Materials Science Forum
ISSN: 1662-9752, Vol. 869, pp 484-489
doi:10.4028/www.scientific.net/MSF.869.484

Submitted: 2016-03-07
Accepted: 2016-03-10
Online: 2016-08-31

Sintered Nickel Casing for Irradiation Targets

R. S. L. Miyano, H. Takiishi, E. P. Soares, A. M. Saliba-Silva, J. L. Rossi

Av. Prof. Lineu Prestes 2242 - Cidade Universitária - 05508-000 - São Paulo - SP

Instituto de Pesquisas Energéticas e Nucleares, IPEN - CNEN/SP

rosatac@gmail.com

Keywords: powder metallurgy; nickel compacted; sintering; irradiation targets, casing.

Abstract: The aim of this work was to develop an alternative way to obtain casings used with irradiation targets containing uranium, for the production of the radionuclide Mo-99-Tc99m. The targets used for the production of Mo-99 are materials containing U-235 designed to be irradiated in a nuclear reactor. Usually these targets are encapsulated in aluminum or stainless steel. The idea here is to obtain casings by encapsulating a uranium button or a metallic cylinder with compacted and sintered nickel powder, this serving as a sealing for the fissile products occurring during U-235 irradiation. The sintered high purity nickel powder samples were compacted in uniaxial hydraulic press at 195 MPa. The sintering of the samples was carried out in an open-air furnace in an atmosphere with a certain control using titanium-machining chips at 600 ˚C. The samples bulk density was evaluated by the Archimedes' principle. The porosity of 20.08% was measured by mercury porosimetry. The microstructure was investigated by scanning microscopy revealing interconnected porosity and nickel oxide at the particles boundary surface. The results obtained by sintering of nickel powder according to the experimental undertaken, indicate the feasibility of achieving a casing for uranium targets.

Introduction

The continued study of nuclear reactions has provided the development of new and interesting techniques and applications. Among these, because of their growing use by society, can be highlighted the radioisotope production for use in nuclear medicine for diagnosis and therapy, also for usage in biology and industry. The studies aimed at their applications, mainly as radiopharmaceuticals require them to be available in sufficient quantity and with high purity.

The modified technetium-99m (Tc-99m) a molybdenum-99 decay product (Mo-99) 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 repeated in Brazil, which has a routine demand of approximately 360 Ci of Mo-99 a week. The nuclear characteristics of Tc-99m allow 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. The 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 contain the uranium fission decay products inside it [2]. Sintered nickel rather than aluminum can be an alternative route for targets production. 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 and the neutron recoil range for the Cu and Ni is about 7 μm. A barrier 10 μm thick should be sufficient to stop the recoil and prevent atoms from reaching the target surface.

The present work is a study of an alternative way to obtain casings for targets irradiation containing uranium, for the production of Mo-99-Tc-99m radionuclide. Hence, the powder metallurgy was used and the sintered part, or casing, serving as a means of encapsulation for uranium button to be irradiated. Powder metallurgy is a manufacturing process that combines compacting with a densification by sintering aiming to achieve the physical and metallurgical properties required for a particular component [3].

Experimental

The procedure adopted in the present work was to evaluate the densification of sintered nickel in an open-air furnace in which the oxygen was, as an attempt, to be controlled by using titanium-machining chips. The powder used was a commercial purity nickel powder, with particle size mesh # 325, particle size below 44 μm. The densification was necessary to allow the sealing of the fissile content within the compacted.

The characteristics of a metal powder are required to understand the behavior of the sintered part in service, to determine the specifications tolerance and related properties and the most important factor, to ensure the reproducibility of the powder behavior during processing. For this control, it is necessary to know: the chemical composition and purity, the microstructure, particle size distribution and shape, porosity, bulk density, green strength, dimensional changes of the compacted parts, among others. The commercial nickel powder purchased was chemically analyzed, see Tab. 1. The chemical analysis shows that the major component was nickel with minor contaminations of elements due to the production route.

