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INFLUENCE OF INITIAL TEMPERATURE AND HEATING METHOD IN THE TEMPERATURE PROFILE DURING ALKALINE DISSOLUTION OF Al FOR THE PRODUCTION OF Mo-99

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ABSTRACT

Radionuclides in nuclear medicine can be used for diagnosis and therapy. The ^{99m}Tc, son 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. Aiming to resolve the dependency of Brazil with respect to the supply of ^{99}Mo was created the Brazilian Multipurpose Reactor project (BMR), 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. The initial reaction temperature is an important parameter, since it has great influence on the value of the maximum temperature and dissolution time. According to literature, for security reasons the dissolution process must have its temperature controlled so that the maximum temperature has to be around 90°C. The behavior of the temperature during dissolution using three different methods of heating in order to minimize the fluctuation of temperature during dissolution, keeping its maximum value at around 90°C was studied. The three methods of heating chosen were: a) initial temperature of 85ºC with continuous heating, b) heating water bath until it reaches the initial temperature (70 to 95ºC), turning off after that, and c) external heating until it reached the starting temperature (60-95°C). The alkaline solution used was 3 mol.L^{-1}NaOH and 2 mol.L^{-1} $NaNO_3$. In the first study it was observed that after 1 minute of dissolution the solution temperature reached 100°C on average, up to a maximum of 109°C, ending with values around 95°C. In the second study after 3 minutes of dissolution the maximum temperature was 106ºC and the minimum 100ºC. In the third study the temperature rise during dissolution increased with increasing initial temperature which practically remains constant until the end of the dissolution.

1. INTRODUCTION

Radioisotopes play an important role among the peaceful uses of nuclear energy. Radionuclides in nuclear medicine can be used for diagnosis and therapy. 99mTc from 99Mo is the tracer element most commonly used in nuclear medicine because of its favorable nuclear properties, accounting for about 80% of all diagnostic procedures *in vivo*. 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. So Brazil had to seek other suppliers, such as Argentina and South Africa to meet their needs, even partially.

In order to solve this problem of dependence on the producing countries, Brazil develops now the project named Brazilian Multipurpose Reactor (BMR), which began on September 2008, for research in nuclear area and for the production of ^{99}Mo, estimated in 1,000 Ci/week.

Within this project we intend to study two lines of research to obtain ^{99}Mo from the fission of ^{235}U. The two surveys cover different types of targets with low enrichment (< 20% of ^{235}U). The first study will be with targets of UAl_x-Al via alkaline dissolution and the second with metallic uranium foils via acid dissolution.

The Nuclear Fuel Center (NFC) at Nuclear and Energy Research Institute (IPEN-CNEN/SP) dominates the manufacturing technology of UAl_x-Al miniplates, for this reason the studies preferably started with this type of target. So far there is no country that produces ^{99}Mo from metallic uranium targets commercially. Among the countries that are studying the fabrication of this type of target, South Korea and more recently USA have the knowledge of this manufacturing technique and may likely be distributors for other users.

2. GENERAL

This work is part of the research on alkaline dissolution of UAlx-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 UAlx-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 UAlx-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.

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 $NaNO_3$. The amount of NH_3 and $NaNO_2$ formed as reaction products depends on the amount of $NaNO_3$[1] in the reagent.

3. EXPERIMENTAL

3.1. Materials and Reagents

- ✓ Scraps of Al (1050);

- ✓ NaOH p.a.;
- ✓ $NaNO_3$ p.a.;
- ✓ 3L jacketed borosilicate glass flask;
- ✓ Thermocouple.

3.2. Dissolution of Aluminum

The dissolution time and the gas volume were chosen to evaluate the results, since they are important parameters in the process development as a whole. To simulate UAlx-Al targets, hot dissolution studies with Al 1050 scraps were carried out. The experiments were performed in triplicate to confirm reproducibility.

The temperature profile during the dissolution is a function of the initial temperature of the reagent and the kind of cooling or heating. In this study three different methodologies for the purpose of maintaining the maximum temperature during dissolution in around 90°C were used:

1. The first method comprises heating the alkaline solution in water bath until the established initial temperature (85°C) [2]; keeping the water bath connected during the entire dissolution (97°C).

