

# MANUFACTURING LOW ENRICHED URANIUM METAL BY MAGNESIOTHERMIC REDUCTION OF UF<sub>4</sub>

M. DURAZZO, A. M. SALIBA-SILVA, I. C. MARTINS, E. F. URANO DE CARVALHO

*Nuclear and Energy Research Institute – IPEN/CNEN-SP, São Paulo – Brazil*

H. G. RIELLA

*Chemical Engineering Department, Santa Catarina Federal University, Florianópolis, Brazil*

## ABSTRACT

This work presents an experimental description of thermal and physical studies to attain a practical manufacturing process of uranium metal enriched to 20% U<sup>235</sup> (LEU–Low Enriched Uranium) by metallothermic reduction of UF<sub>4</sub>, with nuclear purity, for reduced amounts (1000g of uranium). The magnesiothermic reduction is influenced by the thermal conditions for UF<sub>4</sub> reduction. These variables are investigated. The physical arrangement of the crucible/reduction reactor/furnace system and the management of the furnace thermal input in the reduction reactor during the heating were studied. Thermal simulation experiments provided delineation for the reactants' thermal progress before the ignition of the reaction. The heat input to the reduction system has proved to be the main variable that influenced the efficiency of the process. The levels of metallic yield and reproducibility have been improved. The typical yield in the production of uranium metal was above 80%.

## 1. Introduction

The production of uranium metal is necessary to produce the intermetallic U<sub>3</sub>Si<sub>2</sub> which is the modern basis for manufacturing the nuclear fuel used in nuclear research reactors [1,2,3,4]. Uranium metal is also necessary for manufacturing irradiation targets to produce <sup>99</sup>Mo by nuclear fission [5]. These targets are based on the UAl<sub>x</sub>-Al dispersion or thin foils of uranium metal [6,7,8].

There are several possibilities for producing uranium metal [9,10]. In the decade of 1930-40, uranium was produced from metallothermia with Na and Ca from UCl<sub>4</sub>. In the 1950s, in the context of worldwide interest in nuclear technology, several technologies were developed to obtain uranium metal: by electrolysis from the KUF<sub>5</sub> salt, by UO<sub>2</sub> metallothermia with Ca and Mg, and by metallothermia of UF<sub>4</sub> with Ca and Mg. Magnesiothermic reduction of UF<sub>4</sub> is a known process since the early 1940's [11,12].

Normally, calciothermic reduction of UF<sub>4</sub> is preferred worldwide since the exothermic heat is -109.7 kcal/mol if compared to a smaller amount of -49.85 kcal/mol produced using magnesium as the reducer [13]. Nevertheless, the Nuclear and Energy Research Institute – IPEN/CNEN-SP in the nationalization process to produce its own nuclear fuel for research reactors and target fabrication [14,15], decided for the magnesiothermic process of uranium tetrafluoride (UF<sub>4</sub>) since it is easier to be done avoiding handling of toxic and pyrophoric calcium. Moreover, the magnesiothermic process is cheaper since magnesium is cheaper than metallic calcium. Thus, there is an economic compensation despite the unfavorable thermochemistry of reduction with magnesium when compared with reduction with calcium.

Magnesiothermic reduction employs metallic magnesium as a chemical reducer of uranium. Magnesium is mixed with stoichiometric excess to uranium tetrafluoride (UF<sub>4</sub>), which is produced from UF<sub>6</sub> hydrolyzed in HF solution. The hydrolyzed solution is then reduced by stannous chloride to UF<sub>4</sub> [16,17]. UF<sub>4</sub> is a greenish substance which, mixed with magnesium, can be reduced to uranium metal under adequate thermal conditions. This reaction is intensely exothermic. The reaction is:



The reaction products utilize the resulting exothermic heat and melt to form the uranium ingot at the bottom of the crucible and the slag. The supernatant slag, which is formed essentially by MgF<sub>2</sub>, solidifies at the top of the ingot. Considering the calculations of Rand and Kubaschewski [13], the sum of the heat produced by this reaction and the latent heat to melt the products give a reaction heat of -49.85 kcal/mol. Therefore, this is the resulting heat available to heat the products. Assuming that 49 cal/mol is required to raise one degree centigrade of the reaction products, it has been shown that the possible increase in temperature would be only 1017°C. This average heating is based on water, as a calorimetric reference, which uses 7 cal/°C for each atomogram. In the case of 2MgF<sub>2</sub>+U, there are 7 atoms present which, therefore, need 49 cal/°C [13]. However, this heat is not sufficient to reach the melting reaction products if the reaction occurred at room temperature (U melts at 1132°C and MgF<sub>2</sub> at 1255°C). Therefore, these fusions are only possible if there is a preheating of the reactants before the reaction, as predicted by Harper and Williams [9] for this process. In this way, it is seen that the reduction system must be properly designed in order to maximize the thermal efficiency of the system.