Table 1. Results of semi-quantitative chemical analysis (mass%) obtained using the WDXRF (Wave-length Dispersive X-Ray Fluorescence Spectrometry) of the nickel powder used.

Elements	Nickel
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

In the preparation of each casing sample it was used 12 g of nickel powder. All samples were compacted in a uniaxial hydraulic press with a maximum capacity of 10 tons, which was manually operated. The used compaction tooling consists of two punches (top and bottom) with cylindrical geometry, i.e., with an internal diameter of 14 mm, made of tool steel. It was also used zinc stearate as a lubricant on the walls of the die and punches to reduce friction, prevent wear of the tooling and facilitate ejection of the compacted. The pressure used in making the initial compaction of the samples was 195 MPa. After making the green parts, the mass, height and diameter of the samples were measured. The density was determined by the geometric method, by the Archimedes' principle and then by mercury porosimetry.

For the sintering of the casing samples, a temperature threshold was set at 600 °C. As this research was meant for obtaining irradiation targets containing uranium, the 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 phase [4]. The temperature range and the safety margin with respect to the oven have also been taken into consideration, so that the threshold for sintering was not exceeded. A furnace muffle was used and a typical heating and sintering cycle is shown in Fig. 1.

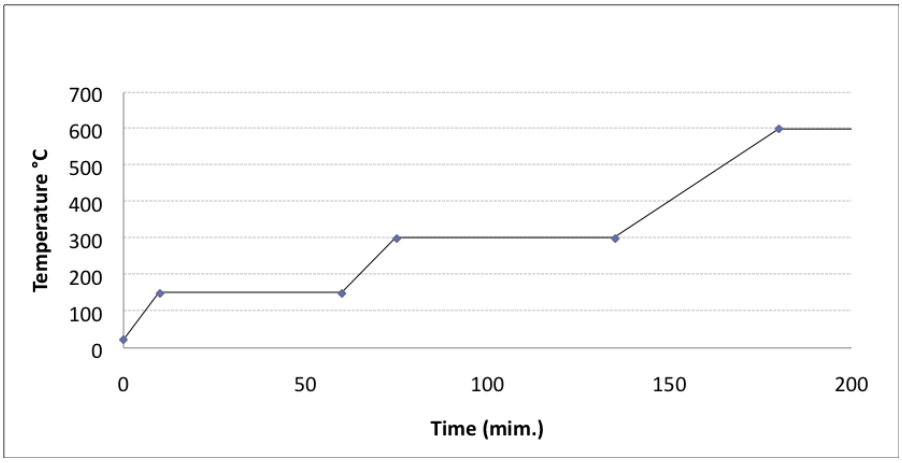


Fig. 1. Typical heating cycle for sintering used in the present work. The heating cycle continued at the 600 ˚C plateau up to 24 h.

The sintered samples were prepared for standard metallographic analysis. The samples were mounted in polyester resin, grinded in SiC paper and polished in diamond paste till grit 1 μm. The samples were carbon coated and observed in a scanning electron microscope - SEM.

Results and Discussion

In order to have a characterization of the nickel powder purchased for this work, its morphology was observed using a scanning electron microscopy (SEM), as is shown in Fig. 2. The nickel powder morphology can be considered as a rounded shape according to the classification suggested by German [3]. There was no evidence from the nickel powder supplier of which production route was used. The nickel powder resembled those obtained by water atomization and can be considered commercially pure.

