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TEMPERATURE PROFILE DURING THE ALKALINE DISSOLUTION OF Al FOR THE PRODUCTION OF Mo-99

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ABSTRACT

Since September 2008 Brazil is developing the project called Brazilian Multipurpose Reactor (RMB), having as main objective to produce about 1000 Ci/week of ^{99}Mo. 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. This study is part of the project to obtain ^{99}Mo by alkaline dissolution of UAl_x-Al targets. Al corresponds to about 79% of the total weight of the UAl_x-Al target. The initial reaction temperature is an important parameter, since it has great influence on the value of the maximum temperature and dissolution time. The heat of reaction must be removed from the dissolving system by controlled cooling. According to literature as a safety condition, the dissolution process must have its temperature controlled so that the maximum temperature to be around 90°C. The behavior of the temperature during dissolution using a thermostatic bath with continuous circulation to maintain its value at around 90°C was studied. The alkaline solution of NaOH 3 mol.L^{-1} and $NaNO_3$ 2 mol.L^{-1}. As initial temperatures were: 70, 75 and 80°C and initial temperatures of the thermostatic bath were: 40, 45, 50, 60 and 70°C. The results indicate that none of the studies it was possible to maintain the temperature of dissolution near 90°C. In the studies where the maximum temperature was around 93°C dissolution was incomplete. It may be necessary an intermittent cooling rather than continuous to ensure that the temperature profile is maintained around 90°C.

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 [1,2].

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 survey cover targets with low enrichment (<20% of 235U). The studies will be with UAlx-Al targets via alkaline dissolution.

The Nuclear Fuel Center (NFC) of Nuclear and Energy Research Institute (IPEN-CNEN/SP) mastered the technology of manufacturing UAlx-Al miniplates, for this reason the studies 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 1050 used for manufacturing the targets. Al is about 79% of the total mass of the UAl_x-Al miniplates [3].

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 [4].

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 [5], 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 $NaNO_3$ in the reagent [6].

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

To simulate UAlx-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.

To verify the temperature profile during the dissolution, the study of the temperature as a function of the initial temperature of the reagent and the temperature of the water bath was performed because according to the literature the temperatures should be around 90°C. This study was carried out using the following procedure: 1) initiating the cooling process with the continuous dissolution in water bath by setting the initial temperature (40, 45, 50, 60 and 70°C); 2) external heating of the dissolution solution until reaches the initial temperature chosen (70, 75 and 80°C); 3) introducing the Al sample to be dissolved into the solution. In this study NaOH 3mol.L^{-1} and $NaNO_3$ 2 mol.L^{-1} solution as dissolution reagent was used.

