CHEMICAL FLOWSHEET FOR RECYCLING IN ADVANCED FUEL CYCLES

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ABSTRACT

The process chemistry for the technical evaluation of TANDEM recycling has been investigated. The purpose of this work is the utilization of electrochemical techniques in order to improve the Purex process. The main advantages are the reduction of the number of cycles, and mostly, the minimization of the generated waste volume. The proposed flowsheet involves four steps of solvent extraction, being that two partitioning ones were performed by an electrolytic mixer-settler MIRELE. The resulting solution achieved a suitable composition for MOX fuel refabrication. An important application of the electrochemical techniques in the new concept of the fuel recycling has been demonstrated.

INTRODUCTION

Nowadays the reutilization of spent nuclear fuels has been largely discussed, mostly to reduce the use of natural uranium and to avoid plutonium proliferation. In these recycling processes uranium and plutonium are recovered toghether, and their utilization as MOX fuel enhances the reactor efficiency. The TANDEM cycle proposed by the canadians researchers has stimulated the countries that have PWR and HWR reactors to develop a number of studies in this field.

With the purpose of conciliating the principles of TANDEM cycle and the electrochemical techniques in the treatment of spent nuclear fuel, a new chemical flowsheet is proposed, where uranium and plutonium are recovered, minimizing both the number of cycles and the waste volume.

The electrochemical techniques was first employed in the U/Pu partitioning at 1st and 2nd cycles of Purex process[1, 2, 3]. Forbicini & Araújo[4] have developed an electrolytic mixer-settler MIRELE (mixer-settler for electrochemical reduction) and have performed several studies on U/Pu partitioning at 2nd cycle.

Based on the flowsheet proposed by Zabunoglu & Spinrad[5] for the treatment of long-cooled light water reactor (LWR) fuel, a flowsheet was proposed using the mixer-settler MIRELE in the two steps involving partial and finish partitioning.

ELECTROCHEMICAL PROCESS

The U/Pu partitioning by electrolyses consists of a liquid-liquid extraction process, using 30% TBP-diluent as an organic phase, loaded with uranium and plutonium, the plutonium being reduced to the practically non-extractable Pu(III). The electrolyte solution (aqueous phase) is composed of HNO_3 and $N_2H_5(NO_3)$ (hydrazine), the latter used to stabilize Pu(III) by the reaction with the NO_2^- formed. In this process, Pu(IV) is reduced both chemically and electrochemically.

The mixer-settler MIRELE

Details of the equipment can be seen in Figure 1. It is composed of eight stages, being each stage coupled to a continuous current source by the platinum anode and titanium cathode. The shape of the settling chamber ensures a large cathode surface, where the reduction occurs. The isolated chamber provides a region free of turbulence and avoids the contact of the anode with the organic phase.

EXPERIMENTAL

A 16-stage mixer-settler, named Celeste-I (Figure 2), was used at the extraction/scrubbing and reextraction steps.

The aqueous feed solution was prepared with about 74gU/L, 75mgPu/L and 3.5M HNO_3 and the organic phase a 30% TBP-diluent solution. The flow rate used for each solution was carefully studied, principally at the extraction step, to avoid Pu accumulation in the mixer-settler.

The current employed in the electrolytic mixer-settler was settled in a previous work[6] in the way to keep a reliable condition for Pu reduction. As electrolyte a 0.2M HNO_3 / 0.2M $N_2H_5NO_3$ solution was used.

Chemical flowsheet

The flowsheet proposed for the recycling of a PWR fuel to be reutilized in a HWR reactor can be seen in Figure 3. Table 1 shows the composition of each solution.

The solutions leaving the mixer-settler MIRELE must be reoxidized to decompose hydrazine and bring Pu(III) back to its +4 oxidation state. An electrolytic equipment named CELOX used. It is a simple flow cell, where the aqueous phase in contact with the electrodes is oxidized. In this case, an higher anode surface compared to cathode surface is used to ensure anodic oxidation.

Results

The evaluation of the results has demonstrated that the utilization of electrochemical techniques for the treatment of spent nuclear fuel in advanced fuel cycles is suitable for the purposes of this work.

The mixer-settler MIRELE, developed previously for the study of U/Pu partitioning, proved its efficiency and flexibility by the use in the recycling process of advanced fuel cycles, where uranium and plutonium were separated together from the other elements, for further fabrication of MOX fuel. The Pu/U ratio has increased 2.2 times and the losses of plutonium, during the whole process, were below 0.1%.