The distribution of the average size of the nickel powder particle was measured using a laser diffraction particle size analyzer. Fig. 3 show the particle size distribution curve obtained for the nickel powder, and the average median size of the powder particle was 23.8 μm. Usually powders suitable for conventional powder metallurgy are typically smaller than 170 μm (-80 mesh). This upper limit is intended for the production of porous sintered parts. Most of PM powders have 30% to 40% fractions not exceeding 44 μm (-325 mesh). This is required by the need of a powder to flow in order to fill the die cavity. Moderate amounts of finer particles also provide a good balance between sintering speed and shrinkage [5]. The present powder size was bellow -325 mesh, e.g., $D_{90\%}$ = 41.31 μm, with a finer particles bellow $D_{10\%}$ = 10.29 μm, which complies with most powder metallurgy particle size distribution.

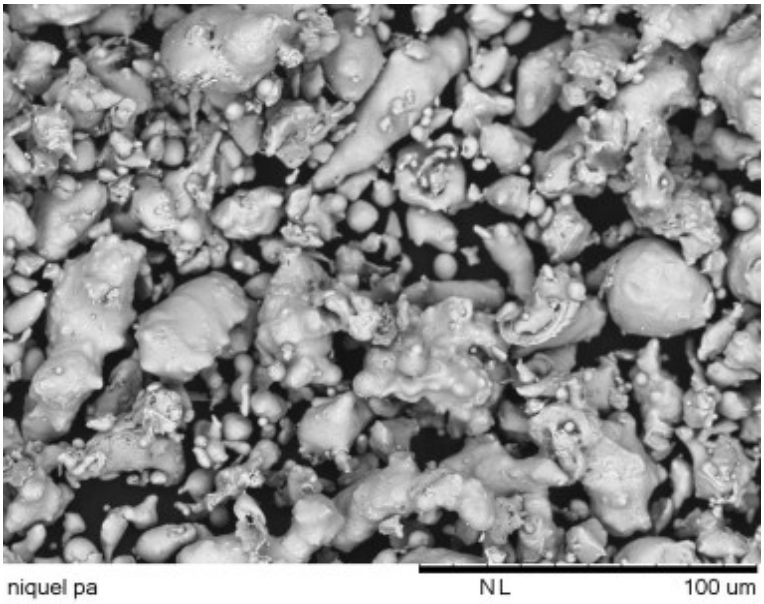


Fig. 2. Scanning electron micrograph with backscattered electron showing the morphological appearance of the nickel powder. It can be seen that the particles are predominantly equiaxed and rounded.

All samples were compacted at 195 MPa. After measurements, the average increase in density after sintering was of the order of 0.09 g/cm^3 for a sintering time of 3 h and 0.26 g/cm^3 for a sintering time 24 h. The graph on sintered samples densification at 600 °C for different elapsed times is shown in Fig. 4 as measured by the Archimedes' principle.

The mechanical compression test was conducted on samples compacted and sintered at 600 °C for different times of sintering. Ten samples were compacted and sintered at different times. From the obtained results it can be observed an increase in the mechanical strength of the nickel compacted samples with increasing sintering time, as shown in Fig. 5. For a sintering time 24 h 600 °C, the compressed compression load was 5645 kgf, giving a compression limit of 367 MPa before the compact collapsed.

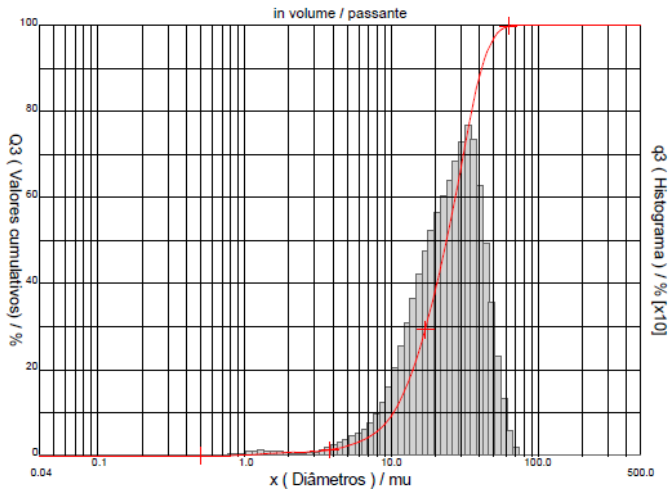


Fig. 3. Particle size distribution curve of the nickel powder. $D_{10\%}$ = 10.29 µm. $D_{50\%}$ = 23.80 µm. $D_{90\%}$ = 41.31 µm.