IPEN/CNEN-SP decided to use the magniothermic route in 1970-80's for producing 100 kg ingots of natural uranium. However, for low enriched (LEU) U-production, it is necessary to handle safe mass (less than 2.2 kg U) to avoid possible criticality hazards. We started studying the magnesiothermic reduction in the late 1990's to produce small amounts of enriched uranium (around 1000 g LEU ingots) [18,19]. This range of LEU U weight is rather small if compared to big productions of natural uranium. Uranium metal is reported [20] to be produced with 94% metallic yield when producing bigger amounts of raw material. The magnesiothermic process downscaling to produce LEU has small possibilities of achieving this higher metallic yield. This is due to the design of crucibles, with a relatively high proportion of surrounding area, which is more prone to withdrawing evolved heat from the exothermic reaction during uranium reduction. The reduction reaction initiates by means of spontaneous ignition. As magnesium thermodynamics is less prompt to ignite than calcium, the batch reactor must be heated up to a temperature around 620°C. The routine shows that this ignition normally happens some degrees below this temperature [20].

IPEN/CNEN-SP designed an infrastructure to produce small amounts of LEU metal. However, in the initial reduction tests, the process did not prove to be reproducible since the yield in produced uranium metal varied over a wide range, from 20% or less to 90% or even more.

The objective of the present work was to investigate the possible causes that affected the yield of the process in order to achieve a stable and reproductive process for the production of LEU metal with the best possible yield. Some variables related to UF<sub>4</sub> powder were investigated. A physical adaptation of the furnace and reactor set was performed and thermal parameters were developed in order to establish a routine for the production of LEU metal.

## 2. Experimental Procedures

A standard reactant charge was fixed at 1815 ± 5g of the UF<sub>4</sub>+Mg mixture (1540 ± 1g UF<sub>4</sub>), containing 15% excess Mg in addition to the stoichiometric amount. In this charge, there is a theoretical mass of 1154g of uranium metal in the form of UF<sub>4</sub>. The objective was to obtain around 1000g of LEU metal.

The preparation of the charge was done keeping a homogeneous mixture of  $UF_4$  with Mg. As these materials have a great difference in densities ( $\rho_{UF_4} = 6.72 \text{ g/cm}^3$  and  $\rho_{Mg} = 1.74 \text{ g/cm}^3$ ), a stratified charge was made. For homogenization purpose, the charge of  $UF_4+Mg$  was divided into 10 layers tapped one by one inside a graphite crucible, each layer containing the blend in the desired proportion. This operation was performed inside a glove box using a V type homogenizer mixer.

The height of the charge inside the graphite crucible varied mainly due to the variability of the tapping operation of the charge layers, which was performed manually. To fill the empty space between the top of the charge and the graphite crucible cap, a variable amount of  $CaF_2$  was tapped over the  $UF_4+Mg$  charge in the crucible to fully complete the reaction volume.  $CaF_2$  was used for being readily available.

The charge bulk density was calculated based on the volume of the crucible filled with the charge. The occupied volume was calculated from the measured value of the distance from the top of the charge to the edge of the graphite crucible, which has a known height. This measurement was carried out using a ruler inside the glove box. Due to the difficulty in reading the measured value, the error in the reading was considered to be 2 mm. The maximum associated error in the calculated charge bulk density was considered to be around  $0.05 \text{ g/cm}^3$ . The magnesium used was in the form of granules of size -12+50 mesh (0.3-1.7 mm). The purity was 99.8%.

The crucible was made of fully machined graphite volume with enough resistance to produce safe nuclear uranium amounting to around 1000g. This crucible was designed to withstand the blast impact of metallothermic reaction, as well as thermal cycles of heating and cooling without excessive wear in order to be used in several batches. The graphite crucible was not lined.

