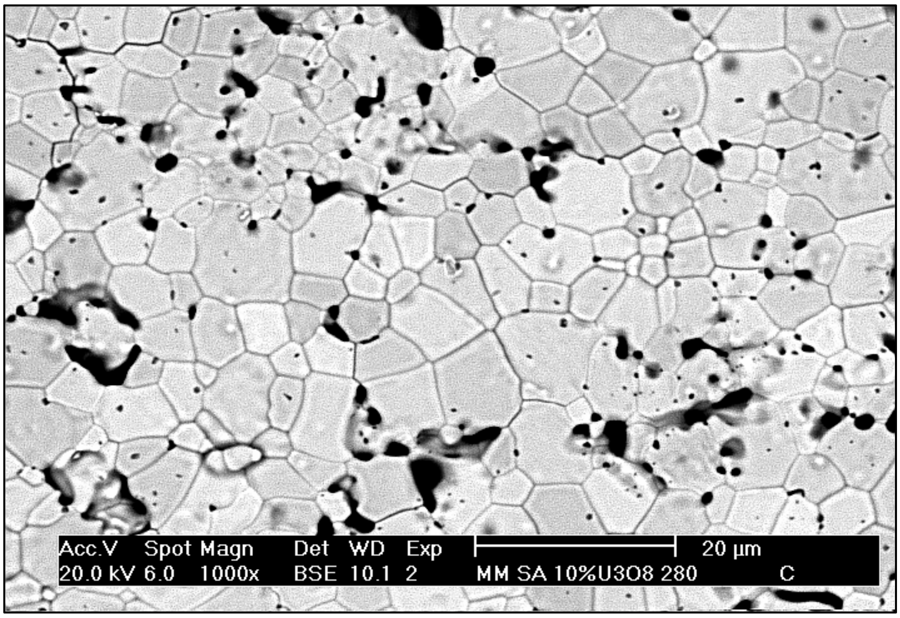

Figure 1. UO_2–7wt%Gd_2O_3 sintered pellet with U_3O_8 sp - 10 wt%, without densification additive

For the analysis of Table 2, all pellets of UO_2-7wt%Gd_2O_3 produced have physical characteristics in accordance with the specification. It is observed that, in comparison with Table 1, that the addition of $Al(OH)_3$ promotes the growth of grain. It is presented in Fig. 2 a pellet with of $Al(OH)_3$ - 0.20 wt% micrography. It is this concentration is obtained greater density average. There is the uniformity of grain size and porosity preferably interstitial. The values quoted in Table 2 refer to the average of 5 pellets for each concentration of Al $(OH)_3$.

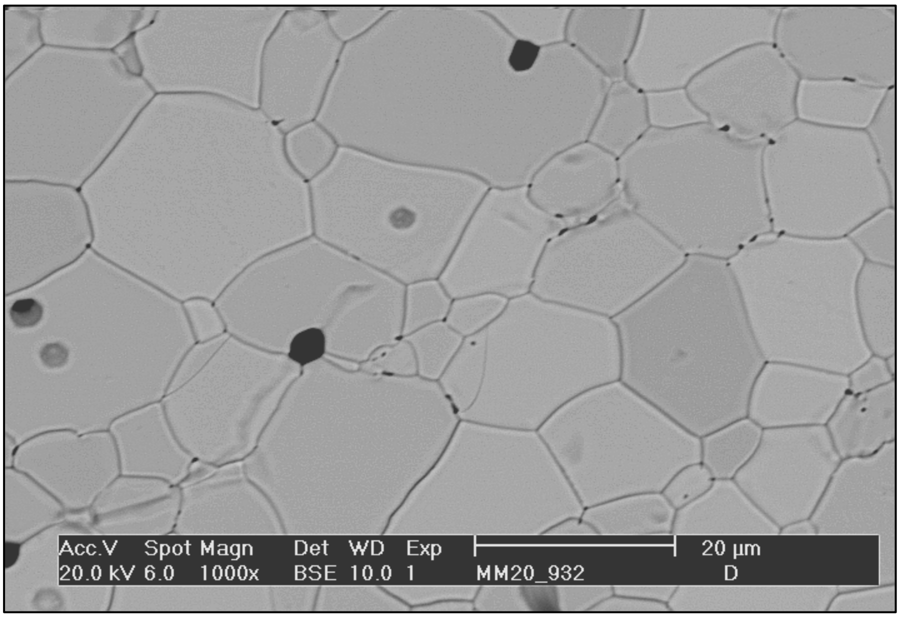

Figure 2. UO_2-7%Gd_2O_3 sintered pellet with $Al(OH)_3$ - 0,20 wt %
(without recycle addition)

The Table 3 illustrates the results obtained, with their standard deviations, of UO_2-7wt%Gd_2O_3 pellets, which were added 3, 7 and 10 wt% of U_3O_8 sp recycle and also used the Al $(OH)_3$, in concentrations of 0.10, 0.15, 0.20, 0.25 and 0.30 wt%.

