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STUDIES ON THE GASES EVOLUTION DURING THE ALKALINE DISSOLUTION OF UAL_2-AL LEU TARGETS FOR THE PRODUCTION OF ^{99}MO IN BRAZIL

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ABSTRACT

Since 2009, Brazil is developing the Brazilian Multipurpose Reactor (RMB), which goal is the construction of a nuclear reactor for research and production of radioisotopes, including ^{99}Mo from LEU targets. For this purpose, in the present work, the volume of gases released during the alkaline dissolution of 6061 aluminum (to simulate UAl_2-Al LEU targets) using molar ratio of NaOH, and $NaOH/NaNO_2$ was evaluated. This parameter is also important since some radioactive gases that evolve with H_2 cannot be released directly to the atmosphere. For the dissolution with NaOH a linear increase in the gases volume (400-1880mL) with the aluminum mass (0.15-0.81g) was observed, and the dissolution time varied from 22 to 29 minutes, independently of the Al mass. The use of $NaOH/NaNO_2$ solution showed a logarithmic increase of the gases volume relating to aluminum mass (0.24-3.5g), and also reduced both the gases volume (310-1520mL) and the dissolution time (11-12min).

1. Introduction

Faced with global crisis in the production of radioisotope ^{99}Mo, 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, since 2009 Brazil is developing the project called Brazilian Multipurpose Reactor (RMB). Within the Brazilian Nuclear Program (PNB) the construction of the RMB, is seen as a long term solution to meet all domestic demand relative to supply of radioisotopes and radiopharmaceuticals. The project will enable the development of production technology of fission ^{99}Mo from targets of low-enriched uranium (LEU) for nuclear research in the area.

The targets to be used in RMB for the production of ^{99}Mo are made of UAl_x, with high percentage of UAl_2, about 81.5% enriched to 19.75 ± 0.2 wt% in ^{235}U and coated with an alloy of aluminum (6061) . The fabrication of the targets by the CCN (Center of Nuclear Fuel) at IPEN-CNEN (Institute for Energy and Nuclear Research-National Commission of Nulear Energy), uses a technique called "picture-frame" consisting of briquette, frame and coating.

In IPEN / CNEN two lines of research with different types of targets (UAl_2-Al and metallic U) will be studied for the development of production technology of ^{99}Mo from LEU targets. The alkaline line uses UAl_2-Al targets, where Al is in greater quantity at the target, and can be dissolved from the following solutions: NaOH, $NaOH/NaNO_3$, $NaOH/NaNO_2$ or KOH.

The alkaline dissolution of the coating (Al) follows a first order kinetics [2,3], represented by the following equation:

$$Al + NaOH + H_2O \rightarrow NaAlO_2\ 3/2H_2 \quad (1)$$

The release of hydrogen can be minimized by adding $NaNO_2$, according to the following equation:

$$Al + NaOH + 1/2NaNO_2 + H_2O \rightarrow 1/2NaAlO_2\ NH_3 \quad (2)$$

The overall process for the recovery and purification of ^{99}Mo present in the UAl_2-Al target, after irradiation in the reactor, involves the following steps: dissolution, filtration, recovery and purification of ^{99}Mo. The processing time should be minimized, considering that the half life of ^{99}Mo is 66h and the half life of ^{99m}Tc is about 6.01 h. This makes the dissolution time a significant parameter in the process development. This work is part of the research of alkaline dissolution of UAl_2-Al targets. The experiments were carried out with scraps of 6061 Al, commonly used to make the targets, while Al is about 79% of the total weight of UAl_2-Al.

2. Materials and Methods

In alkaline dissolution of scraps of Al (6061) with 3M NaOH [1.4], the mass interval from 0.15 to 0.81g were used and 1 Al : 2.16 NaOH molar ratio [5]. For the dissolution with $NaOH/NaNO_2$ 3M solution, the mass range was 0.24 to 3.5g and the molar ratio was 1AL: 2.16 NaOH: 2.16 $NaNO_2$. The initial temperature of dissolution in all experiments was 88°C and complete dissolution arrangement consisted of a reactor with a capacity of 2L, a glass double condenser coil and jacketing, and glass thermometer.