The mercury intrusion porosimetry is an important technique for the quantitative description of the porous structure of a solid [6]. This technique was developed from observing the behavior of a liquid on a porous solid, which is not wetted by the concerned liquid at atmospheric pressure. Mercury porosimetry test revealed a porosity of 20.08%, a median pore diameter (area) of 2.04 µm^2 and a total pore area of 0.060 sq-m/g.

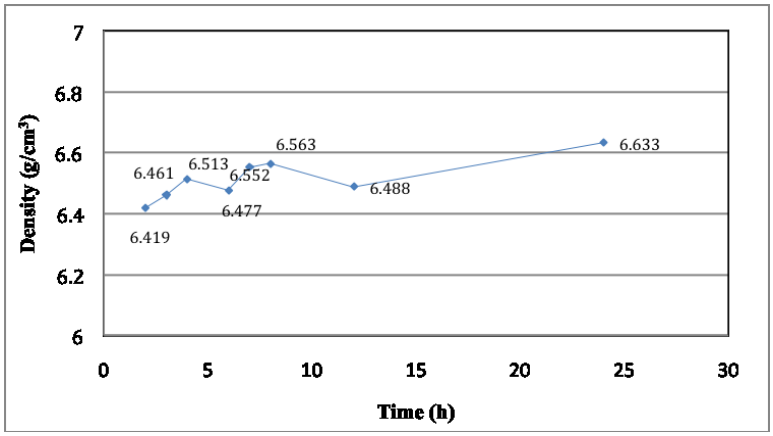


Fig. 4. Variation of density of compacted with sintering time at 600 °C in a protective atmosphere with titanium chips.

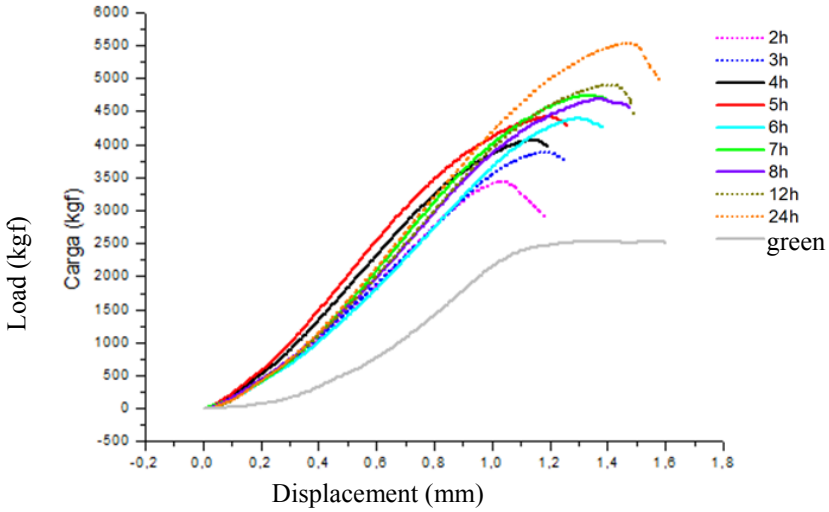
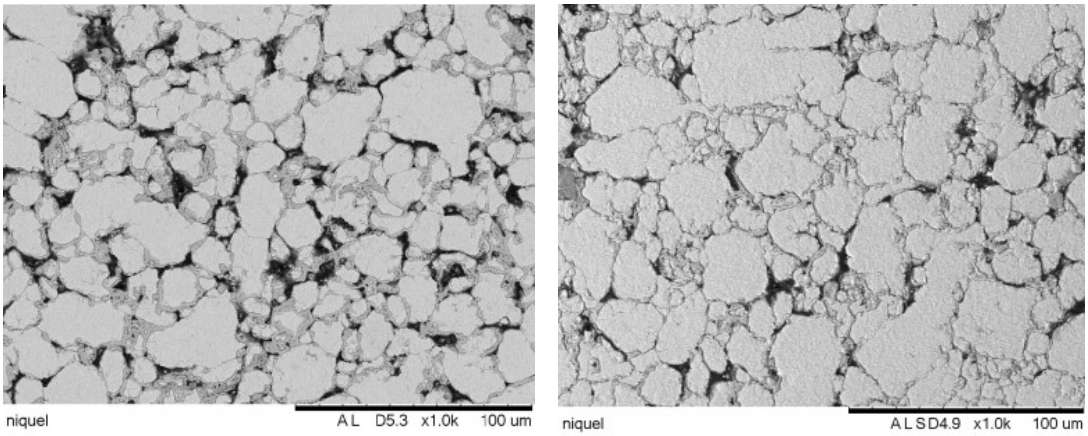