2. The second method comprises heating the alkaline solution in water bath until the established initial temperature (70, 75, 80, 85, 88, 90 and 95°C), turning off the water bath after the solution reaches the desired temperature.

3. The third method comprises the external heating of the solution to the established initial temperature (60, 65, 70, 75, 80, 85, 88 and 95°C), followed by transferring the solution to the reactor without any external heating, only in contact with air at room temperature.

4. EXPERIMENTAL

4.1. Results obtained with the first methodology for Al 1050 samples and NaOH 3mol.L^{-1} + $NaNO_3$ 2 mol.L^{-1} dissolution solution at initial temperature of 85ºC and continuous heating by water bath at 97ºC

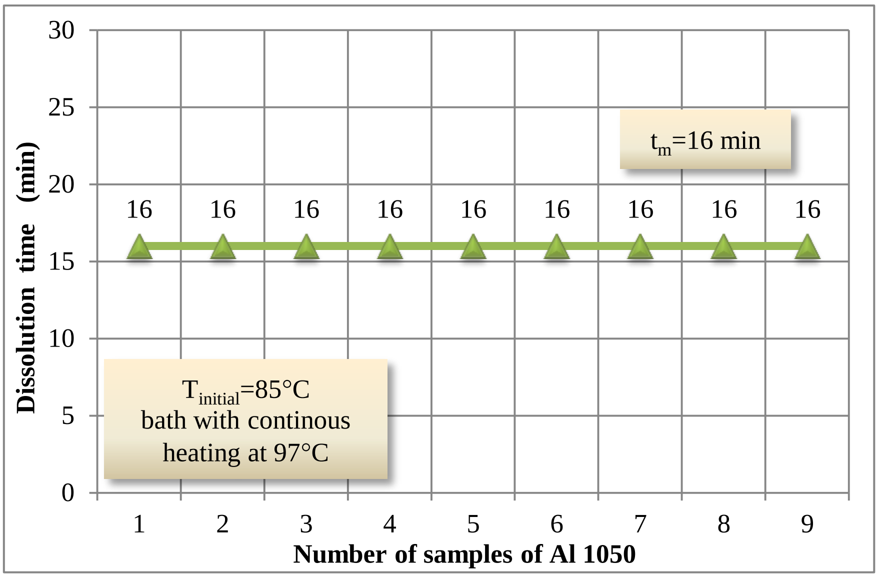

Figure 1: Dissolution time of Al 1050 samples in NaOH 3mol.L^{-1} + $NaNO_3$ 2 mol.L^{-1} solution with initial temperature of 85°C and water bath at 97°C.

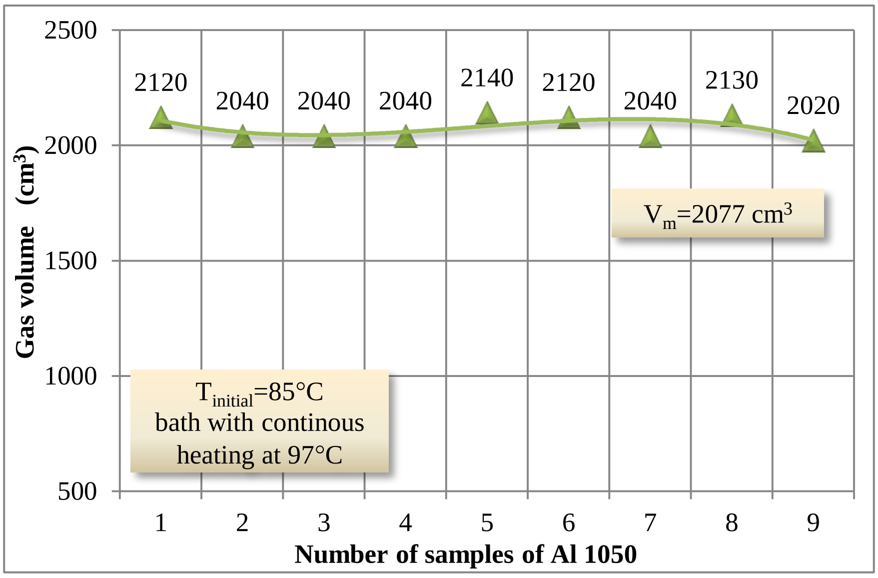

Figure 2: Gas volume in the dissolution of Al 1050 samples in NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} solution with initial temperature of 85°C and water bath at 97°C.