The volume of gas released in each experiment was collected in a graduated glass column with a capacity of 2000 ± 20 mL, which allowed the reading of the values found.

3. Results e Discussion

Figure 1 shows that the volume of gas released during the dissolution of Al (6061) with 3M NaOH ranged from 400 to 1880mL. In this case the increase was linear over the range of mass studied (0.15-0.81g). Fig 2 shows that the addition of $NaNO_2$ reduced the volume of gas released. During the dissolution of Al (6061) with the solution of 3M $NaOH/NaNO_2$ the volume of gas ranged from 310 to 1520mL. The curve that represents the increase of volume compared with the mass of Al showed a logarithmic behavior. In this case it was not possible to verify the composition of the gas released.

Figure 3 shows the variation in dissolution time depending on the weight of Al and the reagent used. One can see that the dissolution time varied from 22 to 29 minutes with 3M NaOH solution, however, when we used the solution $NaOH/NaNO_2$ of 3M (Fig. 4), besides the reduction in the volume of gas released; there was a significant reduction in the time of dissolution, in which case the dissolution time was almost constant, around 12 minutes.

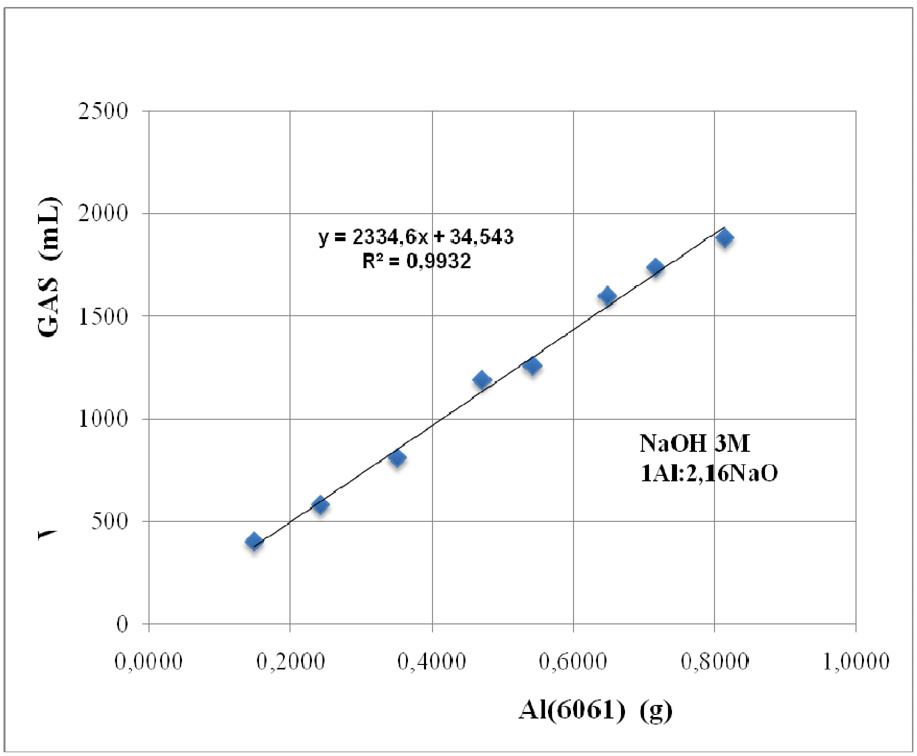

FIGURE 1: Influence of the mass of Al (6061) in the volume of gas released with 3M NaOH and molar ratio of 1AL: 2.16 NaOH.

