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THE SHIELDING AGAINST RADIATION PRODUCED BY POWDER METALLURGY WITH TUNGSTEN COPPER ALLOY APPLIED ON TRANSPORT EQUIPMENT FOR RADIO-PHARMACEUTICAL PRODUCTS

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ABSTRACT

Safety is mandatory on medicine radiopharmaceutical transportation and dependent on radiation shielding material. The focus of the present work is to minimize the use of harmful materials as lead and depleted uranium usually used in packeges transportation. The tungsten-copper composite obtained by powder metallurgy (PM) is non-toxic. In powder metallurgy the density and the porosity of the compacted parts depends basically upon particle size distribution of each component, mixture, compacting pressure and sintering temperature cycle. The tungsten-copper composite, when used for shielding charged particles, X-rays, gamma photons or other photons of lower energy require proper interpretation of the radiation transport phenomena. The radioactive energy reduction varies according to the porosity and density of the materials used as shielding. The main factor for radiation attenuation is the cross section value for tungsten. The motivation research factor is an optimization of the tungsten and cooper composition in order to achieve the best linear absorption coefficient given by equation $I_{(x)} = I_0 e^{(-ux)}$. Experiments were conducted to quantify the effective radiation shielding properties of tungsten-copper composite produced by PM, varying the cooper amount in the composite. The studied compositions were 15%, 20% and 25% copper in mass. The Compaction pressure was 270 MPa and the sintering atmosphere was in 1.1 atm in N_2+H_2. The sintering temperature was 980 ˚C for 2 h. The linear absorption coefficient factor was similar either for the green and the sintered compacts, due the amount of porosity did not affect the radiation attenuation. Thus the sintered was meant for size reduction and mechanical properties enhancement.

1. INTRODUCTION

The obtention of tungsten cobalt, nickel, iron alloys and compound by powder metallurgy - PM is worldwide studied. However, sintered tungsten-cooper alloys literature is scarce. Since tungsten and copper are insoluble together the technique of liquid-phase infiltration is a cost effective obtention route for composites, it is not possible attain a true alloy by casting. The difficulty to obtain a full density composite arises as tungsten contents arise too [1]. For purpose of shielding against radiation the density of composite is a very important item. The first technical choice indicates lead as a good shielding material then copper and tungsten. The lead has a specific density of 11.34 g.cm^{-3} and copper has 8.95 g.cm^{-3} and tungsten is

19.25 $g.cm^{-3}$, but copper is not harmful to the environment and to humans beings [2]. Copper instead, enhances the mechanical strength of the composite. The risk of crash and fire on transport devices is a possibility and lead has a melting point of 328 ºC [3], and it is easy to loose the shield format and it may occur an environmental contamination. The obtention of high density shielding material has been attempted with system tungsten-lead powders but liquid lead does not wet tungsten so this composite was not possible [4].

The composite materials of W-Cu generally contain a little amount of nickel [4]. This composite is used for electrical applications such as circuit breakers and etc. For shielding purposes, the most important element is tungsten due to its high specific density.

