2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-06-9**

COMPACTION AND SINTERING OF NICKEL POWDER USED ENCAPSULATION OF IRRADIATION TARGETS

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ABSTRACT

The objective of this study was to develop an alternative way to produce targets for irradiation containing uranium, for the pair of Mo-99 production-Tc99m radionuclide. These targets were obtained by powder metallurgy, the compact serving as means for encapsulation a uranium cylinder to be irradiated. The targets were compacted in an axial hydraulic press applying different pressures up to 800 MPa. The sintering temperature was 600 °C in hydrogen atmosphere and it was used two sintering cycles, one for 4 h and the for 4 h plus 8 h time. The nickel powder was of high purity, that in order to provide the sealing of the fissile content within the compacted. The bulk density of compacted was evaluated by the method geometric. The porosity was measured by mercury porosimetry technique. The microstructure was investigated by optical microscopy. The results obtained with sintering powders involving confirm the feasibility of achieving a casing for uranium targets.

1. INTRODUCTION

The continued study of nuclear energy and reactions has provided the development of several techniques and applications, which use by society is steadily growing. Among these, it can be highlight the radioisotope production for use in nuclear medicine in diagnostics and therapy, biology and industry. Studies aimed at their usage, require them to be available in sufficient quantity and with high purity.

Molybdenum-99 (Mo-99) and decay product technetium-99m (Tc-99m) is one of the most widely used radioisotope in nuclear medicine. Even with recent technological advances, the participation of radiopharmaceuticals labeled with Tc-99m account for over 80% of all diagnostic radiology medical procedures in the world [1]. These data is also replicated in Brazil, which has a routine demand of approximately 450 Ci of Mo-99 a week [2]. The most important characteristic for the Tc-99m is the obtention of high-quality images providing low radiation doses to patients.

After irradiation in nuclear reactors, the Mo-99 is separated from the uranium and other

fission products by chemical processes and prepared for distribution to consumption centers, such as hospitals and clinics. Metal targets used for this type of production are generally encapsulated in aluminum or stainless steel to protect the metallic uranium and alloys of external chemical reactions and to maintain decay products occurring during the process of uranium irradiation with neutrons, within the target.

Compacted and sintered nickel rather than aluminum can be an alternative route for producing targets. These targets would benefit the post-processing for the chemical opening of irradiated material using the acidic dissolution route. The metals copper, nickel and iron are suitable for the acid dissolution procedure. The recoil range for the Cu and Ni is about 7 μm. A barrier 10 μm thick should be sufficient to stop the decrease and prevent atoms from reaching the target surface [3].

The knowledge of the characteristics of metal powders is required to understand its behavior in use. The determination of tolerance, specifications and related properties are important factors to ensure the reproduction powder behavior during processing. For this control is necessary: appropriate sampling technique, powder purity and suitable chemical composition, proper microstructure, particle size, particle shape, porosity, tapped density, flow velocity, specific surface, compressibility, green strength, dimensional changes of the compacted parts, among others.

The present work is a study of an alternative way to produce targets for irradiation containing uranium, aiming at the production of Mo-99-Tc-99m radionuclide. Therefore, the powder metallurgy was used, the compacted serving as a means of encapsulation for a uranium nucleus to be irradiated. Powder metallurgy is a manufacturing process that combines a compaction step with a densification by sintering aiming to achieve physical and metallurgical properties required for components [4, 5].

2. EXPERIMENTAL

The procedure adopted was to evaluate the densification after sintering of nickel powder at several compaction pressures. It was used nickel powder of commercial purity with grain size #325, average median size of 44 μm, aiming the sealing of uranium fissile contents within the compacted. In order to characterized acquired commercial nickel powder, it was chemically analyzed by the technique of energy dispersive X ray fluorescence spectrometry (EDXRFS), see TAB. 1.

The compaction was achieved using a hydraulic uniaxial powder metallurgy press fitted with upper punch and lower punch die system. It was used 2 g of nickel powder that filled a compaction die cavity with an inside diameter of 9 mm. FIG. 1 shows an image of nickel powder compacted sample in green state. The samples were compacted at pressures of 75 MPa, 150 MPa, 225 MPa, 300 MPa, 375 MPa, 450 MPa, 525 MPa, 600 MPa, 675 MPa and 800 MPa. After compaction, the green samples (this is a expression used in powder metallurgy to designate samples or parts that has not yet been sintered) were measured and the density was evaluated, either using the geometric method.