4. EXPERIMENTAL

4.1. Dissolution studies with initial temperature of reagents at 70ºC and water bath at 40, 45, 50, 60 and 70°C.

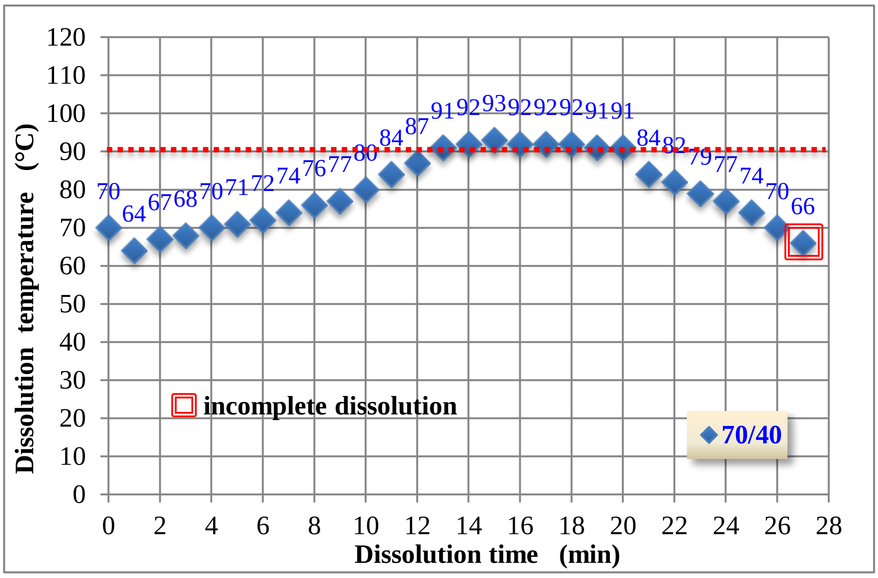

Figure 1: Temperature profiles during the dissolution with initial temperature of 70ºC and water bath initially at 40ºC.

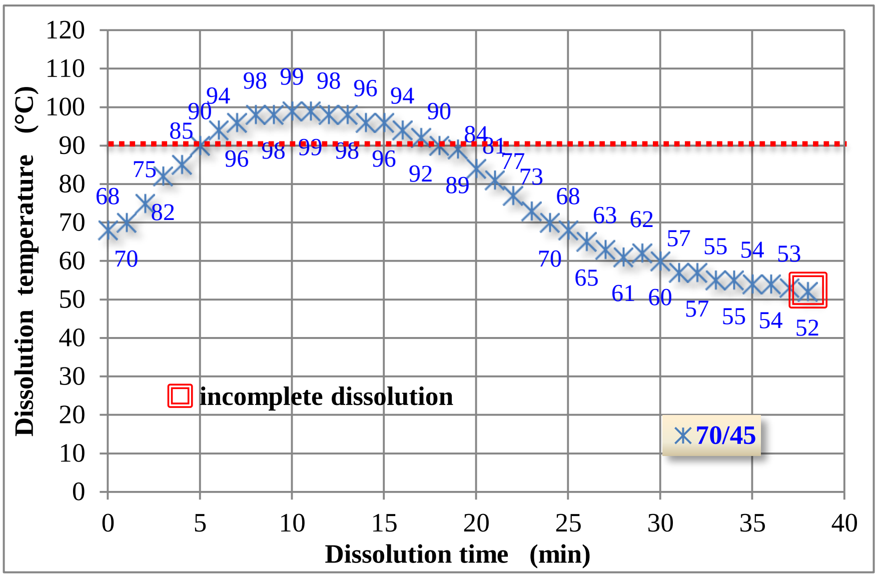

Figure 2: Temperature profiles during the dissolution with initial temperature of 70°C and water bath initially at 45°C.

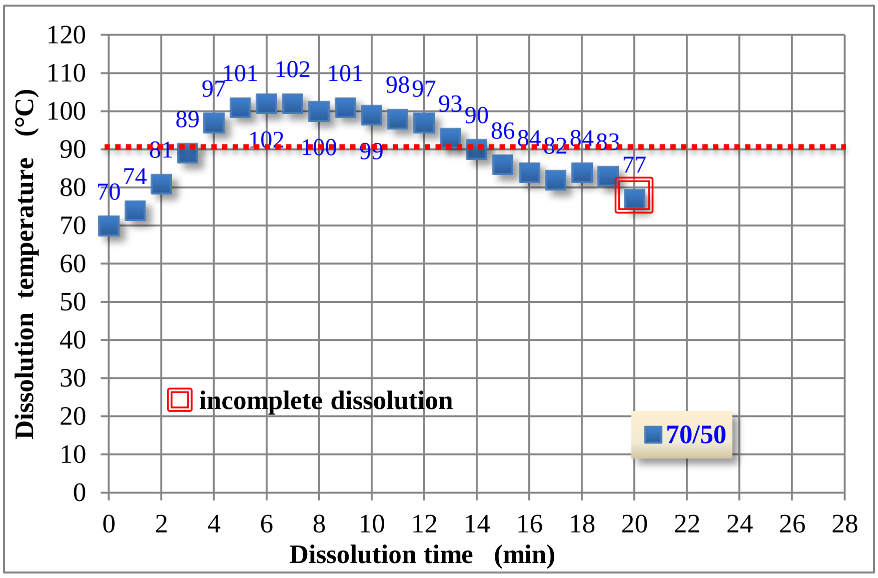

Figure 3: Temperature profiles during the dissolution with initial temperature of 70°C and water bath initially at 50°C.