Some methods of W-Cu-Ni production could be used do produce W-Cu composite. The first method consists in compacting and sintering the tungsten to a skeleton and by capillary action fills the remaining porosity with the melting copper. This method allows the obtention of a composite W-Cu with specific density between 35% and 60% of the theoretical density [4]. The tungsten for shielding purposes need an adhesion element to make the system machinable and at a low processing temperature as possible is desirable. The classical method for preparation of cemented-carbide parts is to cold compact a powder mixture and pre-sinter operation allowing the material to be machined to shape and after undergo sintering. Because of the brittle nature of the powders, the compressed metal powder, green compacted, have a low mechanical strength, thus, substances, which act as both lubricant and binders, are added prior compaction. The zinc stearate is the most widely used additive usually, as a suspension in to an organic liquid. Such solutions contain 5-15% of lubricant and are sufficient to give 0.5% to 2.5% residual lubricant after the solvent volatilization, usually isopropyl alcohol. The compaction pressures used range from 200 to 250 MPa, although 500 MPa is most usual [5]. Presintering is carried out in reducing atmosphere (hydrogen) at the temperatures range from 900 ºC to 1150 ºC [3]. Another problem to care about on powder metallurgy is the porosity. A classical method to determine apparent and theoretical densities is measuring the amount of liquid required to saturate the interconnected porosity. The total porosity will ever be greater than the interconnected porosity. The total porosity on a body produced by powder metallurgy has an unknown amount of closed pores. In order to completely eliminate the closed porosity, sintering to densities in excess to 95% of the theoretical density is required. The interrelationship between total porosity and open pores ignores the fact that the determination of the latter by impregnation is dependent to some extend on the liquid used [5]. Some parameters like yield strength, tensile strength, ductility, and elastic modulus each decreases as the amount of porosity increases. Densities in the range 95-99% theoretical were obtained by hot closed-die forging (repressing). The mechanical properties increase linearly with density up to approximately 98% of the theoretical value. At higher density levels increase mechanical property at the higher rate confirming the strong influence of the last traces of porosity [6].

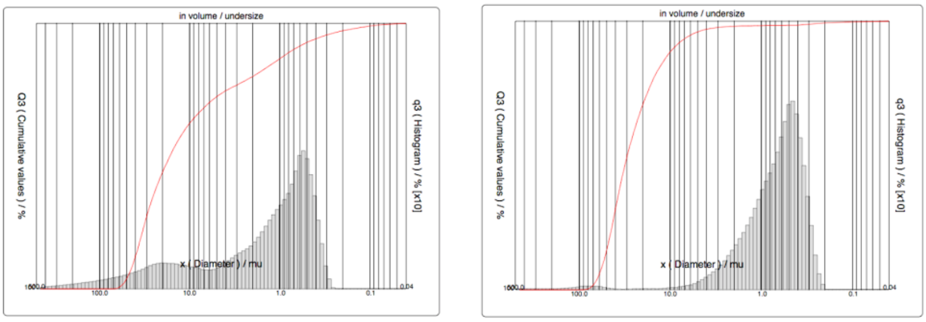
When powder metallurgy materials are used as a shield against radiation, porosity should be well analyzed. Internal porosity makes shielding effect lower than predicted by linear coefficient of absorption or attenuation through the bulk of the materials.

The gamma-ray attenuation by the specific material density follows an exponential law and the ratio of the attenuation coefficient related to the density of the attenuating material is nearly constant for all materials. When gamma radiation of intensity I_0 is incident on an absorber of thickness L, the emerging intensity (I) transmitted by the absorber is given by the exponential equation $I_{(x)} = I_{(0)}e^{(-ux)}$ where (u) is the attenuation coefficient (expressed in cm^{-1}). The ratio $I_{(x)}/I_{(0)}$ is called the gamma-ray transmission. The exponential attenuation for three different gamma-ray energies shows that the transmission factor increases with increasing gamma-ray energy and decreases with increasing absorber thickness (x). Measurements with

different sources and absorbers show that the attenuation coefficient (u) depends on the gamma-ray energy and the atomic number (Z) and specific density (g/cm^3) of the absorber [7]. Alpha and beta particles have a well-defined range or stopping power distance however, gamma rays do not have a unique range. The reciprocal of the attenuation coefficient 1/u has units of length and is often called the mean free path. It is also the absorber thickness that produces a transmission factor of $l_{(0)}/e$ [7, 8]. The action between a photon of gamma ray with matter have many mechanisms of interaction. The main are photoelectric effect, the Compton effect and pair production. In the photoelectric effect the incident photon is totally absorbed with the transfer of its energy to an orbital electron of the inner layers, ejecting it. In the Compton effect the photon is deflected by an electron from the outer layers and the photon transfer part of photon energy to this electron. In the Compton effect the photon is not absorbed and with residual energy continues to interact with other electrons. The pair production (electron and positron) occurs when the energy of the incident photon is higher than the rest mass of these particles, i.e., greater than 1.02 MeV [9]. The occurrence probability of one of these types of interaction depends on the energy of the photon of gamma rays and atomic number of the elements forming the medium [9]. Since the interactions the gamma photon trough the bulk of pure material is well quantified [10], it may be used to calculate the amount of porosity variation with the attenuation coefficient of each element fraction in the green compact.
This study was conducted to produce X%W+Y%Cu composites by PM to be used as an alternative as shielding material. The work task is to reduce copper amount in this composite as low as possible. The amount of tungsten should be optimized because it has effective cross section for successful radioactive absorption coefficient.