After closing the graphite top cover, the crucible was inserted inside an ANSI 310 stainless steel cylindrical reactor vessel, which allows argon fluxing during batch processing (1 L/min with  $2 \text{ kgf/cm}^2$  of pressure). As shown in Figure 1, the whole crucible + reactor was placed in an electric-powered pit furnace with four programmable zones having the possibility of raising the temperature up to  $1200^\circ\text{C}$ . The reaction vessel was heated and the reactants ignited spontaneously. The reaction of  $UF_4$  with Mg produces an intense exothermic heat release inside the crucible. It is considered as an adiabatic reaction. It produces uranium metal and  $MgF_2$  slag in liquid form. Both products deposit in the crucible bottom and are easily taken apart after opening and discharging the crucible. The exact moment of spontaneous ignition and reaction time was detected and recorded by sound waves, using an accelerometer.



Figure 1. Charging of the reactor vessel inside the pit furnace

The whole system developed for the magnesiothermic reduction of  $UF_4$  is shown in Figure 2. This figure shows the graphite crucible set and the reduction reactor inserted in the furnace inner for heating. The dimensions shown in Figure 2 correspond to the current system. The reduction reactor consisted of a cylindrical vessel with pipes necessary for the introduction of argon and the exit of gases inside the reactor. The reactor head is cooled with airflow. This vessel can withstand positive pressure up to 4 atm.

After the reaction, there was a 10-minute wait for full solidification of the reaction products inside the furnace. Then, the furnace energy was turned off and the reactor vessel was lifted out of the furnace. There was another wait for a period of 16 hours to provide complete cooling before its opening for discharging. This avoids uranium metal pyrophoricity, which may happen when this metal is exposed to atmospheric oxygen.

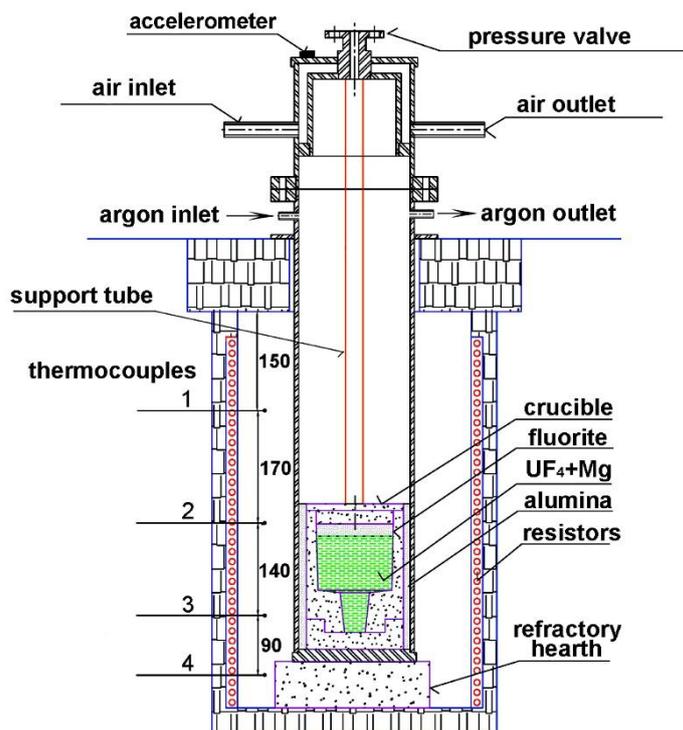


Figure 2. Schematic drawing of pit furnace, reactor vessel and crucible

The unassembly of reduction set was performed inside a glove box inertized with argon. The top and bottom crucible covers were then removed. By means of rubber soft hammering, it was able to withdraw the uranium ingot. The  $MgF_2$  slag was removed by mechanical cleaning. The uranium metal was pickled in nitric acid 65%vol and the final mass of uranium metal was measured and its density evaluated by Archimedes' method.

Following these procedures, the efficiency on uranium metal fabrication was tested by processing 20 precipitation batches of  $UF_4$  made on a large scale (50L) and employing optimized  $UO_2F_2 + HF + SnCl_2$  solution at a temperature range of  $92 \pm 3^\circ C$  with resting time for precipitation during 24h [16,17]. About 3000g of natural  $UF_4$  was produced for each batch. The magnesiothermic process was used to reduce this material to uranium metal by using 40 magnesiothermic reduction operations. After each reduction, the metallic yield was calculated as the ratio between the mass of uranium metal produced after reduction and the total uranium contained in the  $UF_4$  prior to this reduction.