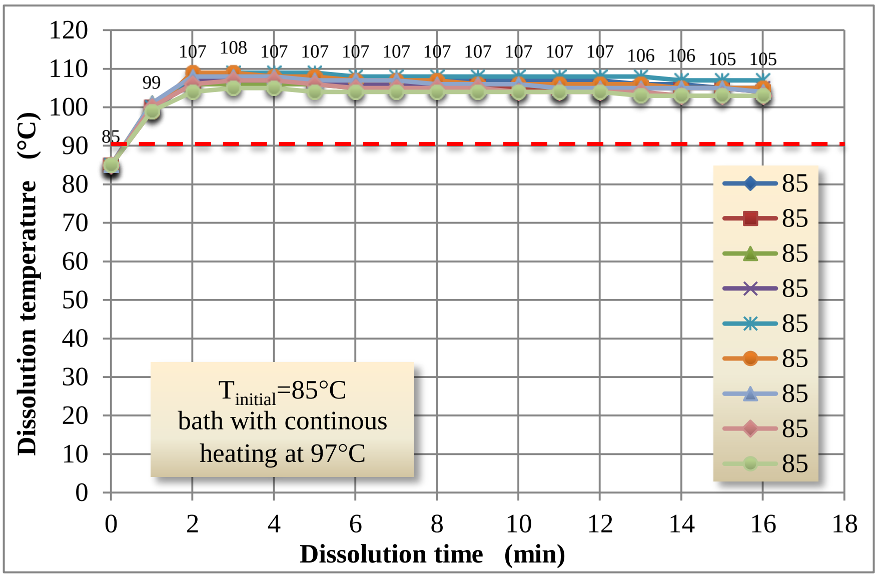

Figure 3: Temperature profile during the dissolution of Al 1050 samples in NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} solution with initial temperature of 85°C and continuous heating by water bath at 97°C

4.2. Results obtained with the second methodology for Al 1050 samples and NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} dissolution solution at initial temperature from 70 to 95°C and water bath being turned off after reaching the established temperature

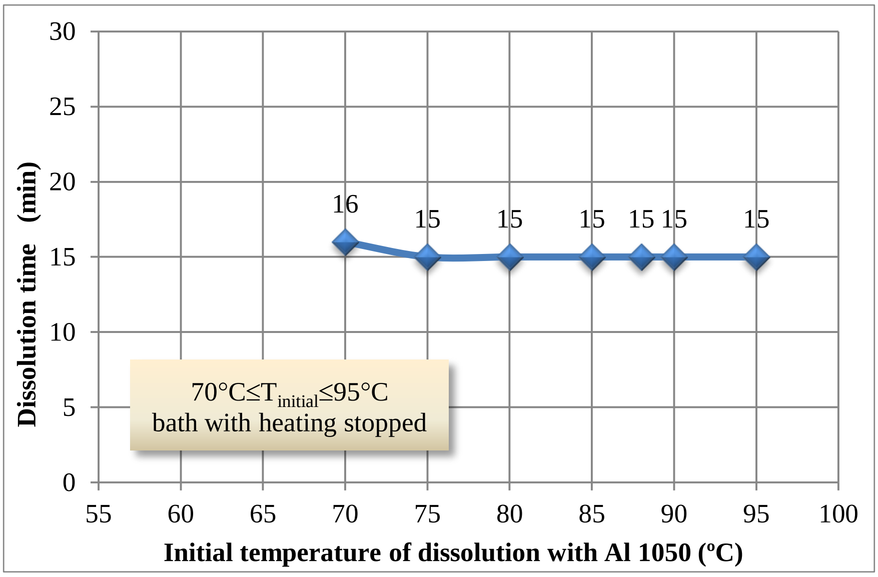

Figure 4: Dissolution time for Al 1050 samples and NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} dissolution solution at initial temperature from 70 to 95°C and water bath being turned off after reaching the established temperature.