2. EXPERIMENTAL

In order to prepare the experimental samples it was used commercial metallic powder. The powders mixture was prepared with tungsten and copper in proportions considered useful to be used as a material for radiation shielding. An initial set of nine samples of W-Cu was prepared with 15%, 20% and 25% in mass of copper.
The commercial powders were particle size analyzed in CILAS equipment and the particle size distribution characteristics of tungsten and copper powders are shown in Fig 1 and are summarized in Tab. 1. For a good homogenization of the particles mixture, the set of samples underwent a 30 min mixing in a ball milling equipment, with stainless steel balls of 55 mm diameter rotating at 90 rpm. After the milling process the powder was mixed again on a rotating Turbula for an additional period of 90 min.



a) b)

Figure 1. Particle size distribution for the used (a) tungsten and (b) copper powders.

Table1. Tungsten and copper powder particles properties

Metal powder	Median particle diameter d_{50} (μm)	Purity (mass) (%)
W	16.6	98.0
Cu	29.7	99.9

The green samples were compacted in an axial hydraulic system with pressure of 250 MPa and pressed again in a cold isostatic press at 250 MPa to attain a green compact with better rigidity. The final characteristics of W-Cu green samples have a composition, weight, diameter and thickness as showed in Table 2.

Table 2. Green compacted samples parameters

Sample #	Powder composition (%)	Total (g)	Tungsten (g)	Copper (g)	Diameter $ø_i$ (mm)	Thickness H_i (mm)	Theor. green porosity (%)
1.1	85W15Cu	50.22	42.55	7.67	25.5	9.8	38.72
1.2	80W20Cu	50.08	40.23	9.84	25.5	9.5	34.27
1.3	75W25Cu	50.10	32.55	17.54	25.5	9.6	25.54
2.1	85W15Cu	40.04	35.52	4.52	25.5	7.3	36.99
2.2	80W20Cu	40.06	34.10	5.95	25.5	7.4	35.54
2.3	75W25Cu	40.12	32.63	7.49	25.5	7.6	34.78
3.1	85W15Cu	30.07	24.01	6.06	25.5	5.9	36.18
3.2	80W20Cu	30.03	22.03	8.00	25.5	5.6	28.77
3.3	75W25Cu	31.63	28.19	3.44	25.5	6.2	39.92

The sintering process was undertaken with three different parameters by samples set. First of all, the samples #1.1, #1.2 and #1.3 was sintered at 980 ºC in furnace with air atmosphere during 120 min and it resulted completes oxidized samples, very fragile. The second process, with samples #2.1, #2.2 and #2.3 was sintered at 980 ºC in a furnace with in vacuum at a pressure of 10^{-2} atm and 180 min. This procedure resulted the same behavior as samples #1.1, #1.2 and #1.3. For samples #3.1, #3.2 and #3.3 was sintered at 980 ºC with N_2+H_2 atmosphere at 1.1 atm and 120 min. The results again showed samples with a poor sintered condition, see Figs. 2 and 3. With this poorly sintered samples, the radiation transmission experimental procedure was performed.
Regarding the theoretical density for the green and sintered samples, samples #3.1, #3.2 and #3.3 could be compared (see Tab. 3). It is noticed that the theoretical density increase of 2%~5% not occur as reported in literature [11]. However, the samples dimension increased by 20% in average and it was due to swelling from the used copper. These effects should be

better studied and others new sample needs to be prepared with low temperature copper alloy instead of pure copper.

