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DISSOLUTION OF TARGETS FOR THE PRODUCTION OF MO-99: PART 2. INFLUENCE OF EXCESS OF REAGENT AND ALUMINUM MASS ON DISSOLUTION TIME

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ABSTRACT

Radioisotopes play an important role in the peaceful uses of nuclear energy. Radionuclides in nuclear medicine can be used for diagnosis and therapy. The ^{99m}Tc, daughter of ^{99}Mo, is most often used in nuclear medicine as tracer element because of its favorable nuclear properties, accounting for about 80% of all diagnostic procedures *in vivo*. Nowadays, the supply of this important isotope is deficient, due to the shutdown of the reactors in Canada and Belgium, the world's largest producers. Aiming to resolve the dependency of Brazil with respect to the supply of ^{99}Mo was created the Brazilian Multipurpose Reactor project (RMB), started in 2008, having as main objective to produce about 1000 Ci/week of ^{99}Mo. This study is part of the project to obtain ^{99}Mo by alkaline dissolution of UAl_x-Al targets. Al, which corresponds to 79% of the total mass, was used to simulate the targets. The reagent used in the dissolution was a 3 mol.L^{-1} $NaOH/NaNO_3$ solution and initial temperature of 88°C. The influence of the reagent on the dissolution time, related to the molar ratio $1Al:1.66NaOH:1.66NaNO_3$, for a range of over 5 to 50%, and the influence of the amount of mass in a range from 16 to 48 g of Al were studied. It was observed that the dissolution time was shorter with the percentage of excess of 30% and the increase of the mass did not change the dissolution time.

1. INTRODUCTION

In order to solve the problem of dependence on the ^{99}Mo producing countries, Brazil develops the project Brazilian Multipurpose Reactor (BMR), which began in September 2008. The project aims to achieve a production estimated at 1000 Ci/week of ^{99}Mo.

Radionuclides in nuclear medicine can be used for diagnosis and therapy. ^{99m}Tc, the product of radioactive decay of ^{99}Mo, is the tracer element most commonly used in nuclear medicine, accounting for about 80% of all diagnostic procedures *in vivo.* This is due to its favorable nuclear properties. Radioisotopes play an important role among the peaceful uses of nuclear energy. Currently, due to the shutdown of the reactors in Canada and Belgium, countries that provide this important radioisotope, the supply is deficient. By this fact, Brazil had to seek other suppliers, such as Argentina and South Africa, to meet their needs partially.

The project BMR with respect to the fuel dissolution studies, aims to study two lines of research for obtaining ^{99}Mo via the fission of ^{235}U. The two surveys cover different types of targets with low enrichment (<20% of ^{235}U). The first studies will be with UAl_x-Al targets via alkaline dissolution and the second with metallic uranium foil via acid dissolution.

The Nuclear Fuel Center (NFC) of Nuclear and Energy Research Institute (IPEN-CNEN/SP) mastered the technology of manufacturing UAl_x-Al miniplates, for this reason the studies preferably started with this type of target.

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.

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

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. The equations involved in the process of dissolution [1] can be seen below:

$$Al + NaOH + H_2O \rightarrow NaAlO_2 + 1.5H_2 \quad (1)$$

$$Al + 0.5NaOH + 0.5NaNO_2 + 0.5H_2O \rightarrow NaAlO_2 + 0.5NH_3 \quad (2)$$

$$Al + 0.625NaOH + 0.375NaNO_3 + 0.25H_2O \rightarrow NaAlO_2 + 0.375NH_3 \quad (3)$$

$$Al + NaOH + 1.5NaNO_3 \rightarrow NaAlO_2 + 0.5H_2O + 1.5NaNO_2 \quad (4)$$

$$Al + 0.85NaOH + 1.05NaNO_3 \rightarrow NaAlO_2 + 0.9NaNO_2 + 0.15NH_3 + 0.2H_2O \quad (5)$$

During the process of dissolution, according to equation (1), 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 [2].

The release of hydrogen can be minimized with the addition of $NaNO_3$. The amount of NH_3 and $NaNO_2$ formed as reaction products depends on the amount of $NaNO_3$ in the reagent. It can be seen in reactions 3-5.