Fig. 5. Compressive load variation graph with the crossbeam displacement of nickel compacted samples in the green state and sintered at 600 °C for different times.

A sample was embedded in resin and prepared for analysis using the scanning electron microscopy technique - SEM. The sample was etched with *aqua regia* 1% for time of 1 s. The observed porosity was the interconnected type, confirming the results of mercury porosimetry that already indicated this kind of porosity, as can be observed in Fig. 6. The SEM showed the nickel particles, the porosity and an particles boundary phase consisting of nickel oxide.



a) b)

Fig. 6. a) Scanning electron micrograph with backscattered electrons from the nickel compact sintered for 3 h showing the nickel grains light contrast, a nickel oxide phase at the particles boundary in gray contrast and dark contrast porosity. b) Scanning electron micrograph with backscattered electrons of nickel compact sintered for 5 h showing densification. *Aqua regia* etching.

The presence of this nickel oxide at the particles boundary surface and certain amount sintering gave some mechanical tensile gain with the sintering elapsed time. This was due to the poor furnace oxygen control. The strength limit was increased from 162 MPa for the green sample to 367 MPa after sintering for 24 h at 600 ˚C.

Conclusion

The sintering process results with the nickel powder, confirm the possibility to obtain a casing for uranium targets. However, for a compacting pressure 195 MPa and sintering temperature 600 °C, with protective atmosphere with titanium chips, varying the sintering time in 2 h to 24 h, were not sufficient to prevent interconnected porosity, the presence of nickel oxide at the nickel particles surface, requiring other possible solutions permitted by powder metallurgy.

References

[1] International Atomic Energy Agency. Production And Supply Of Molybdenum-99 - Ntr2010 Supplement. 54th General Conference, IAEA, Vienna, 2010.
[2] P.J.B.O. Nishiyama: Neutronic And Thermal-Hydraulic Analysis Of A Device For Irradiation Of Leu Ualx-Al Targets For 99mo Production In The Iea-R1 Reactor. Master (Dissertation). São Paulo, 2012. Universidade de São Paulo (USP). (SP). (In Portuguese).
[3] R. M. German: *Powder Metallurgy Science*. (Metal Powder Industries Federation, 105, College Rd. E, Princeton, N.J. 08540, U.S A., 1984).
[4] D.E. Peterson Ni - U (Nickel - Uranium) (1991). In: Asm Handbook, Vol. 3 Alloy Phase Diagrams, Asm International, Materials Park, Ohio, (1992).
[5] C. Flake, F.C. Campbell, Metals Fabrication: Understanding The Basics. Asm International, Materials Park, Ohio, 2013. p..
[6] R.W. Johnson, L. Abrams, R.B. Maynard, T. Amick: Tappi Journal Vol. 82 (1) (1999), p. 239.