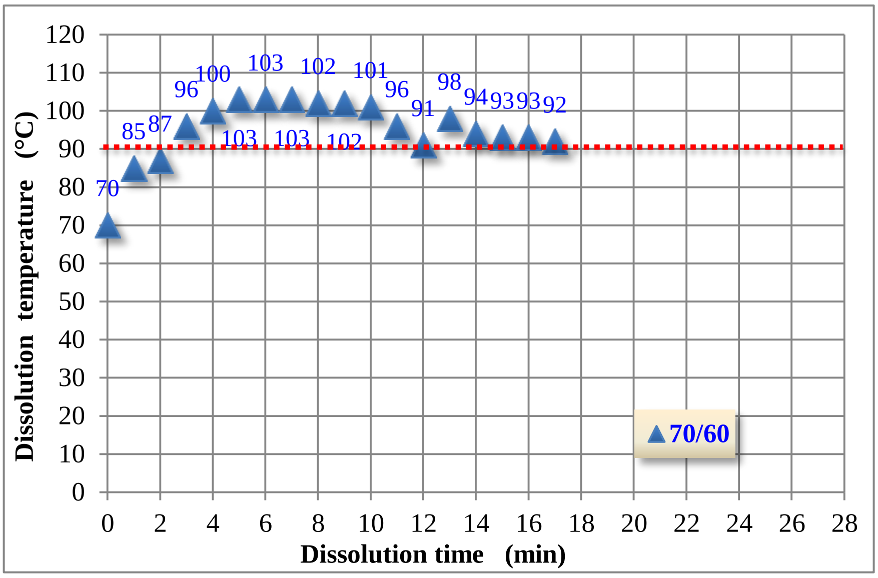

Figure 4: Temperature profiles during the dissolution with initial temperature of 70°C and water bath initially at 60°C.

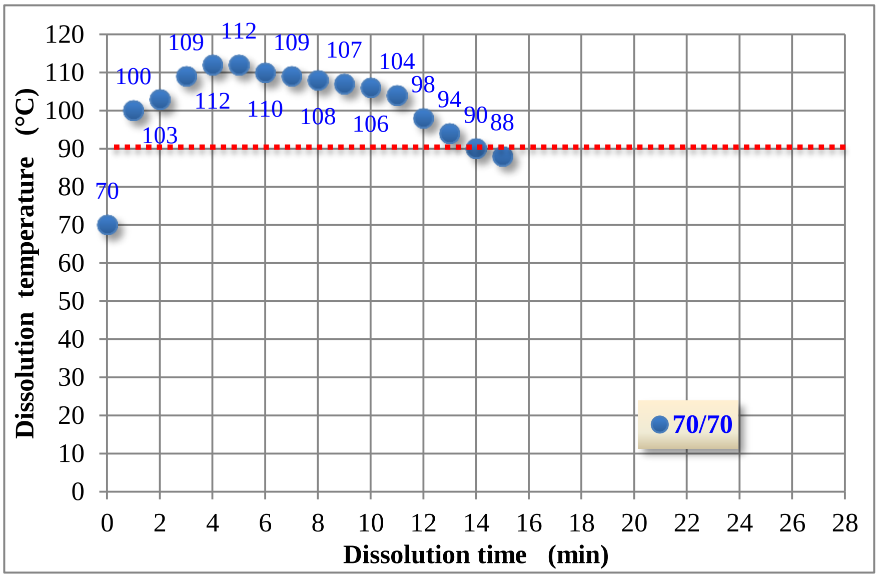

Figure 5: Temperature profiles during the dissolution with initial temperature of 70°C and water bath initially at 70°C.