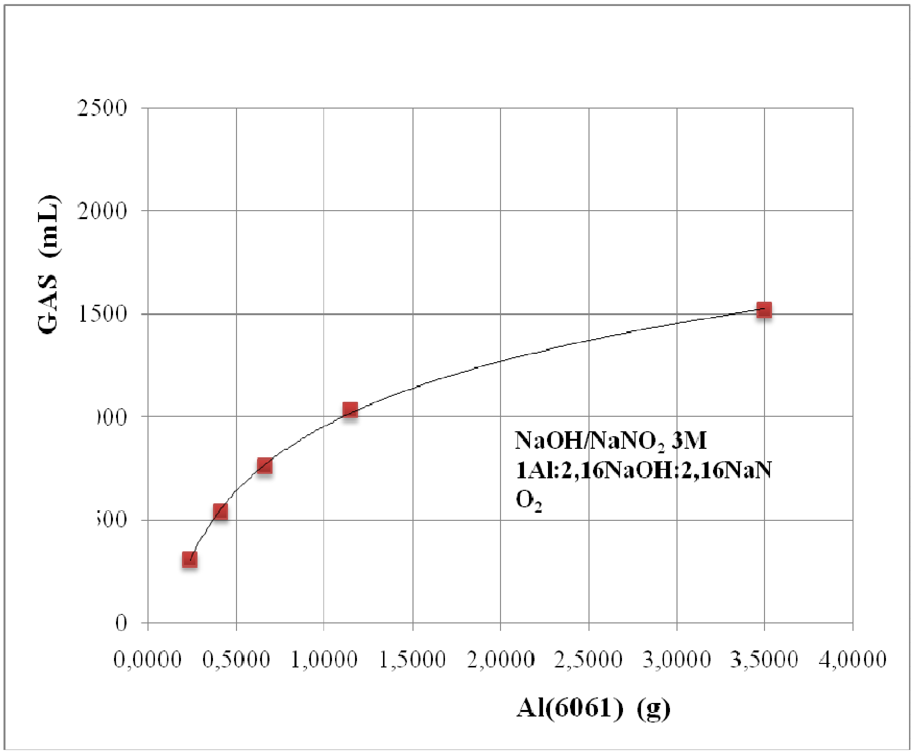

FIGURE 2: Influence of the mass of Al (6061) in the volume of free gas with a solution of 3M and $NaOH/NaNO_2$ 1AL molar ratio: 2.16 NaOH: $NaNO_2$.

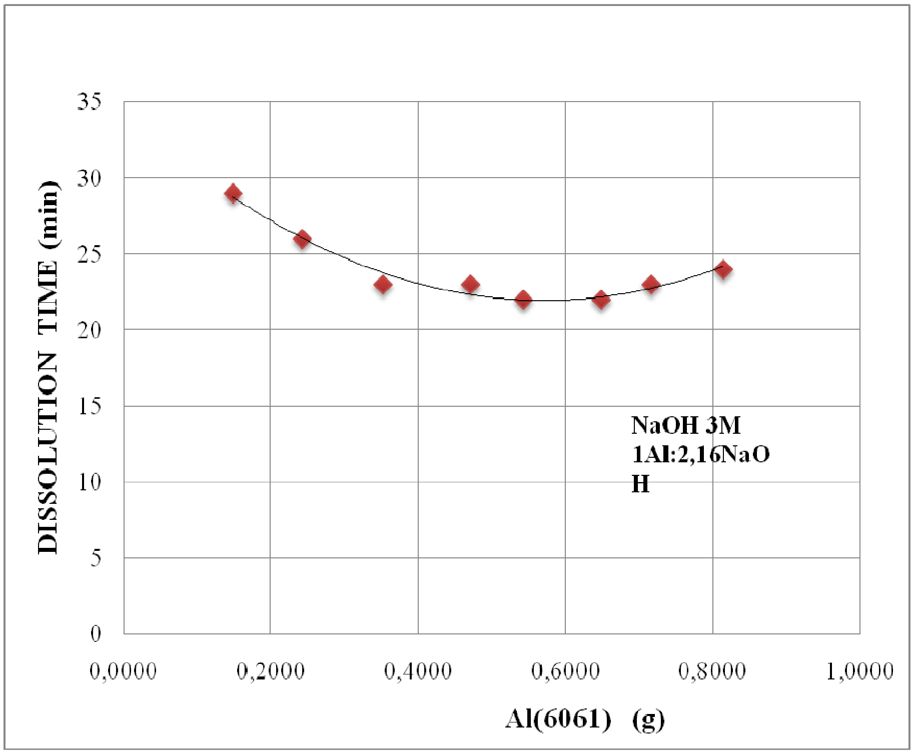

FIGURE 3: Influence of the mass of Al (6061) at the time of dissolution with NaOH 3M and 1Al molar ratio: 2.16 NaOH.

