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Recovery of Actinides from Actinide-Aluminium Alloys: Chlorination Route

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A method for recovery of actinides (An) from An-Al alloys formed by electrochemical separation of metallic spent nuclear fuel on solid aluminium electrodes in molten chloride salts is described. The proposed route consists of three main steps: Vacuum distillation of salt adhered on the electrodes, chlorination of An-Al alloy by pure chlorine gas and sublimation of formed AlCl_3 . A thermochemical study of the route was performed to determine important chemical reactions and to find optimum experimental conditions for all process steps. Vacuum distillation of the electrode is efficient for complete removal of remaining salt and most fission products (FP), full chlorination of the An-Al alloys is possible at any working temperature and evaporation of AlCl_3 is achieved by heating under argon. Experiments have been carried out using U-Al alloy in order to define parameters providing full alloy chlorination without formation of volatile UCl_5 and UCl_6 . It was shown that full chlorination of An-Al alloys without An losses should be possible at a temperature approx. 150°C .

INTRODUCTION

A promising pyrochemical reprocessing option regarding spent nuclear fuels is the grouped-selective separation of all actinides (An's) by electrochemical methods in a LiCl-KCl molten salt. An's are electrochemically grouped-selectively reduced from the mixture of fission products (FP) dissolved in the molten salt. In ITU a process based on electrorefining of metallic alloy fuel and the use of solid reactive aluminium cathodes is under development. In recent work, the efficiency of the process and selectivity over lanthanides has been demonstrated [1, 2]. It has been shown that the use of solid aluminium cathodes leads to the formation of stable An-Al alloys [3], highly loaded in An (up to about 70 wt. %) [4].

Efficient separation of An's from Al-An is a key point for the feasibility of the process. The method to recover An's proposed and studied in this work is a chlorination route based on a three step procedure as presented in Fig.1. Initially, the salt, which is composed mainly of LiCl-KCl and some FP chlorides, adhered on the electrode after the electrorefining process is removed by vacuum distillation. The second step is a chlorination of the An-Al alloy in order to fully convert the alloy to chlorides. The last step consists of the sublimation of AlCl_3 . The final An chlorides can be further chemically processed

to obtain the desired output product, e.g. An alloy. A method involving the chlorination of Al-U spent fuel has been proposed by Bohe et al. [5, 6], but up to now is only based on thermochemical calculations without validation by experimental studies involving Uranium.

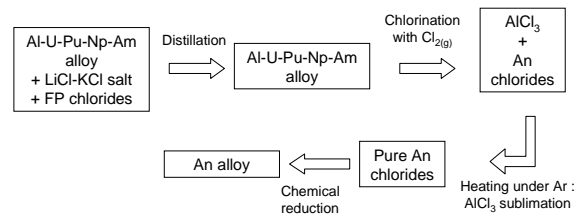


Fig. 1: Principle of the chlorination route (An=actinides, FP=fission products)

In this work, a process using the chlorination of An-Al alloys by pure $\text{Cl}_2(\text{g})$ is investigated. Initially, a detailed thermochemical study of all process steps was carried out. Based on this study, experiments were focussed to optimise process conditions for a full recovery of uranium from U-Al alloys. The reason is that due to the high volatility of UCl_5 and UCl_6 , U is the most difficult element to convert to chloride form without any losses. For that purpose, chlorination experiments on a UAl_3 alloy were carried out in the temperature range $150\text{-}170^\circ\text{C}$.

THERMOCHEMICAL STUDY

The main idea of the chlorination route is to fully chlorinate the alloy and removal of AlCl_3 by sublimation. Thermochemical calculations using the Factsage software were performed, aiming to determine the most suitable conditions for the process. At first, a bibliographic study was carried out to collect available thermochemical data ($\Delta_f H^\circ$, S_m° and $C_{p(T)}$) and phase diagrams. Most thermochemical data concerning the An's chlorides are tabulated, apart from AmCl_n and CmCl_n gaseous compounds [7-11]. Among An-Al alloys, the Gibbs energy of formation of UAl_3 , UAl_4 and PuAl_4 are available, together with phase diagrams for U-Al and Pu-Al systems [12]. In both systems, the melting point of the intermetallic compounds increases with the An content, UAl_2 and PuAl_2 having the highest melting temperature (1620°C and 1540°C, respectively). No phase diagram was found for Np-Al and Am-Al systems. However, three Np-Al intermetallic compounds are reported in the Inorganic Crystal Structure Database [13]. No ternary or quaternary An-Al system was found.