4.2. Dissolution studies with initial temperature of reagents at 75°C and water bath at 40, 45, 50, 60 and 70°C.

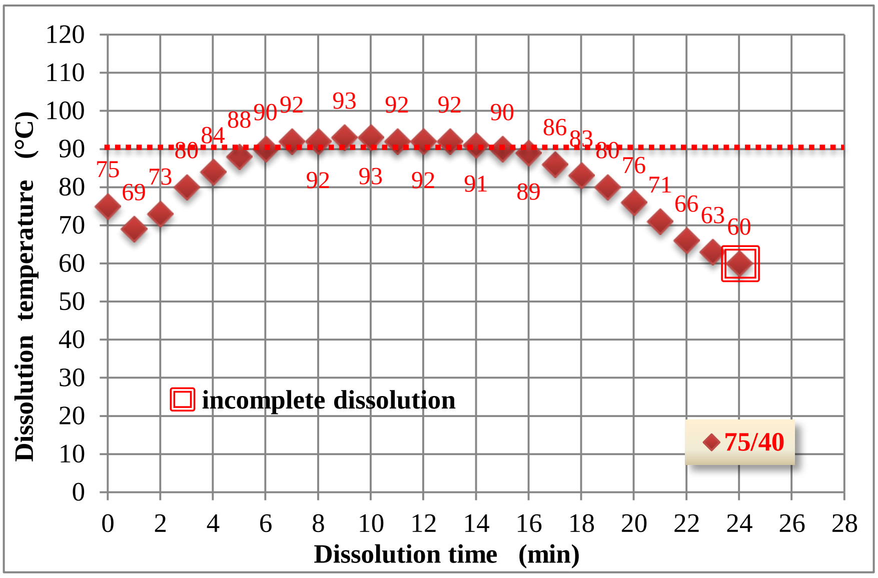

Figure 6: Temperature profiles during the dissolution with initial temperature of 75°C and water bath initially at 40°C.

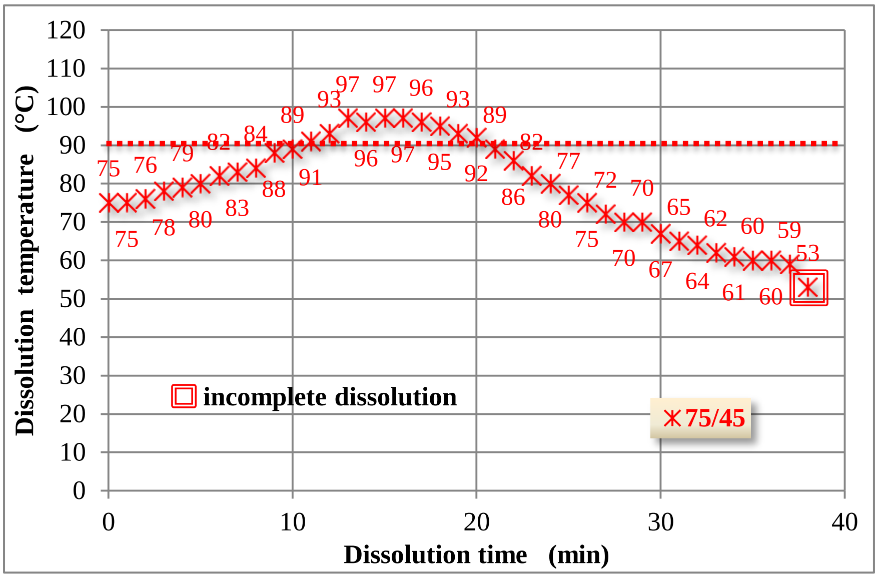

Figure 7: Temperature profiles during the dissolution with initial temperature of 75°C and water bath initially at 45°C.