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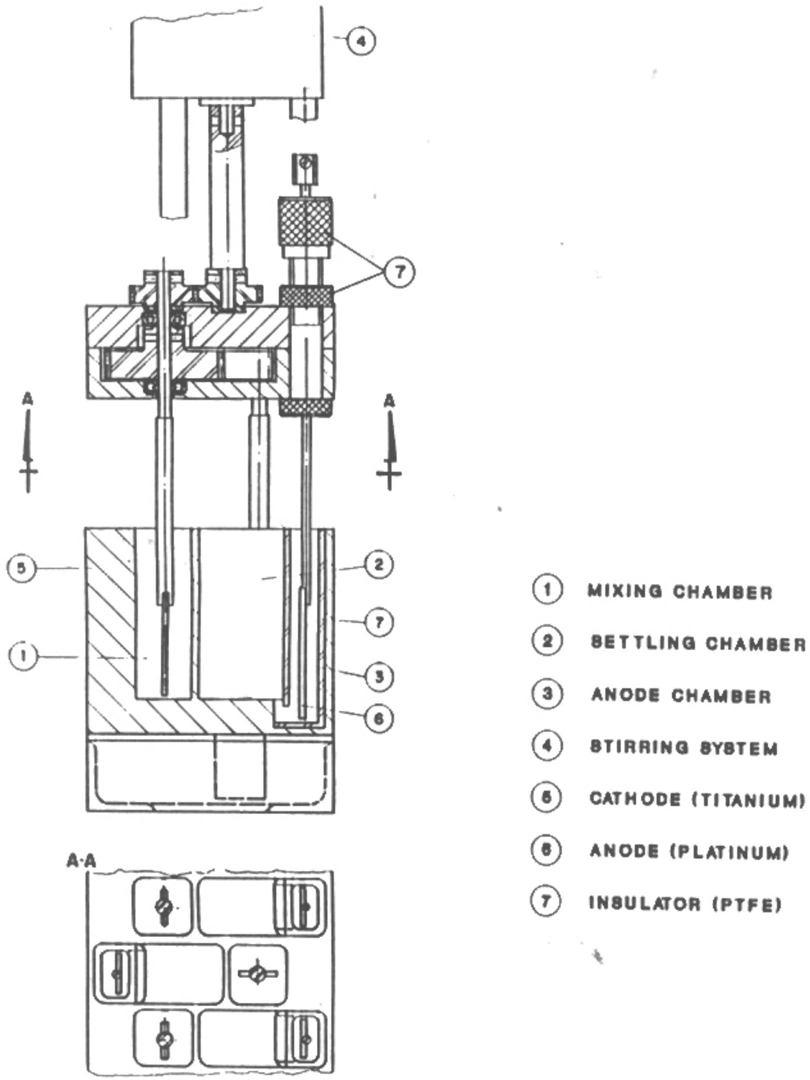