First step: Distillation of remaining salt

To ensure a sufficient purity of both end products (An and Al), it is necessary to clean the cathode from the residual frozen salt adhering at the surface after the electrorefining step. This salt contains a mixture of LiCl-KCl with dissolved FP chlorides. The proposed technique is distillation of the salt from the An-Al alloys and recycling of the salt back in the electrorefiner. This requires sufficient difference in vapour pressures between the salt and the alloy. No thermochemical data exist to fully evaluate the vapour pressure of the salt mixture (LiCl-KCl + FP) and of the alloy (Al-U-Pu-Am-Np-Cm). However, as a first estimation, the vapour pressure, P_M , of each individual compound (FP chlorides and An metals) can be calculated from available data according to:

$$\log P_M = -\frac{\Delta_f G^\circ(T)}{2.303RT} \quad (1)$$

where $\Delta_f G^\circ$ is the Gibbs energy of the equilibrium $M(s, \text{liq})=M(g)$.

Results for the most important An's and FP are plotted in Fig. 2. It can be seen that, except Am, all An metals exhibit a lower vapour pressure than all chloride compounds. As AnAl_3 - AnAl_2 intermetallic compounds have a much higher

melting point than pure metals, their vapour pressures are most likely lower than the one of each individual metal. Therefore, the vapour pressure of alloyed Am will be lower than pure Am.

Pure LiCl-KCl salt is possible to remove by vacuum distillation at temperatures around 800°C, which was confirmed experimentally in the laboratory scale (2g of LiCl-KCl were completely distilled at 900°C and 5.10^{-5} bar). However, other compounds, especially Ln chlorides, need stronger conditions to be removed. Available thermodynamical data indicate that a temperature of 1000°C is required to fully remove all lanthanide (Ln) chloride compounds, with a vacuum of 10^{-5} bar, see horizontal dotted line in Fig 2. However, distillation under these conditions is possibly still not sufficient for complete removal of some FP chlorides (e.g. MoCl_2 , SrCl_2 and BaCl_2).

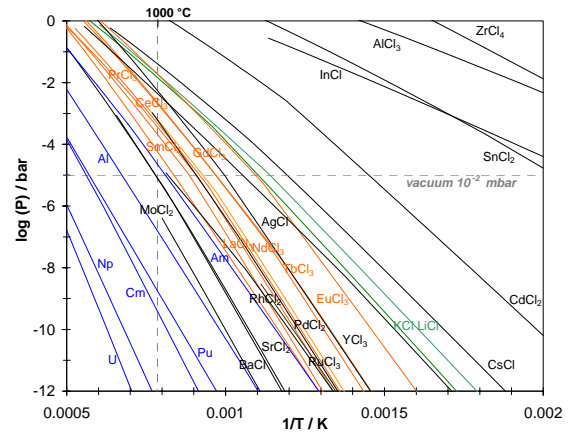


Fig. 2: Vapour pressure vs. reverse of temperature for various metals and chlorides.

Second step: Chlorination of An-Al alloy

The main target of the chlorination is to separate actinides from aluminium. All metal elements (U, Pu, Np, Am, Cm, Al) of the alloy have to be fully converted to chlorides and significantly more volatile aluminium chloride is consecutively evaporated in the following step. Each of presented metals forms a chloride compound in equilibrium with pure chlorine gas, even at relatively low temperatures (25°C). Therefore, the key point of the process is to control working conditions in order to provide formation of desired species only. Formation of gaseous actinide chlorides (e.g. UCl_5 and UCl_6) has to be suppressed to avoid actinide losses by volatilization

The thermochemical study of the chlorination of An-Al alloys is a complex problem, as many compounds can form, and the reactions are strongly dependent on the applied conditions. As a first approach, two main parameters of the process have been studied:

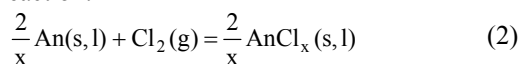
- Temperature of the reaction,
- Molar ratio Cl_2/alloy .

For calculations, a constant volume of 1 dm^3 was used and the Cl_2/alloy molar ratio was fixed by the considered amount of actinide alloy. The following calculations have been performed:

- Relative stability of An chlorides,
- Chlorination of UAl_4 and PuAl_4 at 150°C ,
- Chlorination of UAl_3 in the 100°C - 1000°C temperature range for a fixed Cl_2/UAl_3 ratio.