### 3. Results and Discussion

The bulk density of the charge varied in the range of 1.70 to 2.05  $g/cm^3$ . The height of the  $CaF_2$  layer varied between 12 and 32 mm. Figure 3 shows the influence of the charge bulk density on the metallic yield of the reduction to uranium metal. All individual data are shown. The results show that there is no apparent correlation between the charge bulk density and

the yield of the reduction process. Although the thermal conductivity of the charge is affected by the density variation, the high yield variation cannot be attributed to the variations in the charge bulk density.

Figure show the great variability observed in the metallic yield, which cannot be explained by the charge bulk density variations. Considering the great variability observed in the metallic yields, it cannot be affirmed that there is a correlation between the metallic yield and the charge bulk density. The variation of the charge bulk density also reflects the variation of other physical characteristics of  $UF_4$  that could affect the yield of the reaction, such as particle size and reactivity (specific surface area). Therefore, these physical characteristics do not appear to play an important role in the observed variability in metallic yields. Another powerful variable must be acting.

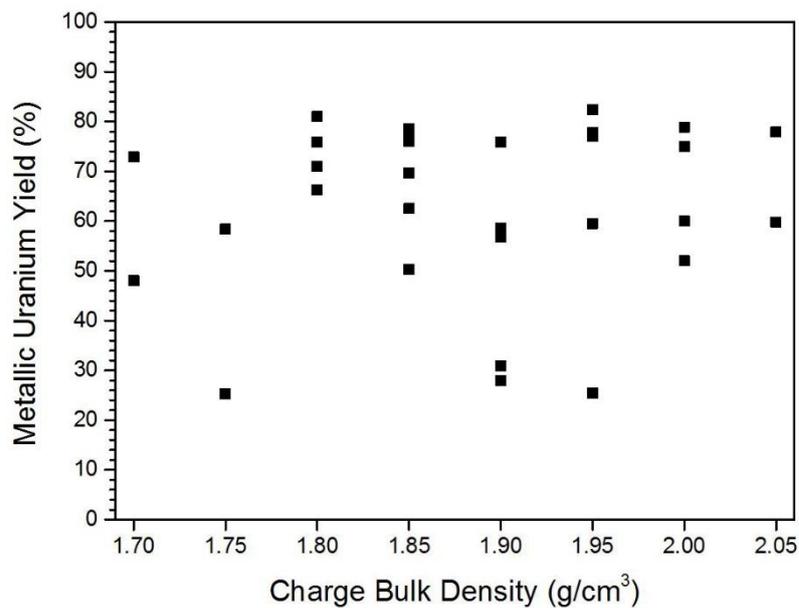


Figure 3. Influence of charge bulk density on metallic yield

Figure 4 shows the variation of the metallic yield obtained in the first 30 reductions performed with natural uranium. A great variability was observed, from 20 to 86%.

Seeking to understand the big variations observed in the metallic yield in the first magnesiothermic reductions, the slags formed after each reaction were fully inspected, trying to correlate any distinctive attribute or aspect with the reaction yield. The magnesiothermic operational routine and evidence after dismounting the crucible to remove uranium ingot lead to the reasoning about the moments before, during and after the ignition. The results of careful inspections of the crucible after reduction showed that the distribution of the  $MgF_2$  slag inside the crucible had a direct relationship with the metallic yield. Three situations could clearly be observed:

- When the yield was good (above 80%), the slag was concentrated at the bottom of the crucible, over the uranium metal ingot. The presence of unreacted  $UF_4$  and uranium metal in the slag was not observed visually.
- In the case of intermediate yields, between 80 and 70%, a considerable volume of adhered slag in the sidewalls of the crucible was observed. A smaller amount of slag was observed over the uranium metal ingot. Droplets of solidified uranium metal were observed entrapped within the adhered slag to the side of the crucible.
- In the case of low yields, below 70%, most of the slag was found on the walls and cover top of the crucible. Large quantities of uranium metal droplets could be seen entrapped within the slag.

