EVALUATION OF ALKALINE DISSOLUTION OF AI 6061 AND AI 1050 FOR THE PRODUCTION OF Mo-99 FROM LEU TARGETS

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ABSTRACT

Since 2008, due to the global crisis in the production of radioisotope ⁹⁹Mo, which product of decay, ^{99m}Tc, is the tracer element most often used in nuclear medicine and accounts for about 80% of all diagnostic procedures *in vivo*. Studies on the alkaline dissolution to obtain ⁹⁹Mo from irradiated UAl_x-Al LEU targets are under development. Processing time should be minimized, considering the short half-life of ⁹⁹Mo and ^{99m}Tc, about 66 h and 6 h, respectively. This makes dissolution time a significant factor in the development of the process. This paper presents the results of alkaline dissolution of "scraps" of Al 6061 and 1050, used to simulate the dissolution process of UAl_x-Al targets. Dissolution time and gas releasing were evaluated using the following alkaline solutions: a) NaOH 3 mol.L⁻¹ and NaNO₃ 2 mol.L⁻¹, b) NaOH 3 mol.L⁻¹ and NaNO₃ 4 mol.L⁻¹. The initial temperature of dissolution was 85°C in all cases. Al 6061 showed values of dissolution with NaNO₃ 2 mol.L⁻¹ showed that the gas releasing for Al 6061 was 2.7% greater than for Al 1050. However Al 1050 showed that gas releasing 9.92% greater than for Al 6061 during the dissolution with NaNO₃ 4 mol.L⁻¹. The decision about what type of alloy has to be used, Al 1050 or Al 6061, it will be up to the group that will manufacture the targets for the RMB.

1. INTRODUCTION

Radioisotopes play an important role among the peaceful uses of nuclear energy. In nuclear medicine radionuclides may be used for diagnosis and therapy. The ^{99m}Tc from the ⁹⁹Mo is the tracer element most commonly used in nuclear medicine by virtue of their favorable nuclear properties, accounting for 80% of all diagnostic procedures *in vivo* [1].

Currently, the supply of this important radioisotope is deficient due to the shutdown of the reactors in Canada and Belgium, the world's largest producers. With that Brazil had to look for other suppliers, such as Argentina and South Africa, to meet their needs, even partially [2]. In order to solve this problem of dependence from producing countries, Brazil develops now the Brazilian Multipurpose Reactor (RMB), initiated in 2008, for research in nuclear and an estimated production of 1,000 Ci/week of ⁹⁹Mo.

The line chosen for the production of ⁹⁹Mo is one that uses the UAl_x-Al targets which uses alkaline dissolution and purification of the product by chromatographic columns (adopted by Argentina) [3]. Brazil through RMB project aims to study and develop the process for the production of ⁹⁹Mo using the alkaline dissolution of UAl_x-Al targets, manufactured in that IPEN-CNEN/SP by the CCN (Centre of Nuclear Fuels). The subsequent steps of separation and purification of ⁹⁹Mo will also be developed at IPEN, until the product reaches the pharmaceutical specifications required for use in the diagnosis of several diseases.

2. GENERAL

This work is part of the research on alkaline dissolution of UAl_x -Al targets. The studies were conducted with scraps of Al used for manufacturing the targets. Al is about 79% of the total mass of the UAl_x -Al miniplates.

The alkaline dissolution is a well-established process and used by some of the 99 Mo producing countries. At the alkaline dissolution of UAl_x-Al targets, aluminum, some fission products and 99 Mo are soluble in this medium, while the uranium remains in the form of a precipitate, thus providing a first separation step [4].

The processing time should be as small as possible, considering that the half-life of 99 Mo is 66h and the half-life of 99m Tc is about 6h. This makes the parameter dissolution time a significant factor in the development of the process [5].

The dissolution of the scraps of Al in an alkaline medium may have different reaction products depending on the reagent used and other factors, such as temperature, concentration, molar ratio, etc.

During the process of dissolution, the release of hydrogen can cause problems with respect to the explosion and fire, which together with the released radioactive gases increases the radioactive risk of gas storage system. The release of hydrogen can be minimized with the addition of NaNO₃. The amount of NH₃ and NaNO₂ formed as reaction products depends on the amount of NaNO₃ in the reagent [6].

3. EXPERIMENTAL

3.1. Materials and Reagents

- ✓ Scraps of Al 1050and 6061;
- ✓ NaOH p.a.;
- ✓ NaNO₃ p.a.;
- ✓ 2L borosilicate glass flask;
- ✓ Thermocouple.

3.2. Dissolution of Aluminum

To simulate UAl_x -Al targets, hot dissolution studies with Al scraps were carried out. The dissolution time and the gas volume were chosen to evaluate the results, since it is an important parameter in the process development as a whole. The experiments were performed in triplicate to confirm reproducibility.

The study aims to determine if there are significant changes in the behavior of dissolution when Al alloys of different types are used in the manufacture of targets. The Center of Nuclear Fuels (CCN), requested an evaluation of the dissolution behavior of the alloys to help in decision of which one to employ to manufacture the targets [7].

These studies were performed using the following procedure: 1) Start the dissolution process with external heating of the solution NaOH 3 mol.L⁻¹ and NaNO₃ 2(or 4) mol.L⁻¹ until the initial temperature reaches 85°C; 2) Introduce in the heated solution the sample of Al to be dissolved. This study did not use cooling water bath.

4. RESULTS AND DISCUSSION

Aluminum alloys are widely used in various industrial applications, because of their high strength and solidity. The copper, magnesium and silicon are some of the best elements forming alloys with aluminum.

Aluminum has an important property for use in nuclear engineering: its low neutron absorption, what means it does not prevent the passage of neutrons, which helps to keep the nuclear reaction of uranium fuel, making it an effective material and of intensive use in the low temperature reactor core [7].