Chlorination of pure An metals

The relative stability of An chlorides was calculated according to the Gibbs energy of the reaction:



In the 100 - 1000°C temperature range, all Gibbs energy values related to Eq. (2) are strongly negative (between -300 and -600 kJ/mol), indicating that the chloride form is thermodynamically promoted. The products of chlorination can be sorted according to their relative stability to the following sequence (ascending Gibbs energy): $\text{AmCl}_2 > \text{AmCl}_3 > \text{PuCl}_3 > \text{CmCl}_3 > \text{NpCl}_3 > \text{UCl}_3 > \text{UCl}_4 > \text{NpCl}_4$.

Provided that the amount of chlorine gas is sufficient (i.e. at least a stoichiometric amount), the chlorination of considered metals will lead to the formation of the following solid chloride compounds at 150°C : PuCl_3 , AmCl_3 , UCl_4 , NpCl_4 and AlCl_3 . U forms gaseous species UCl_5 and UCl_6 in presence of an excess of chlorine gas as described later. At higher temperatures (up to 1000°C), the order of chlorination remains the same, but liquid and then gaseous compounds will form.

Chlorination of UAl_4 and PuAl_4 at 150°C

As mentioned above, the only data available for An-Al alloys are related to UAl_3 , UAl_4 and PuAl_4 . These data were used to evaluate the behaviour of UAl_4 and PuAl_4 under pure chlorine atmosphere at 150°C (Fig. 3).

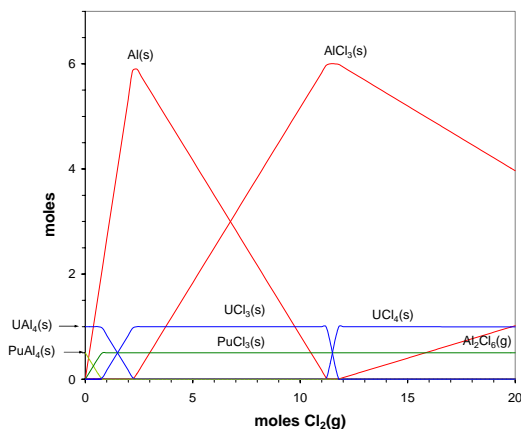


Fig. 3: Chlorination of UAl_4 (1.0 mol) and PuAl_4 (0.5 mol) at 150°C - Influence of the amount of $\text{Cl}_2(g)$.

These calculations show that Pu and U are chlorinated in preference to Al, leaving non-chlorinated Al metal. The behaviour of the alloys is thermodynamically very close to the behaviour of a mixture of pure compounds. However, this calculation shows only the equilibrium state towards which the system should tend. The considered heterogeneous reaction will be most probably limited by kinetic factors due to a surface passivation. Analogous to an oxide layer formation during some oxidation processes, a solid chloride layer is expected to be formed at the surface of the alloy, preventing from further chlorination. This problem can be solved if the electrode is crushed in order to obtain a powder and increase the surface of reaction.

Chlorination of UAl_3 in the 100°C - 1000°C temperature range for fixed Cl_2/UAl_3 ratios

It can be seen from previous calculations that, at 150°C , the chlorination of U yields solid $\text{UCl}_4(s)$, if the Cl_2/U ratio is close to the stoichiometric amount (i.e. 6.5 mol, corresponding to the reaction $\text{UAl}_3 + 6.5 \text{Cl}_2 = 3 \text{AlCl}_3 + \text{UCl}_4$). This temperature is relatively low, and much more favourable reaction kinetics could be obtained, if higher temperatures are used. However, as shown in Fig. 4, an increase of temperature will lead to the formation of gaseous species: $\text{UCl}_6(g)$, $\text{UCl}_5(g)$ and finally $\text{UCl}_4(g)$ will predominantly form with increase of temperature from 150°C to 1000°C .

The formation of compounds starting from UAl_3 alloy and $\text{Cl}_2(g)$ is plotted versus temperature in Fig. 4, using three different Cl_2/UAl_3 initial ratio. It shows that the formation of gaseous uranium

chloride compounds increases with temperature and also with the Cl_2/UAl_3 ratio.