As this work was meant to obtain uranium containing targets for irradiation, the sintering temperature level employed for the nickel sintering was limited to 600 °C due to the fact that the uranium in the presence of nickel at 740 °C forms a eutectic, according to the U-Ni

phase diagram shown in FIG. 2. The chosen temperature allowed a safety margin with respect to the furnace operation and had been taken into consideration, allowing that sintering threshold was not exceeded.

Table 1: Result of chemical analysis (mass %) of the nickel powder used by the technique of energy dispersive X ray fluorescence spectrometry (EDXRFS).

Element	(mass %)
Ni	99.2 ± 0.5
F	0.30 ± 0.05
Mg	0.16 ± 0.05
Si	0.16 ± 0.05
Co	0.09 ± 0.03
Fe	0.09 ± 0.03
Ca	< 0.03
Cr	< 0.03
S	< 0.03
Zr	< 0.03

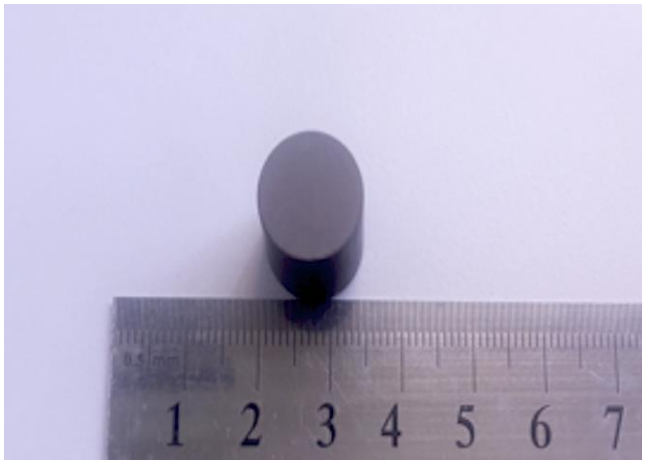

Figure 1: Image of a green compacted nickel sample.

The used thermal sintering was divided into two cycles. One single cycle with heating ramp of 10 °C per minute, a 4 h plateau at 600 °C followed by furnace cooling to room temperature. One double cycle with heating ramp of 10 °C per minute, a 4 h plateau at 600 °C, furnace cooling to room temperature and reheating at 600 °C for 8 h followed by furnace cooling. This was undertaken to observe the densification as a function of compaction pressure and the sintering cycle, see thermal cycles in FIGs. 3 and 4. The samples were sintered under hydrogen atmosphere at pressure of 20 cmHg.

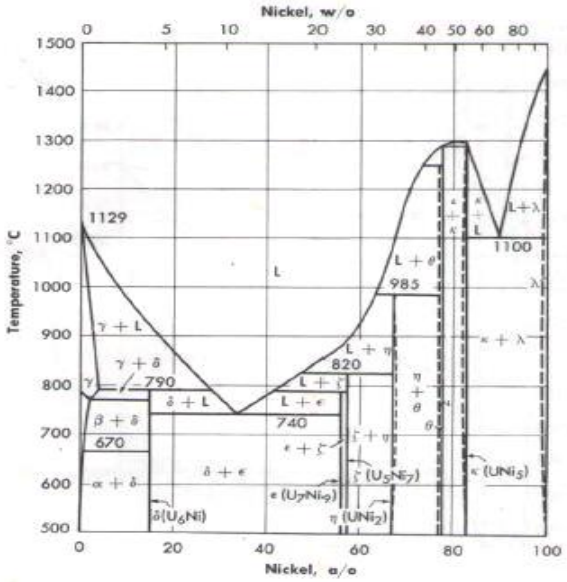

Figure 2: Phase diagram for nickel-uranium after Peterson [6].

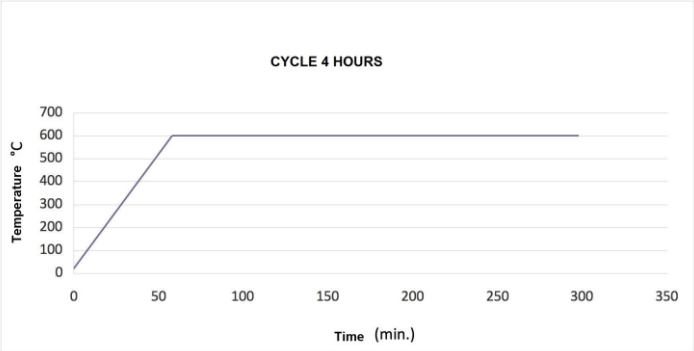

Figure 3: Sintering single cycle with heating ramp adopted for the obtained compacted and sintered at a temperature of 600 °C for a period of 4 h in hydrogen atmosphere.