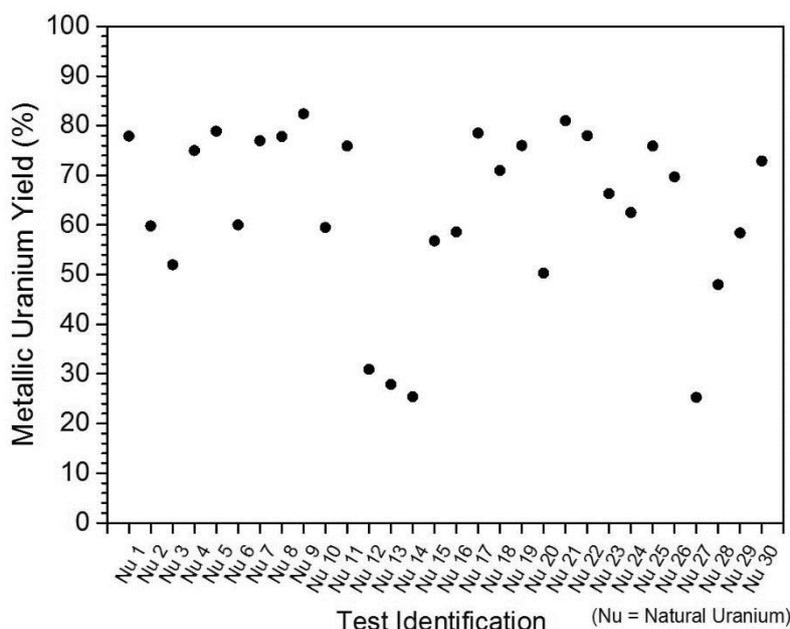


Figure 4. Initial variability of yield in the uranium metal production by magnesiothermic reduction of  $UF_4$

These observations suggested that the most important factor that determines the yield of the reaction is the particular location where the ignition occurs within the charge. If the ignition or the blast onset occurs at the top of the charge, the front of the exothermic reaction shifts from above downwards, providing heat to efficiently melt the reaction products and to promote an effective separation between uranium metal and  $MgF_2$ . The heavy uranium metal goes towards the crucible bottom, and the less dense slag solidifies above it during cooling. The efficient melting of the slag allows the uranium metal droplets, which are produced during the reaction, to separate from the slag duly and also going towards the crucible bottom. The efficiency of uranium metal separation from the slag defines the metallic yield of the reduction process.

When ignition occurs at the bottom of the crucible (or even at the center of the charge), the heat front of the exothermic reaction moves in opposition to the movement of the molten uranium metal produced. The blast throws the  $MgF_2$  slag into the top cover and towards the side walls of the crucible, which solidifies at those places and partially entraps the uranium metal produced in the reaction (see Figure 5). The yield greatly decreases in these cases because, although the reduction occurs, the produced uranium metal is not efficiently separated from the slag.

In order to collect data of the heating profile inside a  $UF_4$  charge, with no Mg reducer, thermocouples were inserted inside the graphite crucible. The thermocouples were duly placed inside de graphite crucible to collect the heating profile at six points; at the top, middle and bottom positions of the charge. The side thermocouples (4, 5 and 6) were at a distance of about 5 mm from the crucible wall. This assembling was schematically made as shown in Figure 5. The furnace has four independent heating zones with four thermocouples (F1tc to F4tc). Inoxidable steel protected k-type thermocouples were used (numbered 1 to 6) to record the temperatures inside the graphite crucible.

A preliminary experiment using the charge of  $UF_4+Mg$  allowed us to get the moment of charge ignition starting the reaction to produce uranium. This was a sacrificial test for the thermocouple. The thermocouple was placed at the top center position of the charge (position 3). The temperature of ignition was found to be around  $620^{\circ}C$ .

A second experiment was carried out using only  $UF_4$ , without Mg, and using six thermocouples placed accordingly inside the reduction reactor (Figure 5). The heating of the reduction reactor started inside a cold furnace. The data for temperature profile lead to the graph presented in Figure 6. All the four heating zones of the furnace were powered up to

step up until 550°C at a heating rate of 5 °C/min. The charge was left to soak for 60 min at this temperature and then the four heating zones of the furnace were powered up to reach 1000°C at a rate of 5 °C/min. This profile is called “wild” heating procedure, since no fine control of heating setups and heat transfer was made.