Table 3. Green compacted samples parameters

Sample #	Powder composition (%)	Total weight (mg)	Diameter $ø_f$ (mm)	Thickness H_f (mm)	Theor. sintered density (%)	Theor. green density (%)
3.1	85W15Cu	29649.2	31.2	7.7	50.3	63.8
3.2	80W20Cu	29928.7	30.5	7.6	53.9	71.2
3.3	75W25Cu	31387.5	31.0	8.2	50.7	60.1

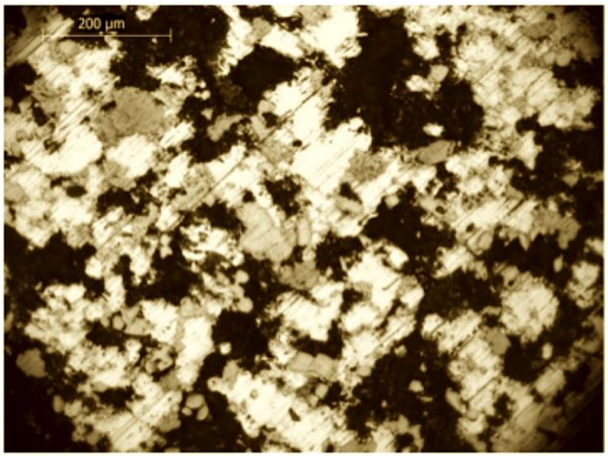


Figure 2. Optical micrograph of sample 75W25Cu surface, showing the W particles (median grey contrast), Cu particles (light gray contrast) and porosity (dark contrast). Not etched.

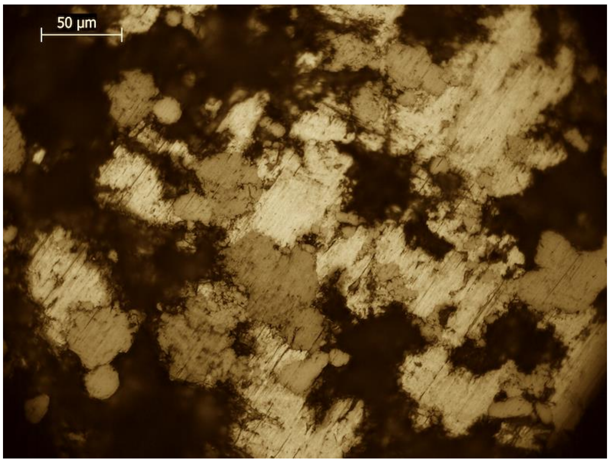


Figure 3. Optical micrograph of sample 75W25Cu surface, at higher magnification, showing the W particles (median grey contrast), Cu particles (light gray contrast) and porosity (dark contrast). Not etched.

The tungsten particle is dispersed in copper after it was melted during sintering process. The sintering process with melting copper did not produced samples with adequate specific density. Even if the densification effect was not achieved, the used samples were used to evaluate the radiation shielding effects. The radiation experiment was affected by the sample porosity directly. The amount of porosity behaved, as air in the attenuation coefficient and the parameter formula of elements compound should be calculated with (u_W+u_{Cu}+u_{Air}). The parameter u_{Air} is known or could be calculated.

The aim of this experimental procedures is to get the difference between the reference value $I_{(x)}$ experimental and $I_{(x)}$ theoretical. The $I_{(x)}$ theoretical can be calculated considering that the sample material is fully dense (in percentage of each element in the composite). This takes into consideration that there is no porosity but a continuous composite material.

The $I_{(x)}$ is obtained with an experimental set-up with a radioactive Co^{60} source of a scintillography, Ortech model 266 PM Base detector and an analog to digital converter for separation power per channel and count (intensity) of photons in each channel. The assembled system (see Fig. 4 and 5) allows the measurement of the value of the radiation attenuation along the way inside the composite.