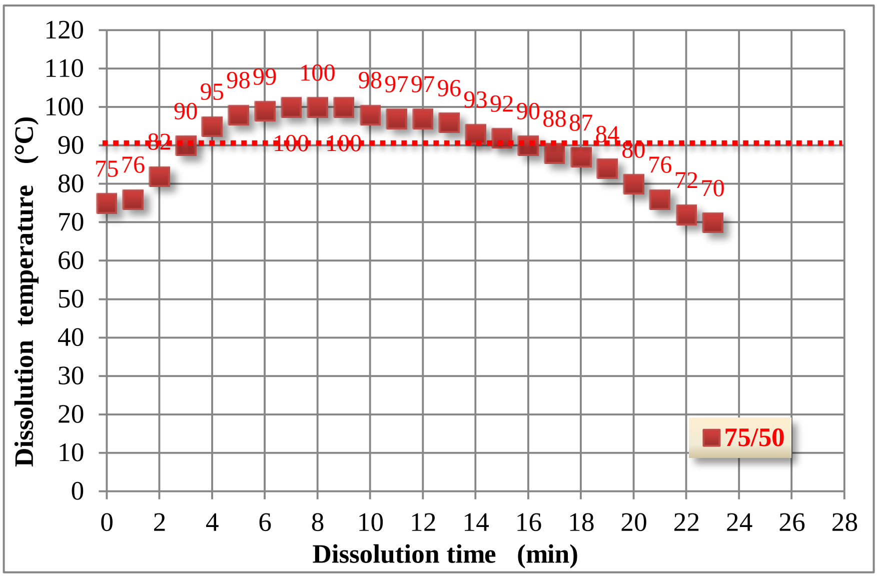

Figure 8: Temperature profiles during the dissolution with initial temperature of 75°C and water bath initially at 50°C.

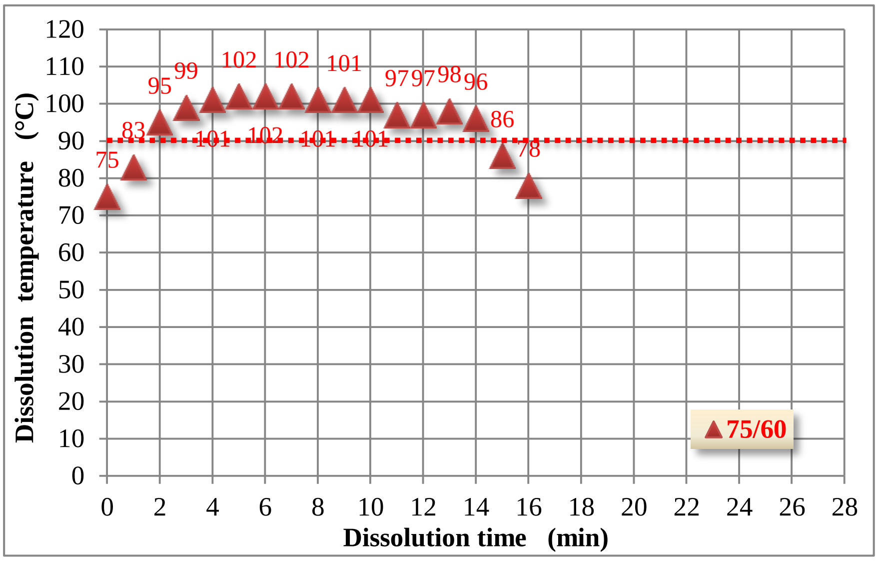

Figure 9: Temperature profiles during the dissolution with initial temperature of 75°C and water bath initially at 60°C.