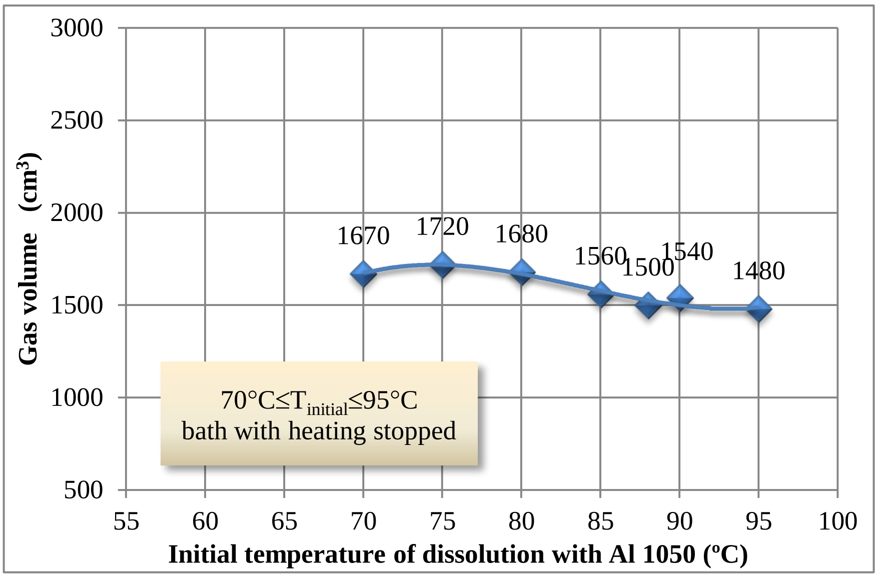

Figure 5: Gas volume for Al 1050 samples and NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} dissolution solution at initial temperature from 70 to 95ºC and water bath being turned off after reaching the established temperature.

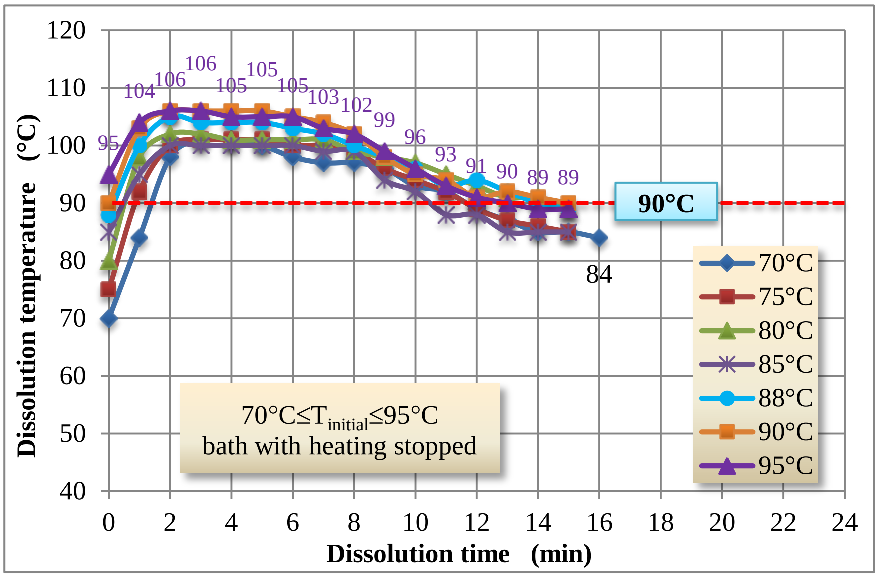

Figure 6: Temperature profiles for Al 1050 samples and NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} dissolution solution at initial temperature from 70 to 95ºC and water bath being turned off after reaching the established temperature.