FIGURE 1: ELECTROLYTIC MIXER-SETTLER MIRELE
CAPACITY: 440ML

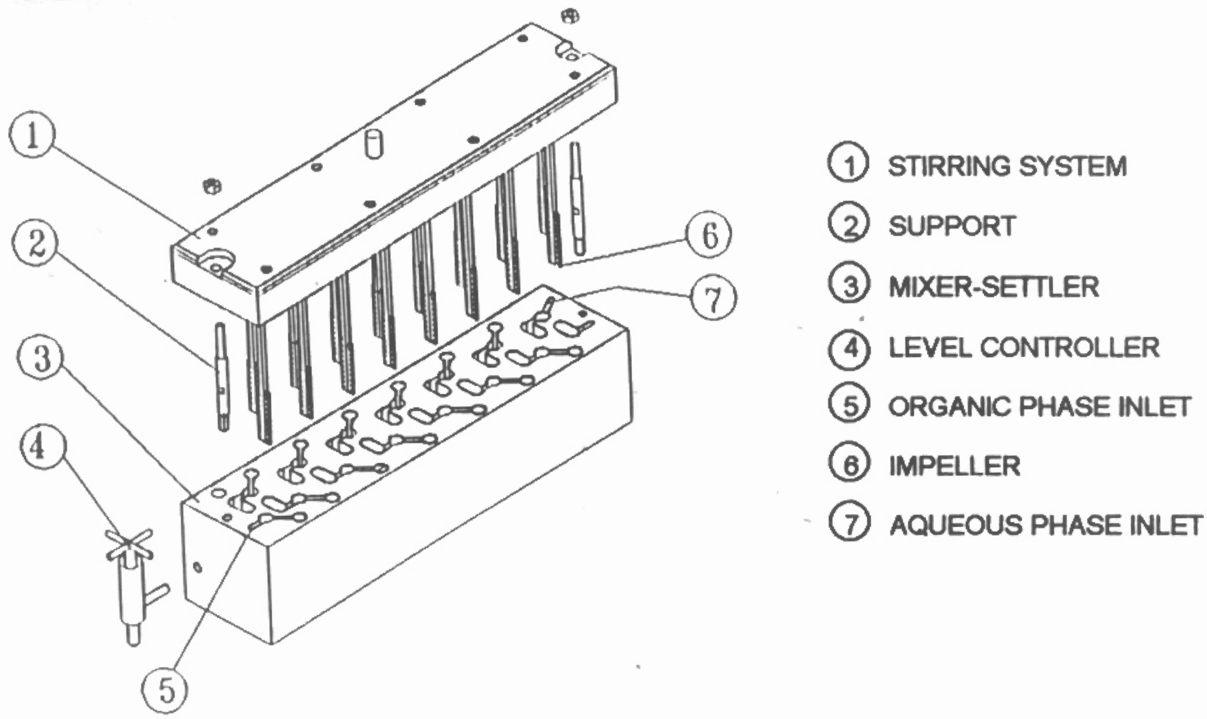

FIGURE 2: MIXER-SETTLER CELESTE-I
CAPACITY: 450ML

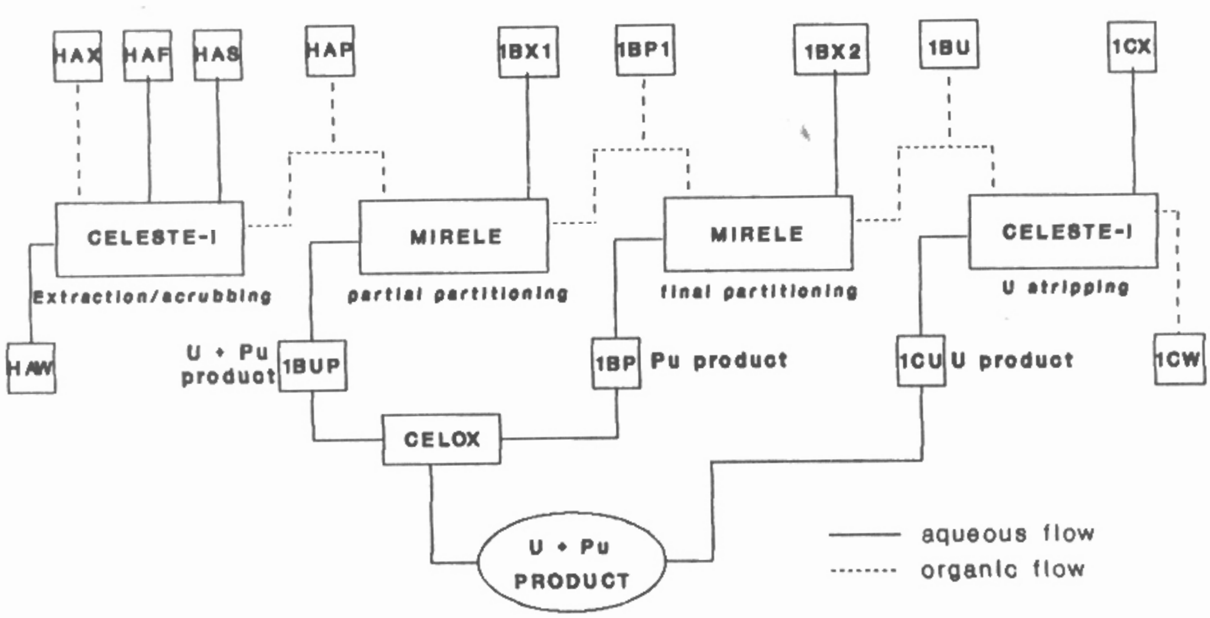

FIGURE 3: PROPOSED FLOWSHEET FOR THE PWR FUEL RECYCLING AS HWR FUEL, EMPLOYING ELECTROCHEMICAL PARTITIONING.

TABLE 1: SOLUTIONS COMPOSITION IN THE FLOWSHEET FOR NUCLEAR FUEL TREATMENT

SOLUTIONS	*TBP-diluent (%)*	*[U] (g/L)*	*[Pu] (mg/L)*	*HNO_3 (M)*	*N_2H_4 (M)*	*FLOW RATE (mL/h)*
HAF: feed solution		73.25	75.10	3.57		150
HAX: organic extracting solution	30					250
HAS: scrubbing solution				0.50		50
HAW: aqueous waste		<0,01	0.01	3.04		200
HAP: co-decontamination extract	30	43.38	44.35	0.19		250
1BX1: partial partitioning reextracting solution				0.20	0.20	200
1BUP: U-Pu product		19.37	43.59	0.36	0.13	200
1BP1: partial partitioning extract	30	24.20	0.01	0.02		200
1BX2: finish partitioning reextracting solution				0.20	0.20	200
1BP: Pu product		6.01	0.009	0.18	0.12	200
1BU: finish partitioning extract	30	18.02	0.001	0.04		200
1CX: U stripping solution				0.01		200
1CU: U product		17.83	0.001	0.02		200
1CW: solvent to be recored	30	<0.01	0.0002	0.01		200