Based on data from Tables 1, 2 and 3 produced by a graph, Fig. 3, which is indicated the hydrostatic density variation in function of $Al(OH)_3$ concentration and its dependencies of U_3O_8 sp recycle concentration.

Table 3. Physical characteristics of UO_2–7wt%Gd_2O_3 sintered pellets with $Al(OH)_3$ and U_3O_8 sp recycle

composition	geometric density (g/cm^3)	hydrostatic density (g/cm^3)	open porosity (%)	pores - average size (µm)	grains - average size (µm)
MB 10 + 3 % U_3O_8 sp	10,19 ± 0,02	10,08 ± 0,02	0,21 ± 0,11	4,4 ± 3,6	9,3 ± 7,6
MB 10 + 7 % U_3O_8 sp	10,16 ± 0,01	10,07 ± 0,01	0,16 ± 0,06	4,3 ± 3,6	8,9 ± 7,6
MB10 + 10 % U_3O_8 sp	10,14 ± 0,04	10,05 ± 0,02	0,14 ± 0,05	4,6 ± 3,8	8,5 ± 7,7
MB 15 + 3 % U_3O_8 sp	10,15 ± 0,03	10,11 ± 0,02	0,16 ± 0,06	5,1 ± 4,1	7,8 ± 6,1
MB 15 + 7 % U_3O_8 sp	10,13 ± 0,07	10,10 ± 0,05	0,12 ± 0,06	5,2 ± 3,9	8,9 ± 9,3
MB 15 + 10 % U_3O_8 sp	10,11 ± 0,01	10,05 ± 0,06	0,48 ± 0,63	5,1 ± 4,0	8,5 ± 6,0
MB 20 + 3 % U_3O_8 sp	10,19 ± 0,02	10,14 ± 0,01	0,17 ± 0,07	4,9 ± 4,6	9,5 ± 7,0
MB 20 + 7 % U_3O_8 sp	10,18 ± 0,07	10,12 ± 0,05	0,12 ± 0,03	5,1 ± 46	9,3 ± 6,4
MB 20 + 10 % U_3O_8 sp	10,14 ± 0,02	10,09 ± 0,02	0,14 ± 0,07	5,2 ± 4,5	9,6 ± 6,7
MB 25 + 3 % U_3O_8 sp	10,16 ± 0,01	10,11± 0,02	0,19 ± 0,07	6,1 ± 4,6	10,4±8,2
MB 25 + 7 % U_3O_8 sp	10,15 ± 0,02	10,08 ± 0,02	0,33 ± 0,17	5,9 ± 4,4	9,7 ± 7,9
MB 25 + 10 % U_3O_8 sp	10,16 ± 0,03	10,07 ± 0,03	0,12 ± 0,04	5,9 ± 4,4	10,6±7,8
MB 30 + 3 % U_3O_8 sp	10,11 ± 0,03	10,07 ± 0,06	0,13 ± 0,01	4,9 ± 4,5	9,5 ± 7,2
MB 30 + 7 % U_3O_8 sp	10,02 ± 0,01	10,03 ± 0,04	0,21 ± 0,18	5,2 ± 4,6	8,5 ± 6,3
MB 30 + 10 % U_3O_8 sp	10,11 ± 0,02	10,03 ± 0,03	0,35 ± 0,13	6,3 ± 6,4	9,4 ± 5,4

MB XX = **M**echanical **B**lending with **0,XX** wt% of $Al(OH)_3$
sp = **s**intered **p**ellet