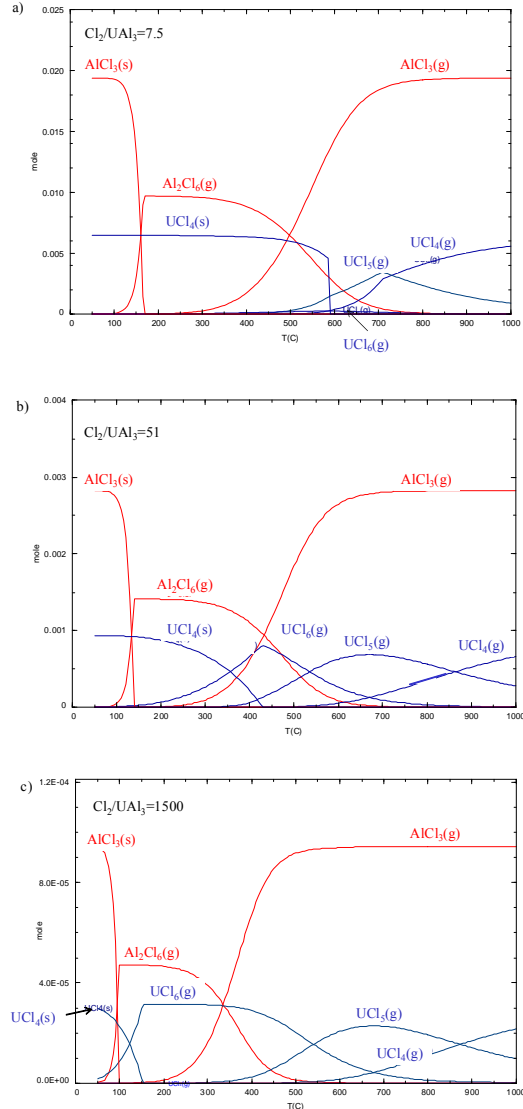


Fig. 4: Chlorination of UAl_3 alloys - Calculation of the evolution of the amount of chloride compounds with temperature and Cl_2/UAl_3 molar ratio. Initial conditions: 1 dm³ reactor containing 2000 - 300 - 10 mg of solid UAl_3 alloy and $4.843 \cdot 10^{-2}$ mol of $\text{Cl}_2(\text{g})$ ($P=1.2$ bars at 25°C).

These calculations indicate that, in order to prevent U losses in the gaseous phase, the reaction should be carried out with the smallest possible excess of Cl_2 . However, from the point of view of the reaction kinetics, the opposite is required, especially because of the reaction type (solid-gas). An experimental study is needed to determine the kinetics of the reaction and to optimise the Cl_2/alloy molar ratio.

Third step: Sublimation of AlCl_3

A well managed chlorination step will lead to a mixture of solid chloride compounds composed of AlCl_3 , UCl_4 , PuCl_3 , NpCl_4 , AmCl_3 and CmCl_3 . The following step is to remove AlCl_3 by sublimation, keeping all An as solids in the reactor. A temperature increase under chlorine atmosphere causes volatilization of U together with Al, but the situation is different using inert Ar atmosphere. As shown in Fig. 5a, the sublimation of AlCl_3 starts at 180°C , while no other chloride compound volatilize before 450°C . At this point, NpCl_4 melts, and quickly volatilizes due to its high vapour pressure. The same behaviour was calculated for UCl_4 , but at a higher temperature ($\sim 550^\circ\text{C}$). As detailed in Fig. 5b, a temperature range of more than 200°C allows to remove AlCl_3 and thus to perform an effective separation of AlCl_3 from the An chlorides.