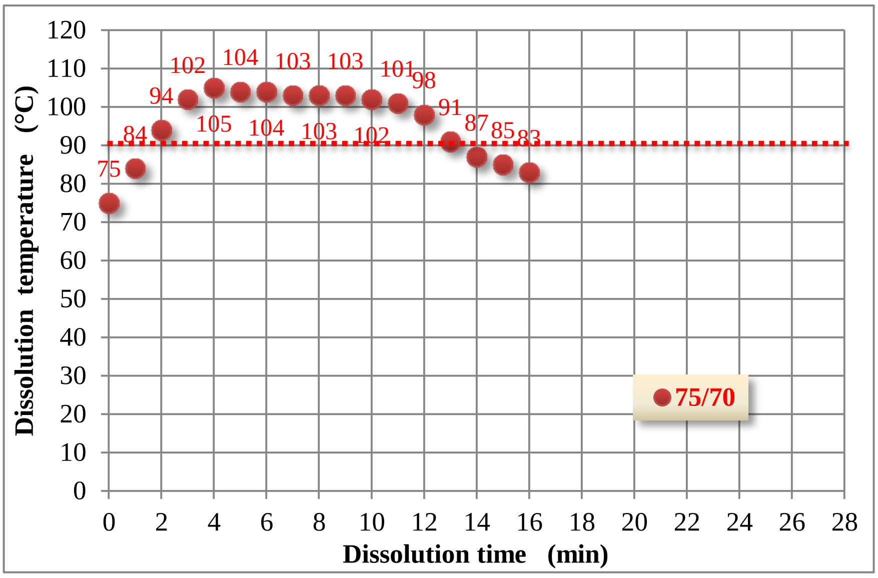

Figure 10: Profiles during the dissolution with initial temperature of 75°C and water bath initially at 70°C.

4.3. Dissolution studies with initial temperature of reagents at 80°C and water bath at 40, 50 and 60°C.

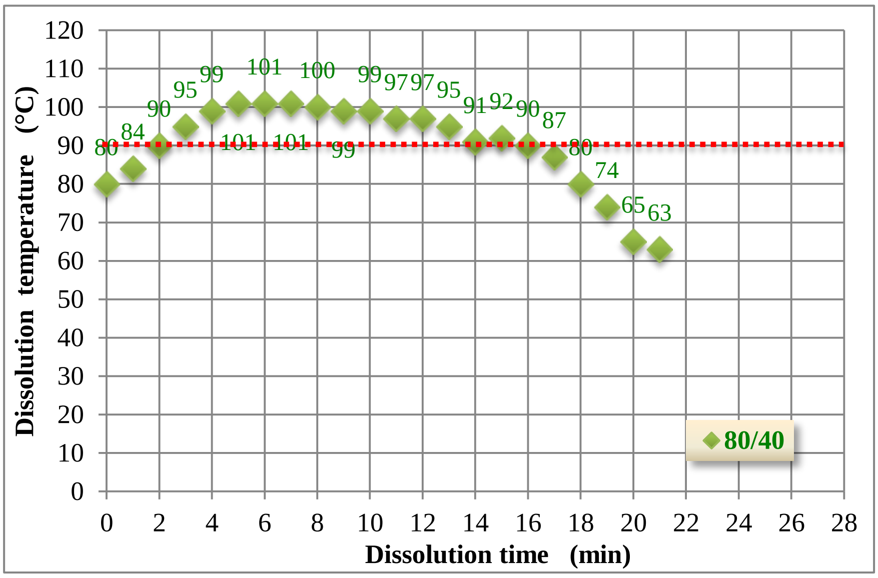

Figure 11: Temperature profiles during the dissolution with initial temperature of 80°C and water bath initially at 40°C.