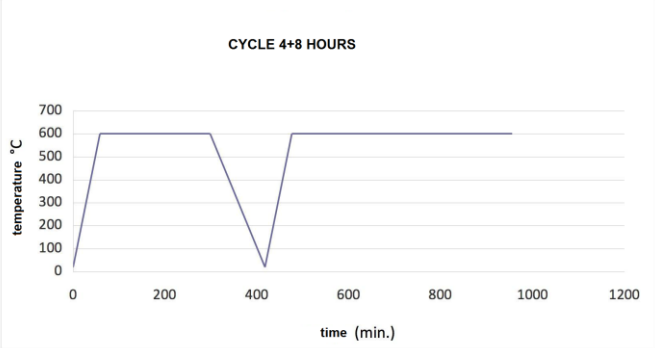

Figure 4: Sintering double cycle with heating ramp adopted for the obtained compacted and sintered at a temperature of 600 °C for a period of 4 h plus 8 h in a hydrogen atmosphere.

3. RESULTS AND DISCUSSION

The purchased nickel powder for the present work was analyzed for its morphology by scanning electron microscopy (SEM). The FIG. 5 shows the morphology of the nickel powder particles adhered on a carbon tape. Their morphology can be considered as a rounded shape according to the classification suggested by German [5]. The nickel powder supplier did not give any evidence of the nickel production route. However it seems that the nickel powder was probably obtained by water atomization followed by reduction and was considered commercially pure.

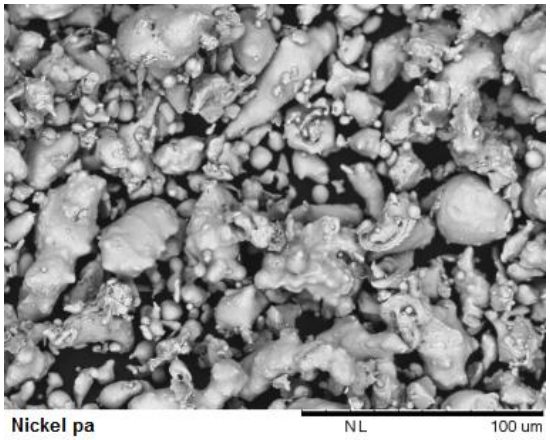

Figure 5: Scanning electron micrograph with secondary electrons of the used nickel powder adhered to a carbon tape.

The nickel powder had its particle size distribution evaluated by laser diffraction, see FIG. 6. The laser diffraction measures particle size distributions by measuring the angular variation in intensity of light scattered as a laser beam passes through a dispersed particulate sample. The distribution curve obtained showed that the median size of nickel powder particle was 23.8 µm.

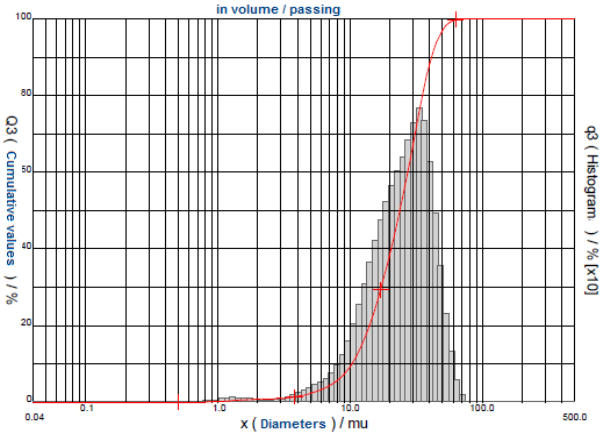

Figure 6: Curve particle size distribution of the nickel powder $D_{10\%}$ = 10.29 µm. $D_{50\%}$ = 23.80 µm. $D_{90\%}$ = 41.31 µm.

The 10 different compression pressures, namely 75 MPa (sample 1), 150 MPa (sample 2), 225 MPa (sample 3), 300 MPa (sample 4), 375 MPa (sample 5), 450 MPa (sample 6), 525 MPa (sample 7), 600 MPa (sample 8), 675 MPa (sample 9) and 800 MPa (sample 10) promoted an increase in the green density of the compacted as shown the graphs of FIG. 7. Maintaining the 600 °C for a period of over 4 h plus 8 h, it was not observed signs of significant changes in the densification of the compacted samples, regardless of the applied compaction pressure. During sintering cycles at 600 °C for 4 h and 4 h + 8 h, it was observed a small pressure raise, passing from the initial 20 cmHg to 24 cmHg and may indicate a reduction of nickel powder. A decrease in samples mass after sintering, was another indication of nickel oxide reduction during the thermal cycles, see FIG. 8. In this case the compacted samples at higher pressures led to smaller mass loss than the less compacted samples. For the calculation of the relative density it has been used 8.902 g/cm^3 for the nickel density.