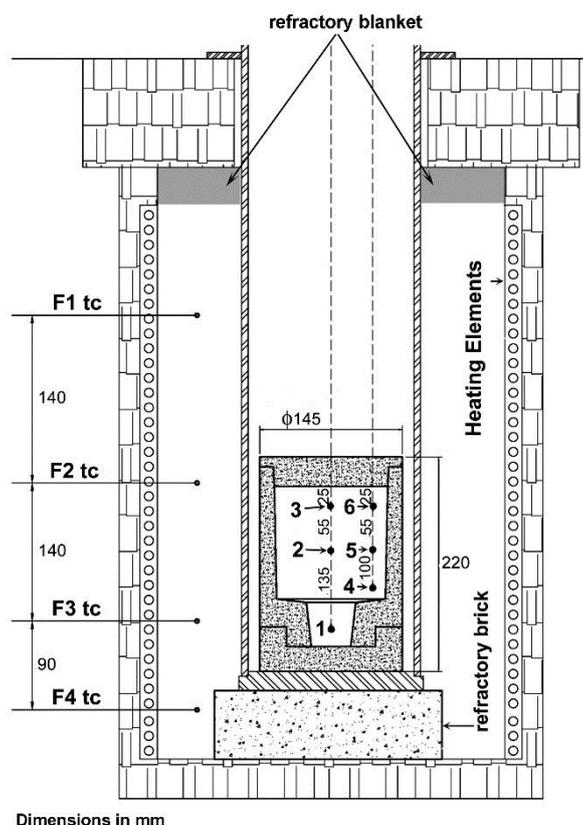


Figure 5. Schematic assembly of the reduction reactor/crucible system inserted in the well

The results display crude differences between temperature evolution inside several regions of the bomb. The major one between top and bottom was 170°C. The time to ignition, as recorded by the accelerometer, also varied over a wide range, from 220 to 330 minutes. Naturally, this is the profile that we would get without the possibility of checking the thermal profile evolution. This is the kind of result we had in the previous history of wide metallic yield variation in the uranium reduction procedures we had carried out, trying to do the reductions with no knowledge of the thermal profile. Figure 6 shows the temperature profiles inside the “pseudo” charge of pure UF<sub>4</sub> following the “wild” heating procedure.

In practice, the reaction products should be heated to a temperature slightly above 600°C for ignition of the reaction. From the data calculated by Rand and Kubaschewski [13] only a preheating of 500°C would be necessary, but in practice this value was around 570-620°C, which indirectly confers a greater fluidity on the reaction products. This has been confirmed in practice by the preliminary experiment using the charge of UF<sub>4</sub>+Mg to determine the temperature of ignition.

Assuming that the ignition occurs at 620°C, as determined empirically, by examining the temperature profiles shown in Figure 6, it is found that ignition occurs at the bottom of the charge. The highest temperatures are indicated by thermocouples 1 and 4, both at the bottom of the graphite crucible. This is an unfavorable situation and should be avoided, because when ignition occurs at the bottom of the crucible the heat from the exothermic reaction moves in opposition to the movement of the produced molten uranium metal. As already mentioned, the blast throws the MgF<sub>2</sub> slag into the top cover and sides of the crucible, which solidifies in these places and entraps the produced uranium metal. The yield declines in this case due to poor separation. The largest variations that were observed in the

yields recorded in Figure 6 should be related to the position in the charge where the ignition occurs. The more the ignition position moves to the top of the charge, the higher the metallic yields will be. If the ignition occurs in the regions at the bottom of the crucible, the yields will be lower.

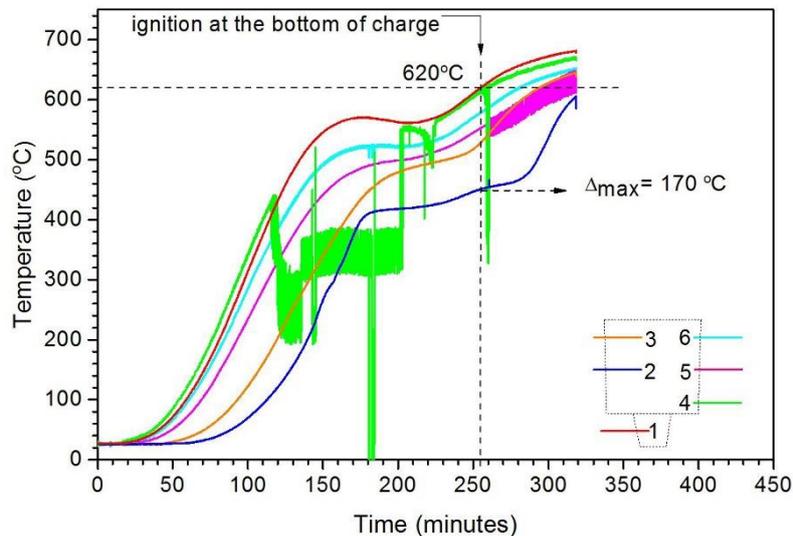


Figure 6. Thermal profiles derived from the “wild” heating procedure

Another unfavorable condition is related to the large temperature variation inside the charge at the moment of ignition, as shown in Figure 6. The temperature difference reaches 170°C between thermocouples 1 (bottom of the crucible) and 2 (center of charge). At the moment of ignition, the central region of the charge is around 450°C, which is below the minimum preheating temperature calculated by Rand and Kubaschewski [13].