3. EXPERIMENTAL PART

3.1. Materials and Reagents

- ✓ scraps of Al;
- ✓ NaOH p.a.;
- ✓ $NaNO_3$ p.a.;
- ✓ 2 L 3-neck borosilicate glass flask;
- ✓ jacketed borosilicate glass condenser with coil;
- ✓ thermostatic bath;
- ✓ water bath;
- ✓ thermocouple.

3.2. Dissolution of Aluminum

To simulate UAl_x-Al targets, hot dissolution [2] tests (88°C) of Al with alkaline solution of 3 $mol.L^{-1}$ NaOH/$NaNO_3$ and molar ratio of 1AL:1.66NaOH:1.66$NaNO_3$ were carried out. Initially, the variation of the reagent 5 to 50% was studied. Next, the variation of the mass of Al remaining constant the molar ratio in 1Al:1.66NaOH:1.66$NaNO_3$ was evaluated [3]. Finally, the best condition for the dissolution of a UAl_x-Al miniplate was chosen. The experiments were performed in triplicate to confirm reproducibility.

4. RESULTS AND DISCUSSION

The excess of reagent with 1Al:1.66NaOH:1.66$NaNO_3$ molar ratio was studied. A 3 $mol.L^{-1}$ NaOH/$NaNO_3$ solution and an initial temperature of 88°C for a range of 5 to 50% were used. Fig. 1 shows that the dissolution time was shorter when a 30% reagent excess was used.

The influence of the mass amount of Al on the time of dissolution for a range of 16 to 48g was studied. A 3 $mol.L^{-1}$ NaOH/$NaNO_3$ solution and an excess of 30% over 1Al:1.66NaOH:1.66$NaNO_3$ molar ratio and initial temperature of 88°C were used. The results indicated that the increase of the mass did not change the time of dissolution. The result can be seen in Fig. 2.

To evaluate the conditions of dissolution defined so far, two dissolutions were performed with UAl_x-Al targets. The following initial conditions were used: a) 3 $mol.L^{-1}$ NaOH/$NaNO_3$ solution, b) excess of 30% over 1Al:1.66NaOH:1.66$NaNO_3$ molar ratio, c) initial dissolution temperature of 88°C [3]. The dissolution times were 14 min for UAl_x-Al targets and 13 min for the Al plates. The results can be seen in Fig. 3.

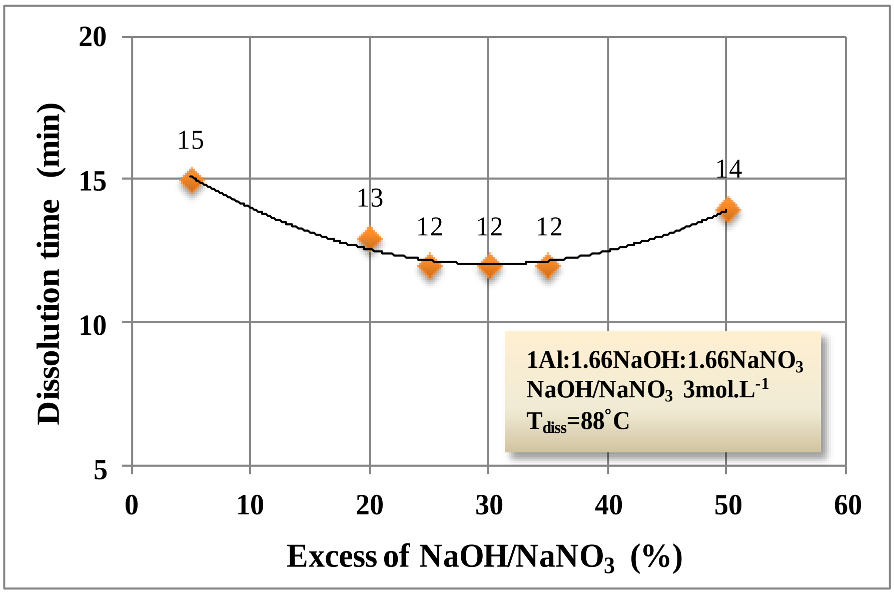

Figure 1. Influence of 3 mol.L^{-1} NaOH/$NaNO_3$ reagent on the time of dissolution for the molar ratio of 1Al:1.66NaOH:1.66$NaNO_3$.