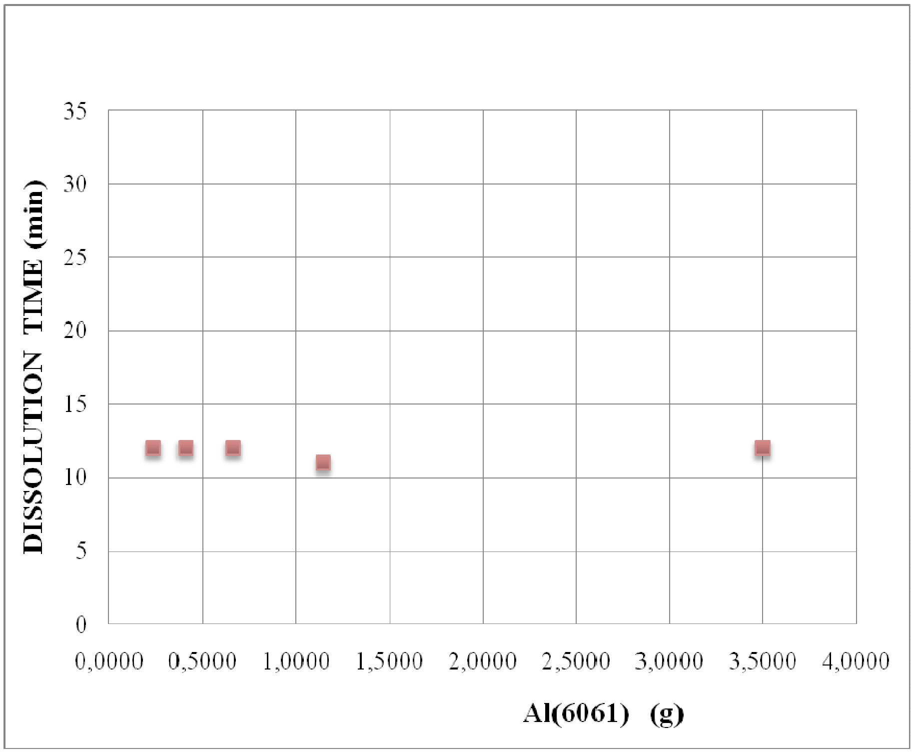

FIGURE 4: Influence of the mass of Al (6061) over the time of dissolution with a solution of 3M $NaOH/NaNO_2$ 1Al molar ratio: 2.16 NaOH: $NaNO_2$.

4. Conclusions

The dissolution experiments indicated that there was an intense and vigorous gas generation when we use only with 3M NaOH reagent dissolution. When the dissolution was performed with a mixture NaOH/NaNO2 3M, a significant improvement in the volume of gas released as well, a great reduction in the time of dissolution.

5. Acknowledgement

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6. References

[1] A.A. Sameh, H.J. Ache, **"***Production Techniques of Fission Molybdenum-99***"**, Radiochimica Acta, 41, 1987.

[2] D.L. Foster, J.E. Savolainen, R.G.Wymer, "Nuclear Reactor Fuel Dissolution." In: Peaceful Uses of Atomic Energy: Proceedings of the International Conference held in New York, v. 9, p. 546-550, Reactor Technology and Chemical Processing, Aug 1956.

[3] BLANCO, R. E. and WATSON, C. D., Head-end Processes for Solid Fuels. In: STOLLER, S. M. & RICHARDS, R. B. eds Reactor Handbook, v. 2: Fuel Reprocessing.: 2ª ed. New York, Interscience, 1961.

[4] B. A. Buchholz and G. F. Vandegrift, "*PROCESSING OF LEU TARGETS FOR 99MO PRODUCTION --DISSOLUTION OF* U_3SI_2 *TARGETS BY ALKALINE HYDROGEN PEROXIDE***",** International Meeting on Reduced Enrichment for Research and Test Reactors, Paris, France, September 18-21, 1995.

[5] P.R. Cristini, H.J. Cols, R. Bavaro, M. Bronca, R. Centurión, D. Cestau, "*PRODUCTION OF MOLYBDENUM-99 FROM LOW ENRICHED URANIUM TARGETS***",** International Meeting on Reduced Enrichment for Research and Test Reactors, Bariloche, Argentina, November 3-8, 2002.