The long time in which the charge remains at high temperatures before ignition should also have a negative impact on the efficiency of the reduction process. In Figure 6 it can be observed that the charge remains up to 150 minutes at temperatures above 400°C, which favors the occurrence of undesirable reactions that may cause the decrease in the metallic yield.

Based on these observations and discussion, it was clear that a new heating program should be developed. The new program must ensure a favorable temperature evolution within the charge, in the following aspects:

- a) the top of the charge should reach the ignition temperature (around 620°C) before the bottom of the charge or even the central region. This ensures that the ignition will occur at the top of the charge, which favors the separation of the uranium metal by the reaction front displacement from top to bottom;
- b) keeping the top of the charge always at a higher temperature at the moment of ignition, the maximum temperature variation between the charge regions should be minimized so that no region is below 500°C, as proposed by Rand and Kubaschewski [13].
- c) the ignition should occur as soon as possible, avoiding exposure of the charge to high temperatures soaking for a long time.

Several heating programs were unsuccessfully tested. All simulations using  $UF_4$  as charge (without magnesium) resulted in faster heating at the bottom of the charge. This was caused by the geometry of the system. The solution of the problem would depend on delaying the bottom heating of the graphite crucible and accelerating the top heating. This was accomplished by inserting a refractory brick (see Figure 5) below the reduction vessel and inserting a refractory blanket into the open space between the reduction vessel and the wall of the furnace above the heating zone. With these small modifications, it was possible to

develop a new heating program that favored the separation of the uranium ingot after the reduction reaction.

The new heating program consisted of raising all four furnace heating zones to 640°C with the highest heating rate allowed by the furnace system, around 10°C/min (varied from zone to zone). All heating zones were left for 240 min at this temperature and then the furnace was powered up to reach 1000°C at a rate of 5 °C/min. The buck mass of the refractory brick placed below the reduction vessel slowed down the bottom heating of the graphite crucible. The better thermal insulation at the top of the reduction vessel accelerated the heating of the graphite crucible top. Figure 7 shows the new heating profiles after the implementation of these modifications.

The thermal profiles achieved after these modifications were satisfactory since all the parameters seeking better heating were met.

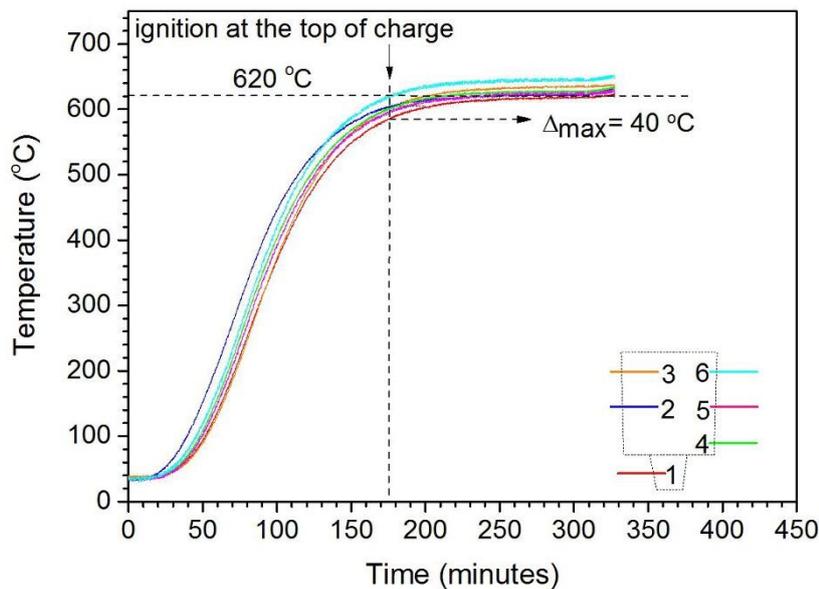


Figure 7. New thermal profiles after adjusting the geometry of the system and heating procedure

The border top of the reduction charge in the crucible was the first position to achieve 620°C (thermocouple 6). This is the likely place of the spontaneous ignition. The charge positions where the temperature was just below 620°C were the center top and center of the charge in the central region (thermocouples 3 and 2), which was also a favorable situation for the ignition path. The bottom of the charge (thermocouple 1) was the position that had the lowest temperature (580°C) at the ignition flash. This means that it has the lowest probability to ignite the reaction at this location, which would be most unfavorable.