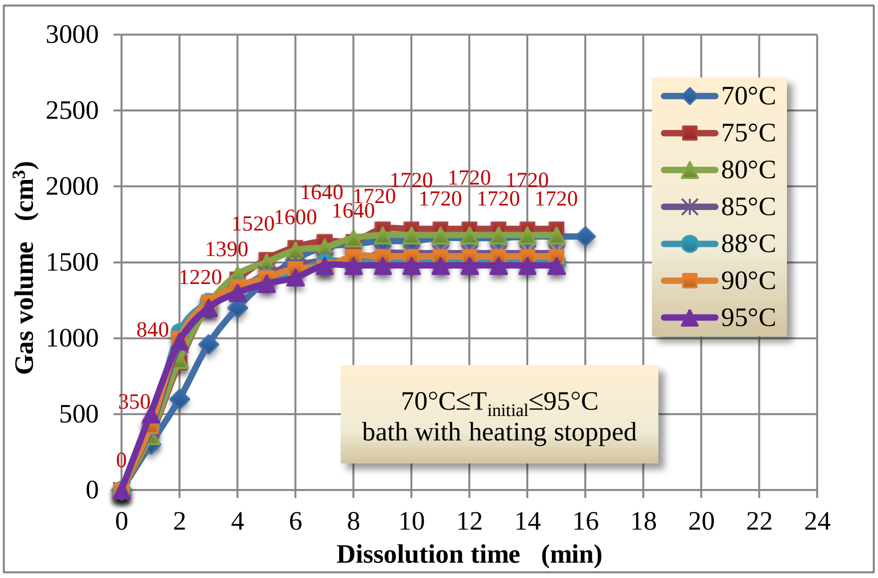

Figure 7: Gas volume profile for Al 1050 samples and NaOH 3mol.L^{-1} + NaNO$_3$ 2mol.L^{-1} dissolution solution at initial temperature from 70 to 95°C and water bath being turned off after reaching the established temperature.

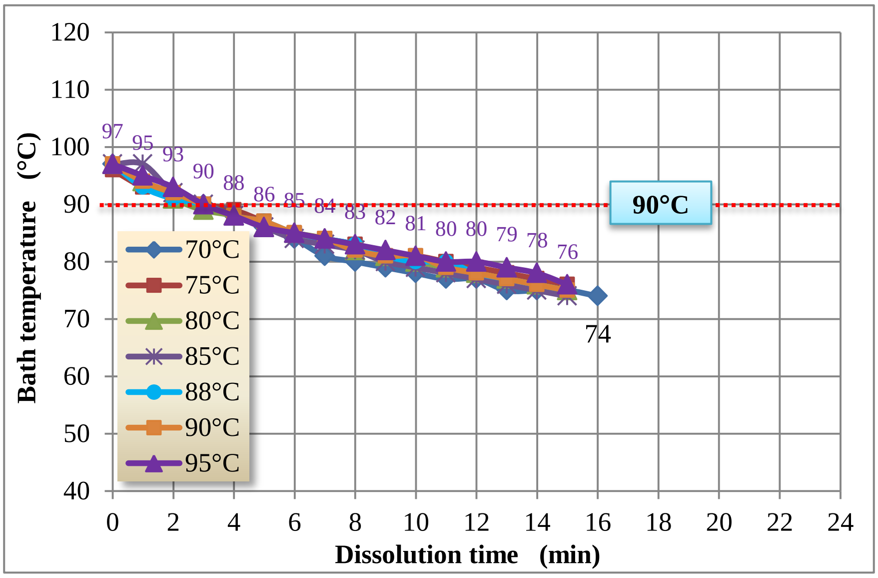

Figure 8: Temperature profile of water bath after being turning off after reaching the established temperature for Al 1050 samples and NaOH 3mol.L^{-1} + NaNO$_3$ 2mol.L^{-1} dissolution solution at initial temperature from 70 to 95°C.

4.3. Results obtained with the third methodology for Al 1050 samples and NaOH 3mol.L^{-1} + NaNO$_3$ 2mol.L^{-1} dissolution solution at initial temperature from 60 to 95°C, without external heating, only in contact with air at room temperature.