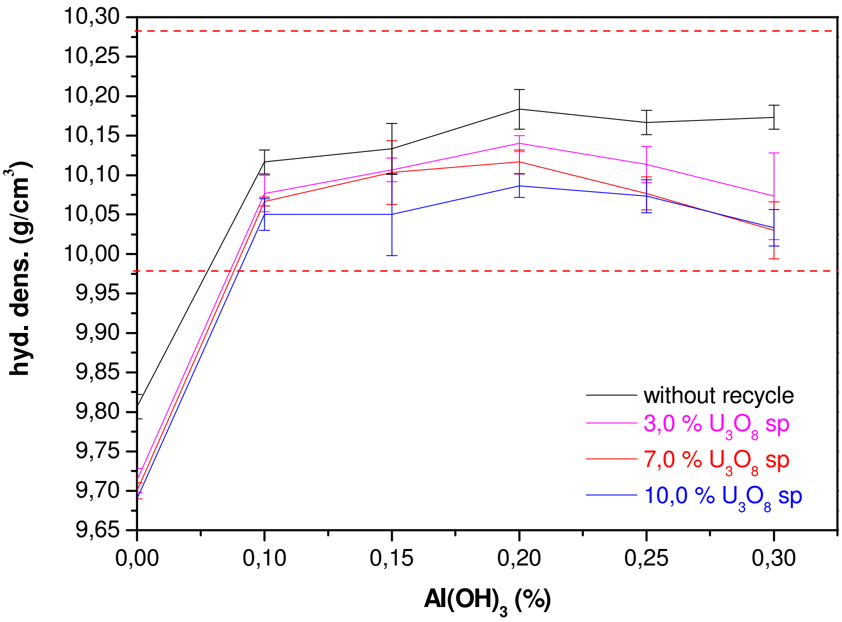

Figure 3. Hydrostatic density (g/cm^3) variation in function of densification additive Al $(OH)_3$ (wt%) and U_3O_8 sp recycle (wt%)

In graph of Fig. 3, for the hydrostatic density average, the addition of U_3O_8 sp recycle, causing a slight decrease in its value for the same $Al(OH)_3$ concentration. When increasing the recycle concentration, decreases the hydrostatic density. It should be noted that this is a trend, because when considering the bar of standard deviations, as the variation is very small, there is overlap of values in most cases. It would also mean that the highest densities are achieved for $Al(OH)_3$ - 0.20 wt%.

In Fig. 4 shows UO_2–7wt%Gd_2O_3 sintered pellet micrography, with $Al(OH)_3$ $^-$ 0.25 wt% and U_3O_8 sp recycle - 7 wt%

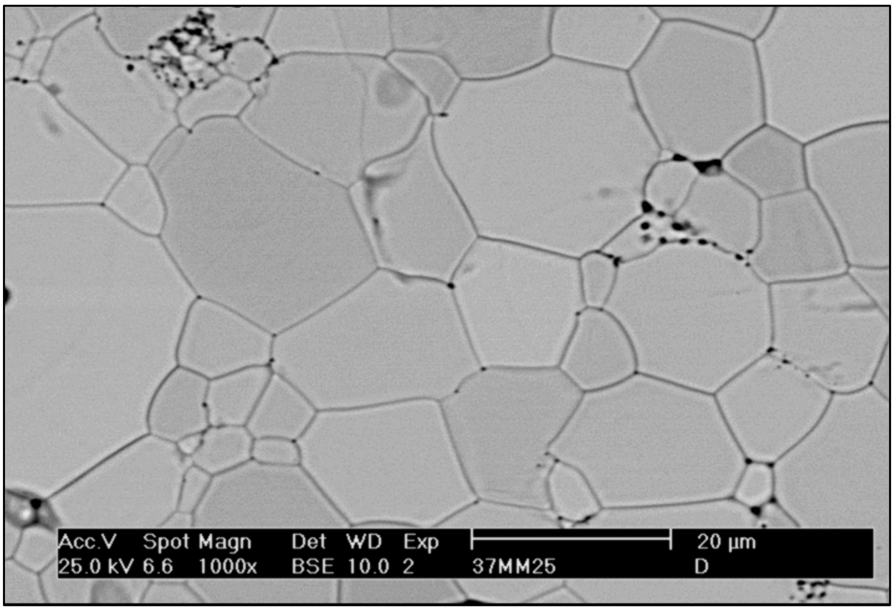

Figure 4. UO_2-7wt% Gd_2O_3 sintered pellet with $Al(OH)_3$ - 0,25 wt% and U_3O_8 sp recycle - 7 wt%

Table 4 illustrates the results obtained, with their standard deviations, of UO_2-7wt%Gd_2O_3 pellets, which were added 3, 7 and 10 wt% of U_3O_8 gp recycle and also used the Al $(OH)_3$, in concentrations of 0.10, 0.15, 0.20, 0.25 and 0.30 wt%.