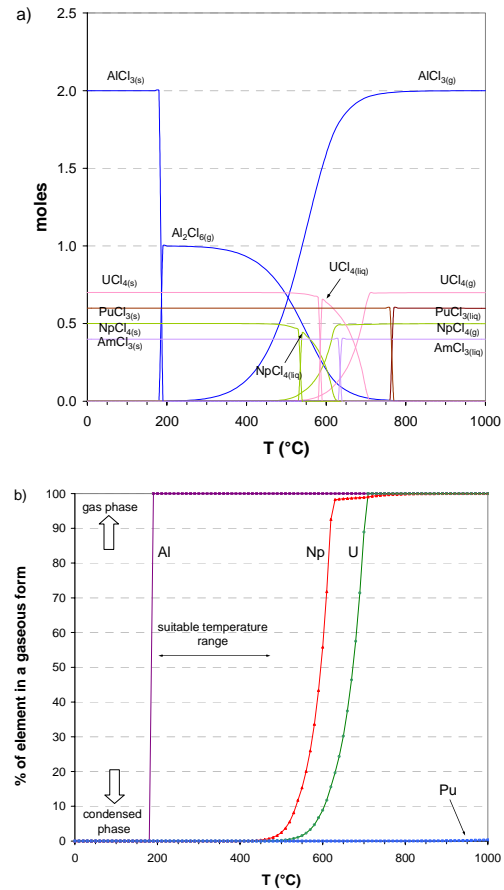


Fig. 5: Sublimation of AlCl_3 from the chloride mixture. a) Evolution of the composition with T. b) Evolution of the volatilization with T. Initial conditions: mixture of 1 mole of An chlorides, 2 moles of AlCl_3 and Ar (1 atm).

CHLORINATION EXPERIMENTS, RESULTS AND DISCUSSION

Although, from a thermodynamical point of view, all steps were proven possible, the applicability of the proposed method depends on the reactions kinetics under working conditions suitable for complete An recovery. The experimental study was carried out to determine realistic conditions for the process and to quantify the chlorination efficiency and actinides volatilization under these conditions.

The experiments were mainly focused on the chlorination step. UAl_3 alloy was selected as input material, since U represents the most difficult element to be completely chlorinated without formation of volatile chlorides. Accordingly, it is expected that the conditions suitable for recovery of U from U-Al alloy should be appropriate for a process including all An.

Experimental

The chlorination experiments were carried out in a glove box under nitrogen atmosphere, specially designed for work with $Cl_2(g)$. The box is equipped with a chlorine gas line ended by a system of gas wash bottles containing KOH solutions (4 and 8 M), where non reacted chlorine gas is absorbed. A quartz reactor placed in a vertical oven was used for the chlorination (see Fig. 6). Chlorine gas was introduced by a quartz tube guided through the lid of the reactor by a gastight connection. The lid was also equipped by sealed ports for a thermocouple and the gas outlet connected to the chlorine off gas treatment. Boron nitride crucibles were used as containers for the chlorinated material and positioned on the bottom of the reactor.

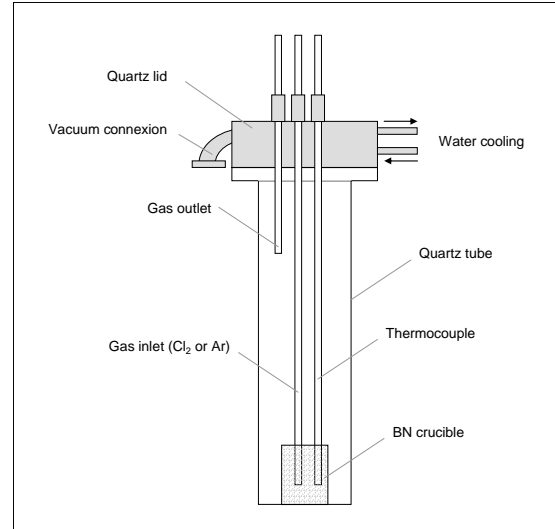


Fig. 6: Experimental set-up for chlorination – Quartz reactor

The input material for chlorination experiments was selected to be UAl_3 alloy. Approx. 3.3g of U-Al alloy was prepared by melting in an arc furnace ($>3000^\circ C$) in 1:3 molar ratio. The composition determined by X-Ray Diffraction (XRD) was 98 wt.% of UAl_3 , 1 wt.% of UAl_2 and 1 wt.% of Al (see Fig. 7). In order to increase the reacting surface, the alloy was crushed into a very fine powder. This was easily achieved manually using a mortar, as U-Al alloys are very brittle.

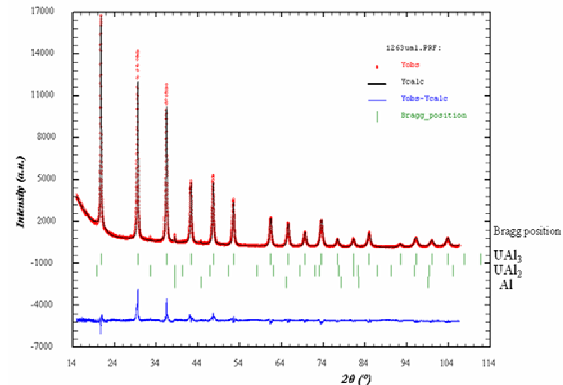


Fig. 7: X-Ray diffractogram of the U-Al alloy prepared with arc furnace and used as input material.