Al 1050 alloys have characteristics of commercially pure aluminum, very ductile in the annealed condition, suitable for cold forming. These alloys have excellent corrosion resistance, which increases with increasing purity of the alloy [7].

The Al 6061 alloy (Al, Mg and Si) are heat treatable, showing an excellent mechanical resistance in the T6 temper [7].

4.1. Study on the behavior of dissolution with NaOH 3 mol.L⁻¹ + NaNO₃ 2 mol.L⁻¹ solution of the samples of Al 6061 e Al 1050 related to dissolution time and released gases volume.

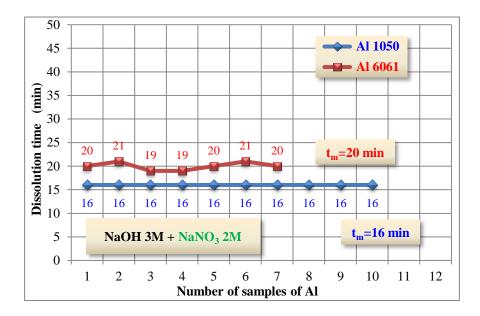


Figure 1: Study of dissolution time for Al 1050 e Al 6061, with NaOH 3 mol.L⁻¹ + NaNO₃ 2 mol.L⁻¹ solution.

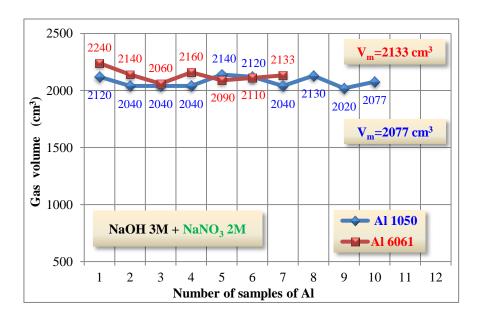


Figure 2: Study of gas volume for Al 1050 e Al 6061, with NaOH 3 mol.L⁻¹ + NaNO₃ 2 mol.L^{-1} solution.

4.2. Study on the behavior of dissolution of samples of Al 6061 e Al 1050 with NaOH $3mol.L^{-1} + NaNO_3 4 mol.L^{-1}$ solution related to dissolution time and released gases volume.

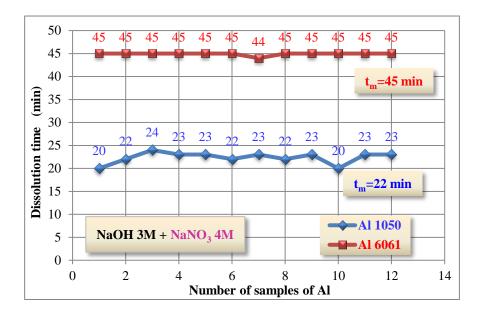


Figure 3: Study of dissolution time for Al 1050 and Al 6061, with NaOH 3 mol.L⁻¹ + NaNO₃ 4 mol.L⁻¹ solution.

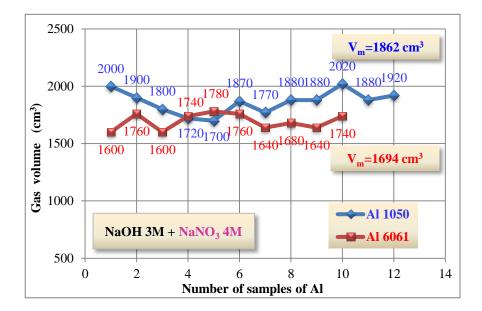


Figure 4: Study of the gas volume for Al 1050 and Al 6061, with NaOH 3 mol.L⁻¹ + NaNO₃ 4 mol.L⁻¹ solution.

5. CONCLUSIONS

The indication of the type Al alloy, 1050 or 6061, by the team of Al dissolution, was based on the results of the dissolution time, the final aspect of the solution after the dissolution, the

economy of reagents and also the chemical composition of the alloys Al 1050 (Figure 5) and Al 6061 (Figure 6).

Al 6061 showed values of dissolution time 25% greater than the Al 1050, and volume of gases was 2.7% higher than the Al 1050, when NaOH 3 mol.L⁻¹ + NaNO₃ 2 mol.L⁻¹ was used as the reagent dissolution.

Al 6061 showed values of dissolution time 104,55% greater than the Al 1050, and volume of gases was 9,92% less than the Al 1050, when NaOH 3 mol.L⁻¹ + NaNO₃ 4 mol.L⁻¹ was used as the reagent dissolution.

These results when compared to the chemical composition of the alloys Al 1050 and Al 6061, helped indicate the choice of Al 1050 for use in the manufacture of the targets of UAl₂-Al.

Preliminary tests of manufactured targets will be important for the decision of CCN regarding the choice of which alloy will be used in the manufacture of targets.

Chemical Composition of Alumi	nium Alloy 1050
Table 1. Typical chemical composition for aluminium alloy 1050	
Element	% Present
Cu	0-0.05
Mg	0-0.05%
Si	0-0.25%
Fe	0-0.4%
Mn	0-0.05%
Zn	0-0.07%
ті	0-0.05%
AI	Balance

Figure 5: Typical chemical composition of Al 1050 alloy.

Component	Amount (wt.%)
Aluminium	Balance
Magnesium	0.8-1.2
Silicon	0.4 - 0.8
Iron	Max. 0.7
Copper	0.15-0.40
Zinc	Max. 0.25
Titanium	Max. 0.15
Manganese	Max. 0.15
Chromium	0.04-0.35
Others	0.05

Figure 6: Typical chemical composition of Al 6061 alloy.

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