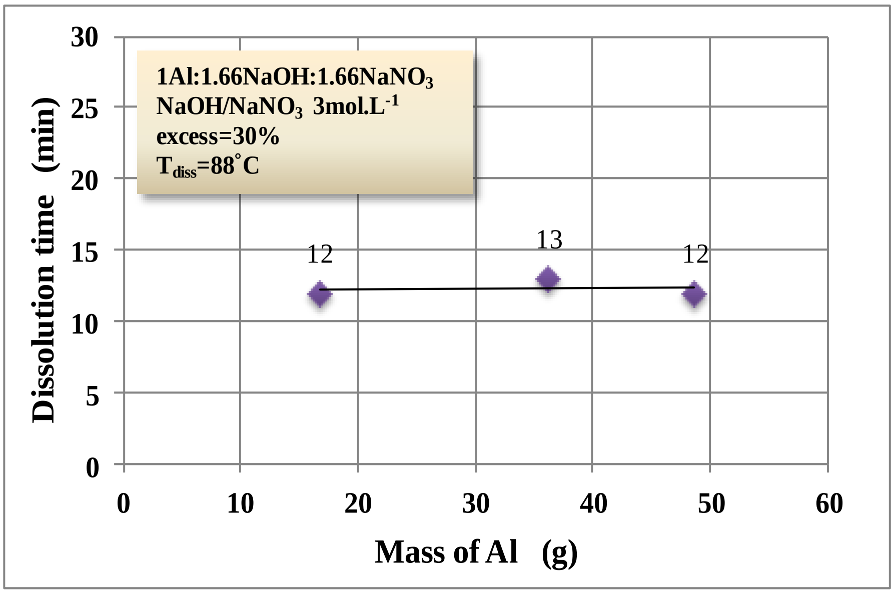

Figure 2. Influence of mass increase in Al dissolution time for 3 mol.L^{-1} NaOH/$NaNO_3$ and 1AL:1.66NaOH:1.66$NaNO_3$ molar ratio.

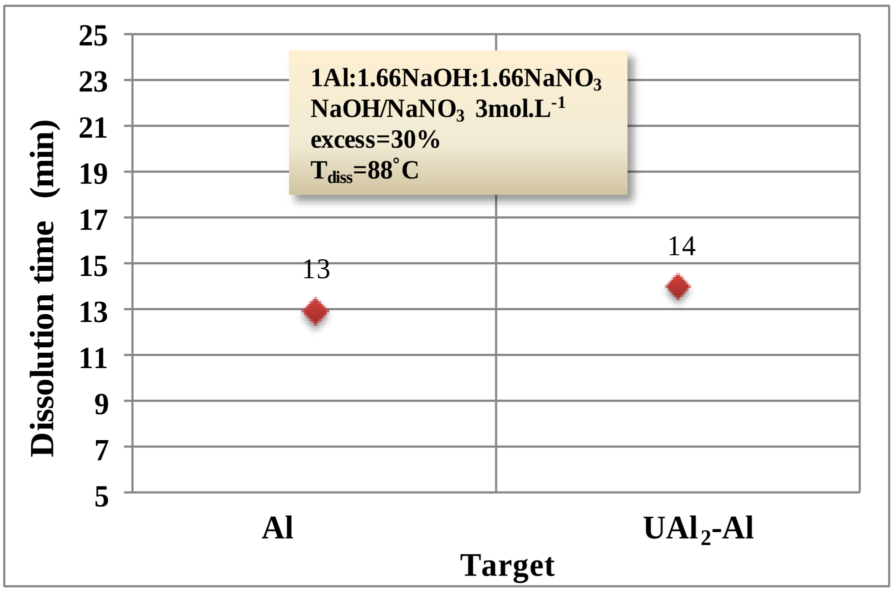

Figure 3. Dissolving time of UAl_x-Al targets and Al with 3 $mol.L^{-1}$ $NaOH/NaNO_3$ solution and $1Al:1.66NaOH:1.66NaNO_3$ molar ratio using 30% in excess.

5. CONCLUSION

Shorter time of dissolution was verified with reagent excess of 30% compared to the $1Al:1.66NaOH:1.66NaNO_3$ molar ratio, which amounts to a final molar ratio of $1Al:2.16NaOH:2.16NaNO_3$. The increasing of Al mass in the range studied did not increase the dissolution time in the same proportion. This was demonstrated when a plate of UAl_x-Al was dissolved.

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