Table4. Physical characteristics of UO_2–7 wt%Gd_2O_3 sintered pellets with $Al(OH)_3$ and recycle U_3O_8 gp

composition	geometric density (g/cm^3)	hydrostatic density (g/cm^3)	open porosity (%)	pores - average size (μm)	grains - average size (μm)
MB 10 + 3 % U_3O_8 gp	10,12 ± 0,02	10,07 ± 0,01	0,18 ± 0,15	4,6 ± 4,2	9,8 ± 8,2
MB 10 + 7 % U_3O_8 gp	10,07 ± 0,01	10,00 ± 0,03	0,27 ± 0,06	4,7 ± 4,2	9,2 ± 7,3
MB 10 + 10 % U_3O_8 gp	10,01 ± 0,02	***9,94 ± 0,02***	0,69 ± 0,16	4,7 ± 4,0	8,7 ± 6,7
MB 15 + 3 % U_3O_8 gp	10,03 ± 0,02	10,10 ± 0,02	0,33 ± 0,25	5,2 ± 4,2	8,9 ± 5,8
MB 15 + 7 % U_3O_8 gp	10,05 ± 0,05	10,08 ± 0,02	0,20 ± 0,07	5,0 ± 4,0	9,2 ± 5,9
MB 15 + 10 % U_3O_8 gp	10,14 ± 0,02	10,07 ± 0,02	0,16 ± 0,04	4,8 ± 4,0	9,1 ± 6,0
MB 20 + 3 % U_3O_8 gp	10,22 ± 0,03	10,16 ± 0,01	0,15 ± 0,02	5,3 ± 4,8	10,3±7,0
MB 20 + 7 % U_3O_8 gp	10,22 ± 0,02	10,15 ± 0,02	0,24 ± 0,08	5,9 ± 4,7	9,0 ± 6,9
MB 20 + 10 % U_3O_8 gp	10,18 ± 0,03	10,14 ± 0,04	0,20 ± 0,16	5,1 ± 4,1	8,0 ± 6,0
MB 25 + 3 % U_3O_8 gp	10,20 ± 0,04	10,15 ± 0,03	0,15 ± 0,10	6,0 ± 4,5	10,8±8,1
MB 25 + 7 % U_3O_8 gp	10,18 ± 0,02	10,11 ± 0,02	0,32 ± 0,05	5,9 ± 4,7	10,4±8,1
MB 25 + 10 % U_3O_8 gp	10,18 ± 0,03	10,10 ± 0,01	0,18 ± 0,02	6,0 ± 4,5	10,6±7,7
MB 30 + 3 % U_3O_8 gp	10,18 ± 0,04	10,14 ± 0,03	0,17 ± 0,16	4,4 ± 3,6	8,4 ± 6,4
MB 30 + 7 % U_3O_8 gp	10,14 ± 0,01	10,09 ± 0,01	0,24 ± 0,07	6,5 ± 5,7	8,2 ± 7,0
MB 30 + 10 % U_3O_8 gp	9,89 ± 0,07	***9,94 ± 0,12***	0,42 ± 0,21	6,3 ± 4,9	7,6 ± 5,8

MB XX = **M**echanical **B**lending with **0,XX** wt% of $Al(OH)_3$
gp = **g**reen **p**ellet

Whereas in Table 4, it is observed that the condition in sintered pellets of UO_2–7wt%Gd_2O_3 + Al $(OH)_3$ - 0.10 wt% + U_3O_8 gp - 10 wt%, not reached the density specified [2] Based on these pellets micrography, Fig. 5, there is intragranulates pores due to a blockage in the process of densification caused by high levels of U_3O_8 obtained by green pellet calcination. The U_3O_8 gp shows high activity and can change the local oxygen potential, transforming ions of U^{+4} to U^{+5} thereby increasing the diffusivity of uranium. As a result it has the Kirkendall effect, as observed in published studies [5]. It is emphasized that the content of 0.10 wt% of Al $(OH)_3$ was not enough to minimize this effect.

Again in Table 4, it is observed that the condition of UO_2–7wt%Gd_2O_3 + $Al(OH)_3$ - 0.30 wt% + U_3O_8 gp - 10 wt%, the hydrostatic density is below the specified value [2] . In this case we should consider an additional effect to explain the reduction of the hydrostatic density, i.e., the high content of $Al(OH)_3$. An excess of additive does not compromise the growth of grain size, but causes a reduction in density due to volatilization of $Al(OH)_3$ [6].