The molar ratio Cl_2 /alloy was kept constant for all the chlorination experiments. The volume of $Cl_2(g)$ was fixed by the volume of the reactor ($1dm^3$) and the amount of powder to chlorinate was set to be 300 mg. The Cl_2 /alloy molar ration was then equal to 36 for all the experiments.

Three experiments were carried out in the temperature range 150 – 170°C. Two experiments, at 150 and 160°C, consisted of two successive chlorinations during approx. 20 hours each. The last experiment at 170°C was a single 20 hours chlorination. In each chlorination experiment, the reactor was filled completely with chlorine gas and then isolated. After chlorination, samples of the chlorinated powder were taken for XRD analysis. In the two first experiments, the rest of the powder was refilled in crucible for further chlorination. Both the initial material and products from the complete chlorination were weighted to calculate a mass balance.

Observations

Experimental results are summarised in Tab. 1. After the first chlorination, a grey/green powder was obtained for all experiments performed. It indicates the presence of UCl_3 and/or UCl_4 . An increase of powder mass was also observed. In two samples, i.e. CIUAL-150 and CIUAL-160, a part of the powder turned into agglomerate that remained stuck at the bottom of the crucible and could not be analysed. The formation of this agglomerate could be induced by the fine powder structure of the initial UAl material. It shows that the efficiency and the optimal conditions for the chlorination route are strongly dependent of the grains size of UAl alloy.

After the second chlorination, a pure green powder was obtained for all experiments performed, indicating the presence of UCl_3 and/or UCl_4 as the major phases (see Fig.8). The agglomerate part, when formed, was still observable but it became brittle which allowed its extraction and analysis.



Fig. 8: Pictures of the powder before chlorination and after second chlorination at 150°C.

TABLE 1 Composition of the input material and the solid chlorination product

	Specie	Concentration [wt.%]	
		Input material	Product
CLUAL150 Temp. 150°C Two chlorinations 20h	UCl_3	0.0	18
	UCl_4	0.0	72.9
	UAl_3	98.0	2.2
	UAl_2	1.0	0.5
	$AlCl_3$	0.0	6.4
	Al	1.0	0.0
	mass [mg]	300.5	371.1
CLUAL160 Temp. 160°C Two chlorinations 20h	UCl_3	0.0	19.0
	UCl_4	0.0	64.0
	UAl_3	98.0	4.7
	UAl_2	1.0	0.0
	$AlCl_3$	0.0	12.2
	Al	1.0	0.0
	mass [mg]	300.2	396.4
CLUAL170 Temp. 170°C one chlorination 20h	UCl_3	0.0	84.5
	UCl_4	0.0	0.0
	UCl_5	0.0	traces
	UAl_3	98.0	0.6
	UAl_2	1.0	0.0
	$AlCl_3$	0.0	14.9
	Al	1.0	0.0
	mass [mg]	301.04	375.5

Composition of the chlorinated product

XRD analysis of the powder, after the first chlorination, indicated a high efficiency of the chlorination, as less than 2% of the initial alloy remained non-chlorinated. After the second chlorination, the same kind of analysis (see Fig. 9) showed the conversion of original product UCl_3 to UCl_4 .

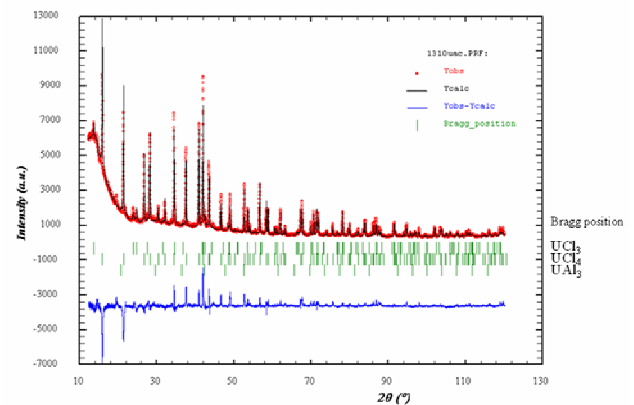


Fig. 9: X-Ray diffractogram of a sample after second chlorination at 150°C.

As expected, the analysis of the agglomerate revealed a lower efficiency of the chlorination, as roughly 15% of the UAl_3 remained in this part even after two chlorinations. This lower efficiency is caused by a limited diffusion of Cl_2 inside the agglomerate, which indicates a drawback in decreasing too much the grain size of the initial powder.