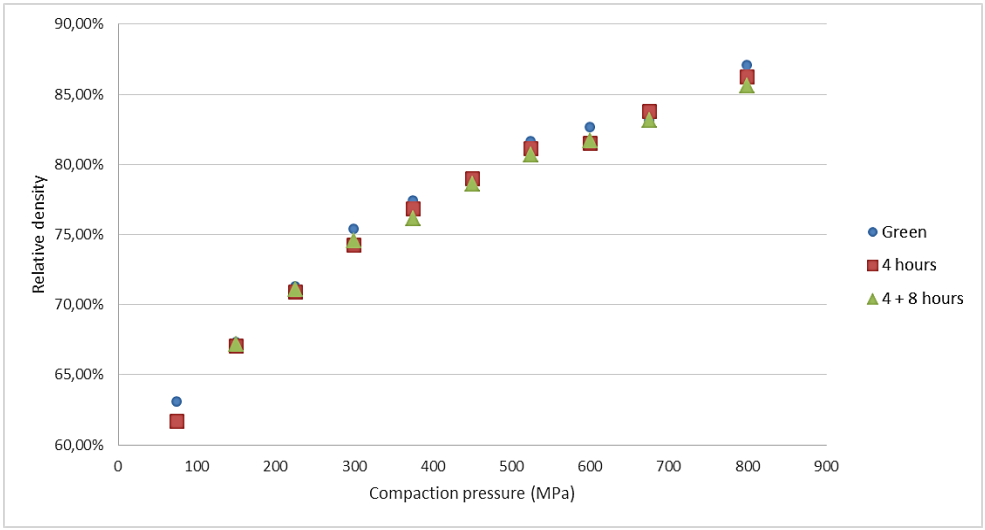

Figure 7: Compressibility chart relating to compaction in 10 different pressures and the relative density after the sintering cycles at 600 °C.

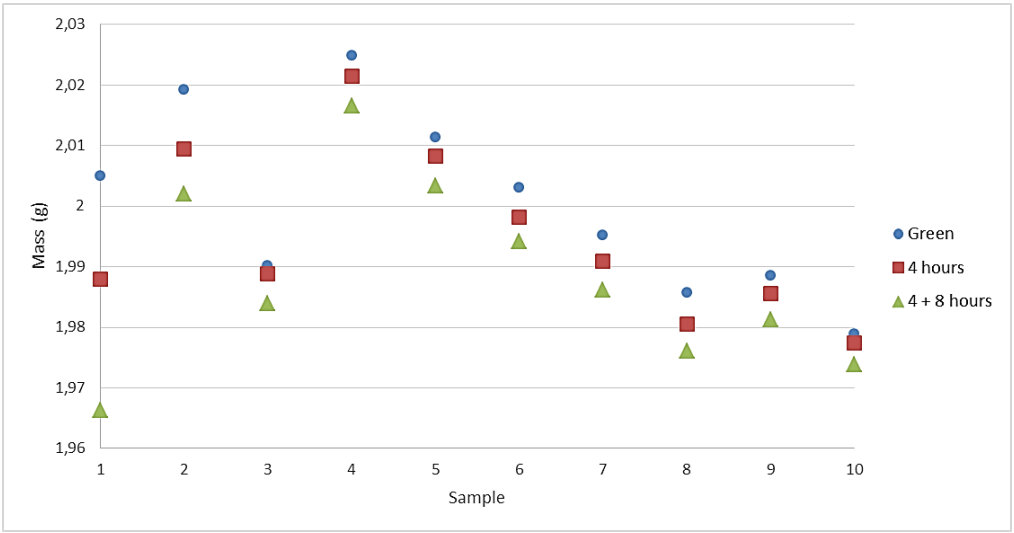

Figure 8: **Graph relating the compacted samples mass loss from green to sintered at 600°C.**

4. CONCLUSION

Maintaining the sintering temperature plateau at 600 °C for a period of over 4 h to 8 h, it was not observed signs of significant changes in the densification of the compacted samples, regardless of the applied compression pressure.

Different compression pressures promoted a greater relative density of the compacted and the pressure 800 MPa compaction showed a relative value of density above 85% of theoretical density of the nickel.

ACKNOWLEDGMENTS

The authors wish to express their gratitude for the financial support by CAPES (Grant Pró-Estratégia n. 2058/2012) and CNPq.

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