From the simulation, the maximum temperature difference between all regions of the reduction charge was only 40°C, ensuring that all regions were above 500°C at the time of ignition, which meets the minimum preheating suggested by Rand and Kubaschewski [13].

The heating time until the ignition decreased significantly with the new heating program was from 255 to 175 minutes. The time at which the charge remained above 400°C decreased from 150 to 85 minutes.

Based on these positive results, a further 10 reduction tests with natural uranium were carried out using this new heating program and the new system conditions. The results showed that the times elapsed until the ignition were reproducible. The ignition time was determined experimentally with the use of an accelerometer. The reproducibility of the ignition time demonstrates that the heating profiles shown in Figure 7 have reproduced in the

actual reductions. As a result, the ignition occurred always at the top of the reduction charge and the metal yields stabilized above 80%.

Once it was demonstrated that the modifications described above stabilized the metallic yield above 80%, reduction programs were started using LEU  $UF_4$ . Fifty nine reduction operations were carried out so far, which confirmed good metallic yields. Figure 8 shows the yields obtained in the production of enriched uranium metal and summarizes all the results obtained throughout the development of the process to produce uranium metal by magnesiothermic reduction of  $UF_4$ .

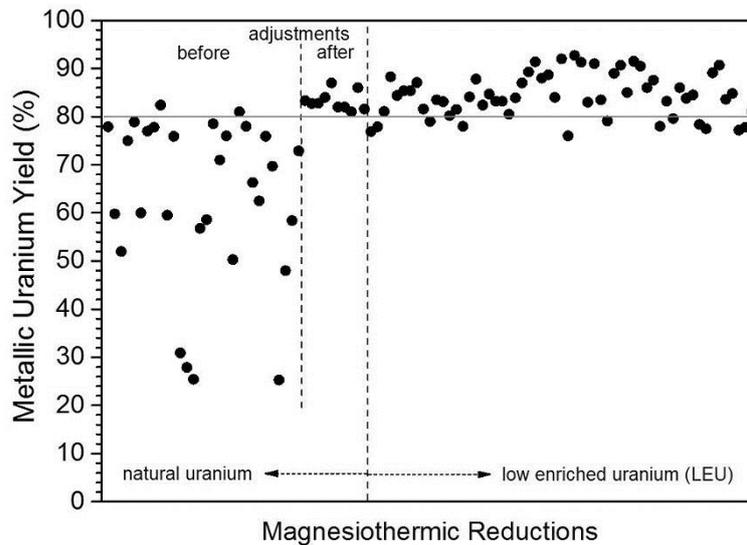


Figure 8. Metallic yields from operations with low enriched uranium and information on the history of reductions

Very few reductions showed metallic yields slightly below 80%, which was considered acceptable. The limit of 80% for the metallic yield was considered acceptable in view of the unavoidable presence of  $UO_2F_2$  and  $UO_2$  in  $UF_4$ . In addition, variations in charge bulk density caused by manual operations and/or fluctuations in the physical characteristics of  $UF_4$  powder are also unavoidable, which causes unavoidable fluctuations in the metallic yield.

#### 4. Conclusions

A suitable route to producing low enriched uranium metal by magnesiothermic reduction of  $UF_4$  was studied. The process was shown to be very sensitive to the temperature distribution within the reduction charge at the sparking of spontaneous ignition. The main factor limiting the uranium metallic yield is shown to be the position in the charge where the ignition occurs. If the ignition occurs at the top of the charge, the front of the exothermic reaction shifts from the top downwards, providing the heat needed to ensure efficient separation of the uranium metal from the slag. Otherwise, if ignition occurs at the bottom of the crucible, the reaction occurs but the separation is impaired as the slag is thrown to the top of the crucible, solidifying and trapping the uranium metal inside.

After making the needed modifications to adjust the temperature profiles inside the reduction charge, the process provided reproducible metallic yields around 80%.

The enriched uranium metal produced has been used to manufacture routine fuel elements for the IEA-R1 research reactor [21]. The natural uranium metal has been used for research work in the field of high uranium loaded fuel [22,23] and target development [24].

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