

URANIUM RECOVERY FROM SLAGS OF METALLIC URANIUM

Fornarolo, F.; Frajndlich, E.U.C.; Durazzo, M.

Centro do Combustível Nuclear - IPEN/CNEN-SP

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The Nuclear Fuel Center of IPEN has already concluded its development program to fabricate uranium silicide fuel type (U₃Si₂-Al) for nuclear research reactors as IEA-R1 starting from 20wt% enriched uranium hexafluoride. The obtaining process of U₃Si₂ involves metallic uranium as an intermediate product which produces slags containing uranium.

The present work shows the results obtained from the development of a process of uranium recovery from slag's lixivium of calcined metallic uranium. Metallic uranium is unstable, pyroforic and extremely reactive. On the other hand, U₃O₈ is a stable oxide with low chemical reactivity, and it justifies the slags calcination of metallic uranium. This calcination occurs in oxidizing atmosphere and transforms the metallic uranium into U₃O₈, as shown by the x-ray diffraction (FIG. 1). Some experiments have been carried out using different nitric molar concentrations, acid excess contents and temperature control of the lixivium process. The nitric lixivium main chemical reaction for calcined metallic uranium slags is represented by the equation:



It was adopted the following process parameters: Calcination of slag of metallic uranium (600 °C /3 h), sieving and granulometric segmentation of the calcined slag (100 - 200 mesh), concentration adjustment of HNO₃ lixivium (1,0 molar), HNO₃ excess (120%) lixivium temperature (40 °C), agitation (300 rpm), turbine stem type (45 degrees inclination). As results there were obtained: lixivium occurrence minimum time: 9 hours; fluoride concentration in lixivium: 0,002g/L%. Lixivium made at low temperatures and low nitric concentrations reduce both the solubility of magnesium and calcium fluoride and the corrosion effect caused by fluoride ions, ensuring a stable and secure lixivium from operational point of view. The nitric dissolution of metallic uranium sludges produces an uranyl nitrate solution, an utilized compound to feed the uranium purification system by solvent extraction method, with diluted n-tributylphosphate, a routinely utilized procedure for fuel plates, compacts and rejected powders recovering on the fabrication process of the fuel element. The purified uranium is precipitated as ammonium diuranate (ADU) at 600C, by injecting gaseous ammonium diluted with air. Aiming at returning of the recovered material to the U₃Si₂-Al fuel element production process, the purified ADU has to be converted to uranium tetrafluoride (UF₄) by U₃O₈ route. The obtained pure ADU was converted into U₃O₈ by calcination. Thus, all the present U₃O₈ reacts with SnCl₂ and HF, so UF₄ is obtained.

Considering the analysis of the x-ray diffraction spectrum of pure U₃O₈ samples and slags of calcined metallic uranium in contrast to impurities samples of calcined metallic uranium slags (FIG.1) it can expressly be verified the separation of these phases. A 99,5% yield testifies the feasibility of the uranium recovery from slags of metallic uranium process by means of prior calcination of these slags in nitric media with both low acid concentration and lixivium temperature.

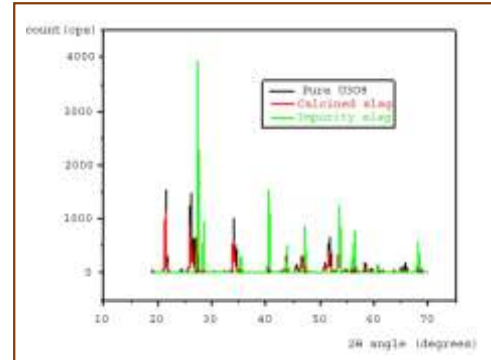


FIGURE 1 - x-ray diffraction spectrum.

ACTIVITIES ON HIGH DENSITY UMO FUELS POWDER FABRICATION

¹Oliveira, F.B.V.de; ²Gracher, R.H.; ¹Durazzo, M.

¹Centro do Combustível Nuclear - IPEN/CNEN-SP; ²Universidade Federal de Santa Catarina - UFSC

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The next step after the UMo alloy obtention is its transformation in the powder form, to its use as a dispersion fuel in the plates. In order to particulate the ingot some methods were studied, and the decision is to use the hydration-dehydration method, called sometimes HDH. This process consists of an initial chemical reaction, in temperatures about 450-500C, between uranium and hydrogen, leading to the formation of uranium hydride UH₃. To avoid uranium oxidation, the chamber where the reaction occurs must be evacuated, then filled with argon prior to the hydrogen insertion. The rate of hydrogen absorption by uranium was monitored with a manometer, which indicates the fall of pressure with time. The system worked semicontinuously, when the hydrogen amount dropped to a certain value, the operator inserted more hydrogen, and so on, until the end of the absorption. When we reach the phase where the pressure doesn't drop itself, it is indicated that we have to proceed to the dehydration (DH) step. DH is carried out without the need to open the chamber, but now the temperature is about a half of the H temperature, and vacuum is applied until the end of the hydrogen release. This indicates that uranium hydride is decomposing itself in hydrogen (gas) and uranium powder, our goal. Here at CCN / IPEN we worked with natural uranium in both phases, alpha and gamma. The former behaves better than the later in terms of the reaction rates, but we showed with our experiments that in both cases the hydration is possible.