During the experiments, a part of the sublimated $AlCl_3$, and eventually other volatile products, was deposited at the cold surface on the top of the reactor. This deposit was collected and analysed for presence of U compounds. No U was detected in the deposit after the chlorinations carried out at 150 and 160°C. Traces of UCl_5 were detected after chlorination at 170°C but quantification was not possible. However, it clearly shows that higher temperatures than 170°C can not be applied due to uranium losses by volatilisation.

It was impossible to make a quantitative evaluation of the $AlCl_3$ content by XRD due to its high solubility in the epoxy-resin used for the preparation of the sample. Accordingly, the concentration of $AlCl_3$ had to be calculated from the real mass of the chlorination product weighted after the experiment, which was deduced from the calculated theoretical mass of U chlorides and non-reacted residual alloy. The mass balance and an evaluated composition of the chlorination product from the three different experiments are shown in Tab. 1 (for the two first experiments, final mass was evaluated taking into account the mass of powder lost after the first chlorination for analysis). For each experiment, the final composition indicates that chlorination is almost fully efficient as less than 5 wt.% of the final product is composed of metallic U-Al, even at 150°C. The results indicate that a major part of $AlCl_3$ was evaporated already during the chlorination process. According to the calculations, a mass increase up to 686 – 719 mg was expected after chlorination of the initial 300 mg of the alloy, if all $AlCl_3$ had remained in the product. The high percentage of $AlCl_3$ remaining in CLUAL170 is due to the shorter chlorination time, i.e. 20h (instead of 40h for the two other experiments). It also explains why only UCl_3 was formed during this experiment.

CONCLUSIONS

Pyrochemical processes for recovery of actinides from spent nuclear fuel using both solid and

liquid aluminium are investigated at ITU, EC-JRC and CEA, France [14]. Final products for the studied techniques, i.e. electrorefining in molten LiCl-KCl eutectic and reductive extraction to liquid Al in molten LiF-AlF₃, are actinide-aluminium alloys. As very high selectivity and efficiency has been demonstrated in laboratory scale for both methods, the key point for their further application is to identify a method for recovery of actinides from the formed alloys. According to the results described in this work, a chlorination process seems to be advantageous to fulfil this task.

Initially, the theoretical study showed thermodynamical possibility of all steps proposed for the process.

The conclusions from the study were:

- Vacuum distillation is efficient to remove the remaining salt including most fission products, except $MoCl_2$, $SrCl_2$ and $BaCl_2$ (proposed conditions: $T=1000^\circ C$, $P=10^{-5}$ bar).

- Full chlorination of the alloy is possible at any considered temperature (studied temperature range: 25 – 1000°C). In this case, as it is a solid-gas reaction, the reaction kinetic is the most important factor.

- Best conditions for the chlorination step are to chlorinate the finely crushed alloy at 150°C, using minimum acceptable Cl_2 / alloy mass ratio in order to avoid the formation of gaseous U compounds.

- Heating of the chlorination products under inert argon atmosphere at 350-400°C allows to evaporate $AlCl_3$ and consequently to recover pure solid An chlorides.

The experimental study was focused mainly on the kinetic aspects of the chlorination step. Three experiments showed that chlorination of UAl_3 can be efficiently performed in the 150-170°C temperature range. Approx. 20 hours of chlorination yielded products mainly composed of solid UCl_3 and UCl_4 even at a temperature of 150°C, which is in good agreement with the conclusions of the thermodynamic study. Further chlorination led to the conversion of UCl_3 into UCl_4 . Traces of volatile UCl_5 were identified in the condensate after the chlorination at 170°C. Also in previous experiments it was demonstrated that important amounts of volatile UCl_5 and UCl_6 are formed during chlorinations at temperatures higher than 180°C. As a

consequence, for optimum efficiency of the chlorination, temperatures higher than 170°C should not be applied. In addition, a single chlorination step running for 20 hrs at 150°C is enough to sublimate the major part of produced AlCl_3 .

On the other hand, the experiments showed that too finely crushed input material can decrease the total efficiency of the reaction, as non-fully chlorinated agglomerates were formed inside the crucible.

The influence of working temperature on the efficiency of the chlorination process was evaluated during the experiments and future investigations will be focused on the optimisation of the molar ratio between $\text{Cl}_2(\text{g})$ and the alloy.

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