Based on data from Tables 1, 2 and 4 prepared by a graph, Fig. 6, which is indicated the hydrostatic density variation in function of the $Al(OH)_3$ concentration and U_3O_8 gp recycle concentration. This figure can be seen that hydrostatic density average, increasing with the addition of U_3O_8 gp recycle, is that there is also a small reduction in its value for the same concentration of Al $(OH)_3$. Again, the highest average densities are achieved for $Al(OH)_3$ - 0.20 wt%.

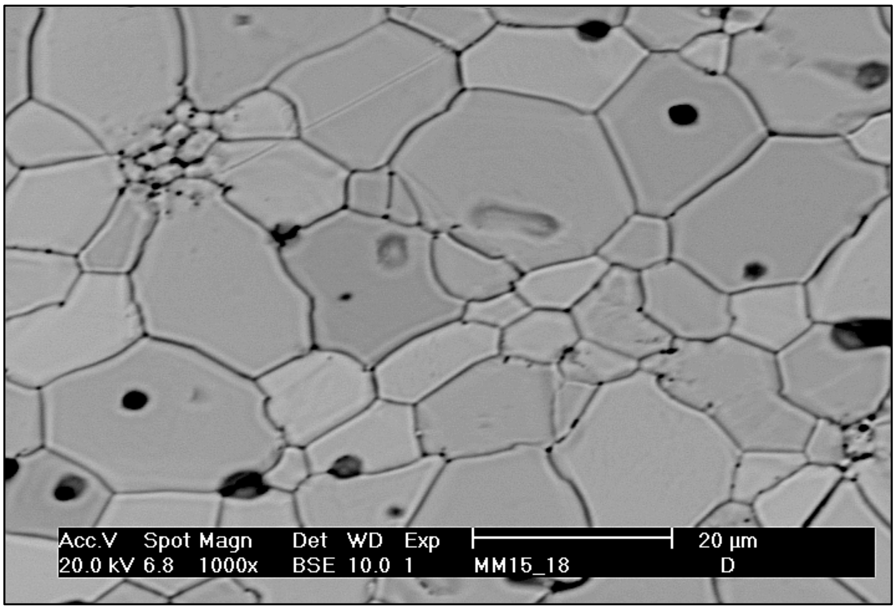

Figura 5. UO_2–7wt% Gd_2O_3 sintered pellet with $Al(OH)_3$ - 0,15 wt % and U_3O_8 gp - 7 wt%

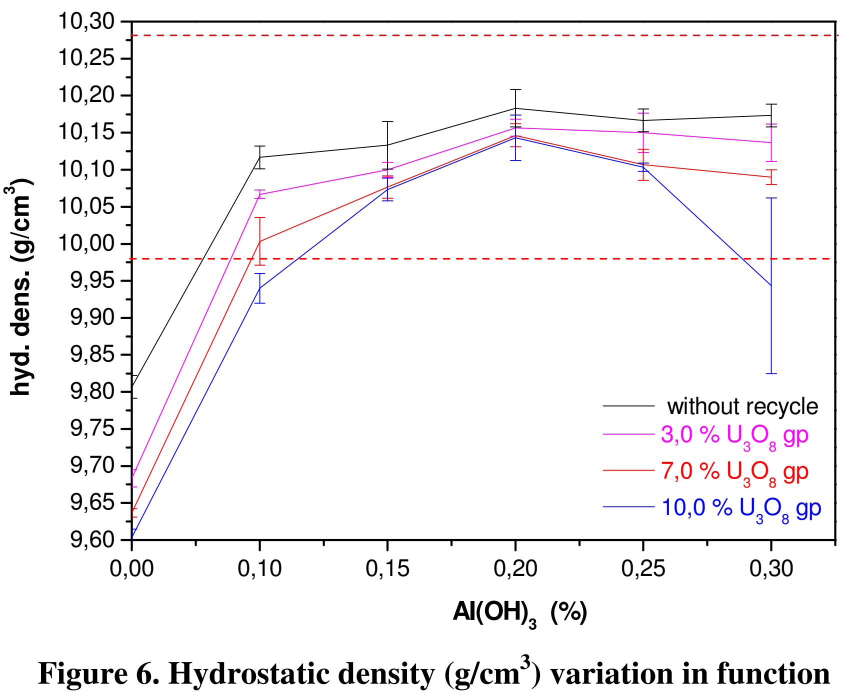

Figure 6. Hydrostatic density (g/cm^3) variation in function densification additive Al $(OH)_3$ (wt%) and U_3O_8 gp recycle (wt%)

Another test with sintered pellets was the resintering test. This test consists of subjecting the pellets at a high temperature for a long time, aiming to simulate its contraction when they are in the reactor core. The analysis of the contraction is done by measuring the hydrostatic density variation. The test was performed at a temperature of 1720 ° C for 24 h in a reducing atmosphere. For the specification, the density increase can not be greater than 0.142 g/cm^3 [2].