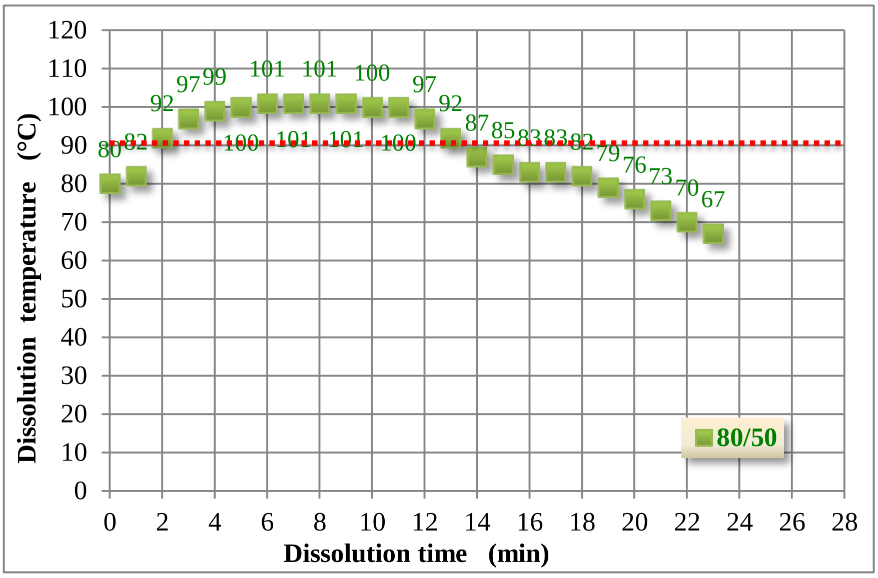

Figure 12: Temperature profiles during the dissolution with initial temperature of 80°C and water bath initially at 50°C.

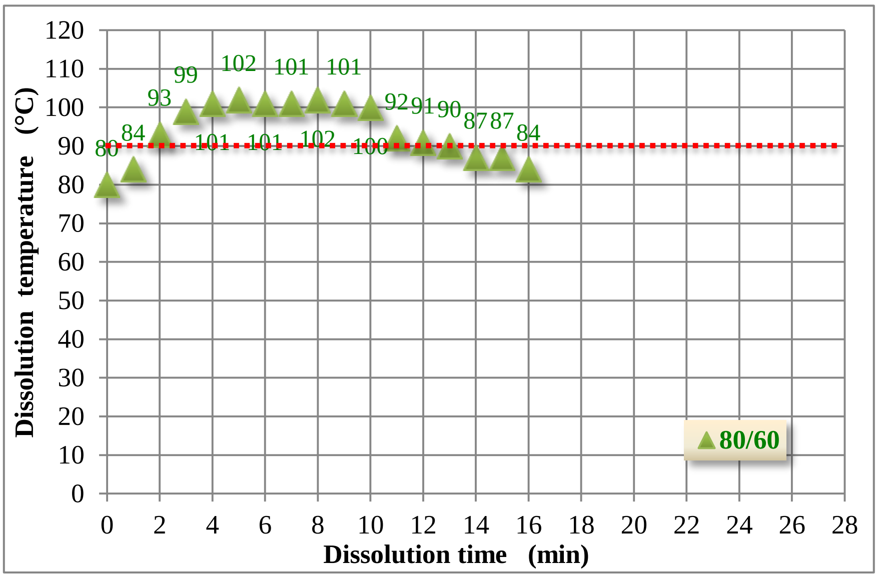

Figure 13: Temperature profiles during the dissolution with initial temperature of 80°C and water bath initially at 60°C.

Table I: Maximum (T_{max}) and final (T_{end}) temperatures in the studies with initial dissolution temperatures of 70, 75 and 80°C and water bath of 40, 45, 50, 60 and 70°C.

T_{diss}/T_{bath}	T_{max}(ºC)	T_{end}(ºC)
70/40	93	66
70/45	99	52
70/50	102	77
70/60	103	92
70/70	112	88
75/40	93	60
75/45	97	53
75/50	100	70
75/60	102	78
75/70	104	83
80/40	101	63
80/50	101	67
80/60	102	84

5. CONCLUSIONS

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

In all graphs the temperature of the cooling bath has influenced on the time of dissolution. The temperature rise of cooling helps reduce dissolution time.

In Table I it is noted that in situations where the maximum temperatures studied were close to 90°C, the dissolution was incomplete. In these studies the cooling of the dissolution solution was continuous.

It is believed that as the process is exothermic; to maintain the temperature profile during the dissolution around 90°C an intermittent cooling must be used.

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