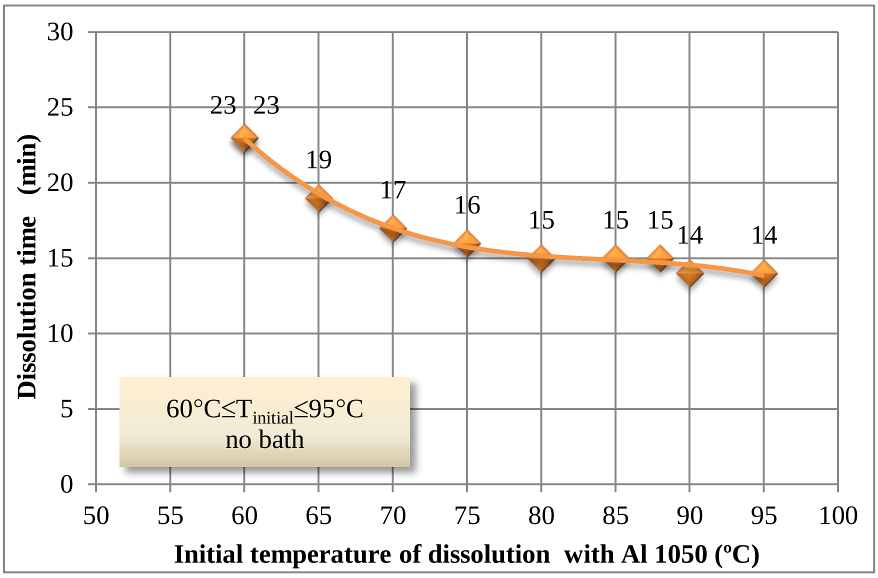

Figure 9: Dissolution time for Al 1050 samples and NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} dissolution solution at initial temperature from 60 to 95°C, without external heating.

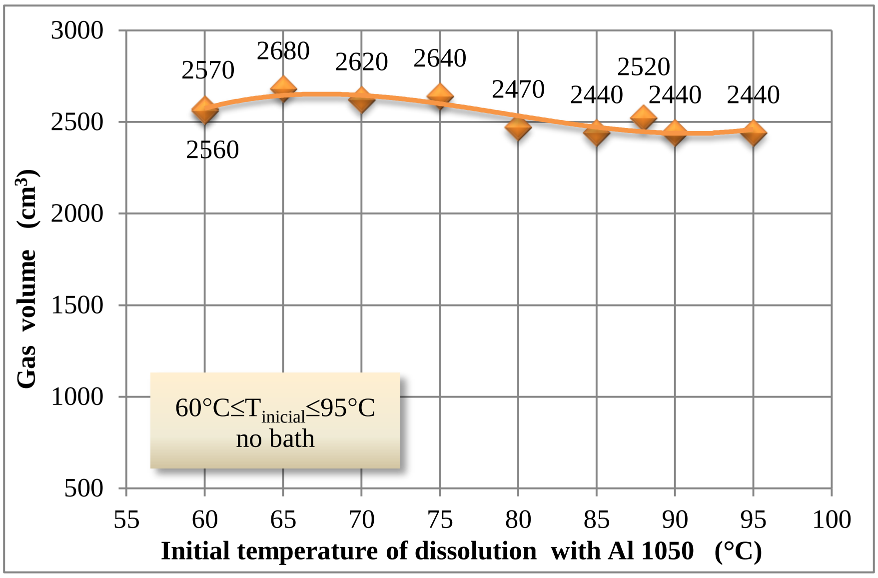

Figure 10: Gas volume for Al 1050 samples and NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} dissolution solution at initial temperature from 60 to 95°C, without external heating.

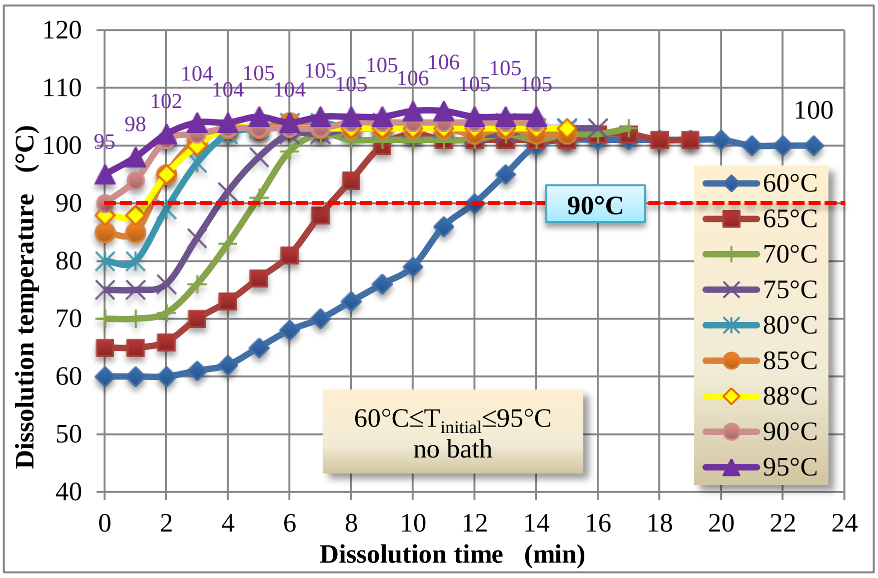

Figure 11: Temperature profile during dissolution of Al 1050 samples in NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} solution at initial temperature from 60 to 95ºC, without external heating.