Resintering test was conducted using the three recycle concentrations (3, 7 and 10 wt%) for $Al(OH)_3$ - 0.20 wt%. For the other $Al(OH)_3$ concentrations (0.10, 0.15, 0.25 and 0.30 wt%) used only 7 wt% for each recycle. Table 5 show the results obtained with this test.

Table 5. Characteristic of the $(U\text{-}Gd)O_2$ pellets after resintering

resintering	h mm	d mm	M g	g. d. g/cm^3	h. d. g/cm^3	r. h. d. g/cm^3	r. o. p. %	den. var. g/cm^3
MB 10+7 %U_3O_8 sp	10,230	9,290	6,9479	10,17	10,07	10,09	0,1309	0,02
MB 10+7 %U_3O_8 gp	10,350	9,285	6,9599	10,08	9,99	10,00	1,7443	0,01
MB 15+7 % U_3O_8 sp	10,265	9,280	6,9277	10,12	10,11	10,11	0,0585	0,00
MB 15+7 % U_3O_8 gp	10,380	9,280	6,9210	10,00	10,08	10,09	0,1022	0,01
MM 20	10,320	9,230	6,9869	10,27	10,18	10,18	0,1480	0,00
MB 20+3 % U_3O_8 sp	10,270	9,255	6,9205	10,16	10,14	10,16	0,0441	0,02
MB 20+7 % U_3O_8 sp	10,325	9,230	6,9310	10,18	10,12	10,12	0,1064	0,00
MB 20+10 % U_3O_8 sp	10,425	9,210	6,9331	10,13	10,09	10,11	0,0438	0,02
MB 20+3 % U_3O_8 gp	10,315	9,240	6,9846	10,25	10,15	10,16	0,1020	0,01
MB 20+7 % U_3O_8 gp	10,320	9,235	6,9736	10,24	10,13	10,14	0,0583	0,01
MB 20+10 % U_3O_8gp	10,370	9,215	6,9369	10,18	10,18	10,18	0,1024	0,00
MB 25+7 % U_3O_8 sp	10,380	9,235	6,9610	10,16	10,06	10,10	0,0873	0,04
MB 25+7 % U_3O_8 gp	10,380	9,220	6,9475	10,17	10,09	10,13	0,1023	0,04
MB 30+7 % U_3O_8 sp	10,385	9,260	6,8968	10,00	10,04	10,07	0,3803	0,03
MB 30+7 % U_3O_8 gp	10,265	9,265	6,9274	10,16	10,10	10,13	0,0879	0,03

MB XX = **M**echanical **B**lending with **0,XX** wt% of $Al(OH)_3$
sp = **s**intered **p**ellet
gp = **g**reen **p**ellet
h = pellet height
d = pellet diameter
m = pellet mass
g. d. = geometric density
h. d. = hydrostatic density
r. h. d. = hydrostatic density after resintering
r. o. p. = open pores after resintering
den. var. = hydrostatic density variation

3. CONCLUSIONS

Without the addition of the additive for densification $Al(OH)_3$ does not get the desired density and open porosity.

When the recycle are not added, you get $(U\text{-}Gd)O_2$ sintered pellets in accordance with the specifications for the five Al $(OH)_3$ concentrations.

The reuse up to 10 wt% of recycle in the form of U_3O_8, obtained from the calcination of sintered pellets, provides the burnable poison $(U\text{-}Gd)O_2$ pellets, according to the specifications for all concentrations of Al $(OH)_3$ studied.

To recycle the U_3O_8, obtained from the calcination of green pellets, it is observed that when adding 10 wt% of this recycle, in the mixtures with $Al(OH)_3$ - 0.10 and 0.30 wt%, the average density of not meet the specified limits.
For the concentration of $Al(OH)_3$ - 0.20 wt%, is obtained pellets $(U\text{-}Gd)O_2$ with higher average densities.

The use of the additive $Al(OH)_3$ not only promotes the densification of the pellets of $(U\text{-}Gd)O_2$, but also promotes the growth of grain.

4. REFRENCE

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