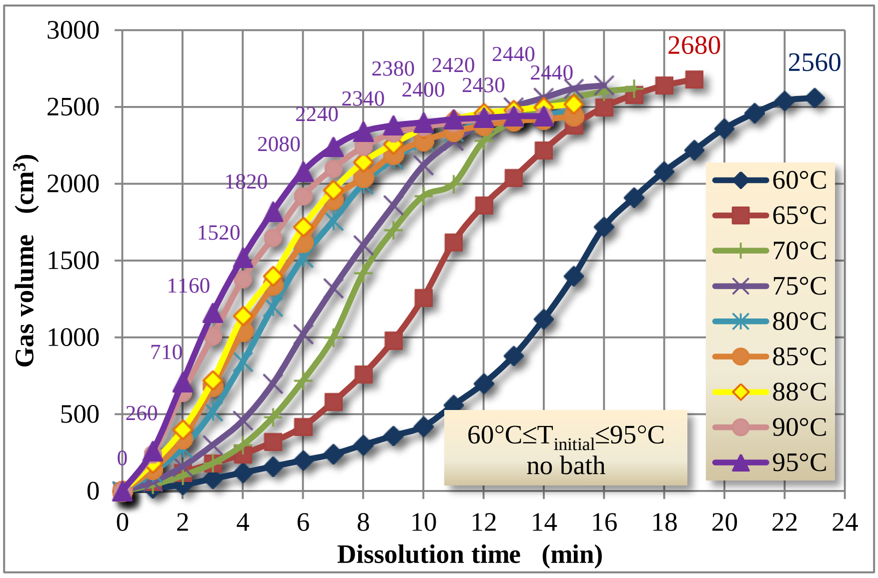

Figure 12: Gas volume profile during dissolution of Al 1050 samples in NaOH 3mol.L^{-1} + $NaNO_3$ 2mol.L^{-1} solution at initial temperature from 60 to 95ºC, without external heating.

5. CONCLUSIONS

The initial temperature of the process has an important role in the temperature profile during the dissolution, since from a certain minimum initial temperature of the dissolution process is exothermic.

In all experiments that used the first method (Fig. 1 to 3), the temperature profile in Figure 3 showed that in 1 minute the solution temperature reached 99°C on average, up to values around 108°C and ending with values around 105ºC. The dissolution time in all experiments was 16 minutes (Fig. 1) and the average volume of the gases (Fig.2) was 2077 cm^3.

In all experiments (in triplicate) using the second method (70 to 95°C), it was noted that after 2 minutes the solution reached 99°C on average, the values varying up to a maximum of 106°C and a minimum of 84ºC. The dissolution time was 15 minutes at temperatures of 75 to 95ºC, but at a temperature of 70°C the time was 16 minutes [3].

It is observed in the temperature profile of Figure 3 that the decrease in temperature is slow because the bath is kept on during the entire dissolution, unlike Figure 6, where the temperature is turned off immediately after the reagent reaches the desired temperature. In this case the reduction is more pronounced because of heat exchange between the bath (Fig. 8) and the reagent.

In all experiments in which the third method was used (60 to 95°C), it was observed that the maximum temperature achieved during the dissolution was 106°C and the final values of temperature ranged from a maximum 105ºC and a minimum of 100ºC. The dissolution time ranged from 14 to 23 minutes, however, it was 15 minutes at temperatures from 80 to 88°C. In this methodology a temperature rise much slower during the dissolution was observed, however, in all experiments the profile tends to a plateau around 100°C.

It was observed that the dissolution process is strongly exothermic and the dissolution rate increases as temperature increases, confirmed by the results obtained with the third method (Fig. 9). According to established studies the dissolution process must be controlled so that the dissolution is complete within about 1 hour under conditions such that the temperature remains around 90°C [4].

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