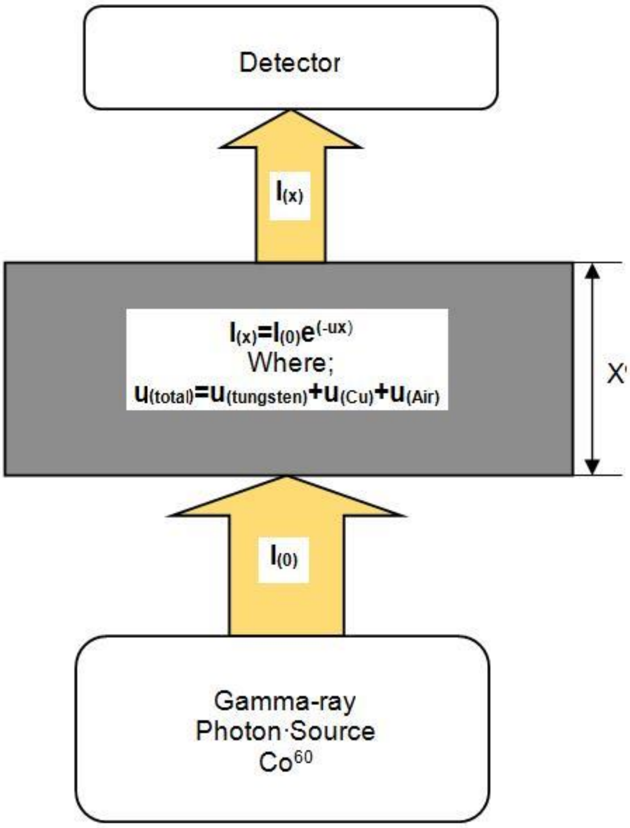


Figure 4. Schematic set-up diagram for the linear attenuation coefficient measurement.

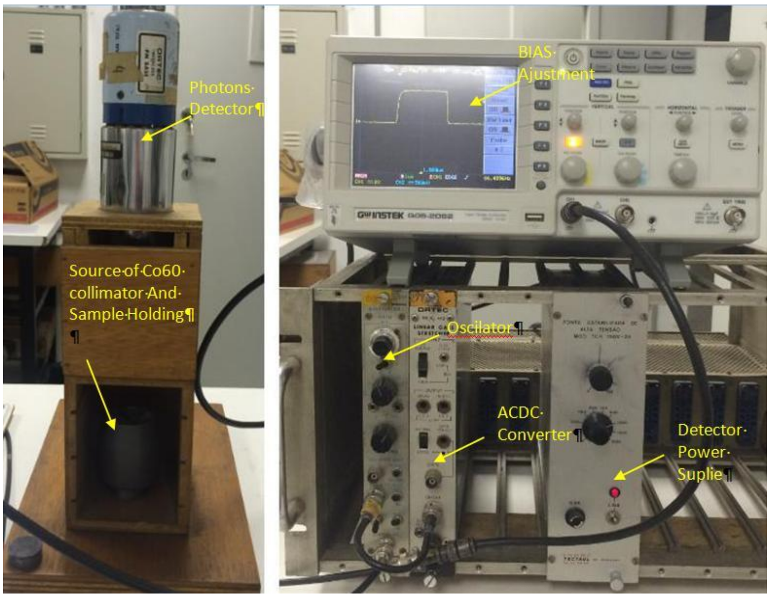


Figure 5. Experimental set-up showing the main used equipments.

3. RESULTS AND DISCUSSION

The first step adopted for the experimental assay was to measure the maximum emission value condition ($I_{(x)}/I_{(0)} = 1$) without absorbing material between the emission source and detector. This experimental condition showed maximum intensity of Co^{60} source in two distinct energy peaks. The intensity ($I_{(0)}$) at the peak of 957 photons with energy 1130 keV and 701 photons with energy 1330 keV. To increase the accuracy of the results of the tests were repeated twice at 5 min interval (see Fig. 6).

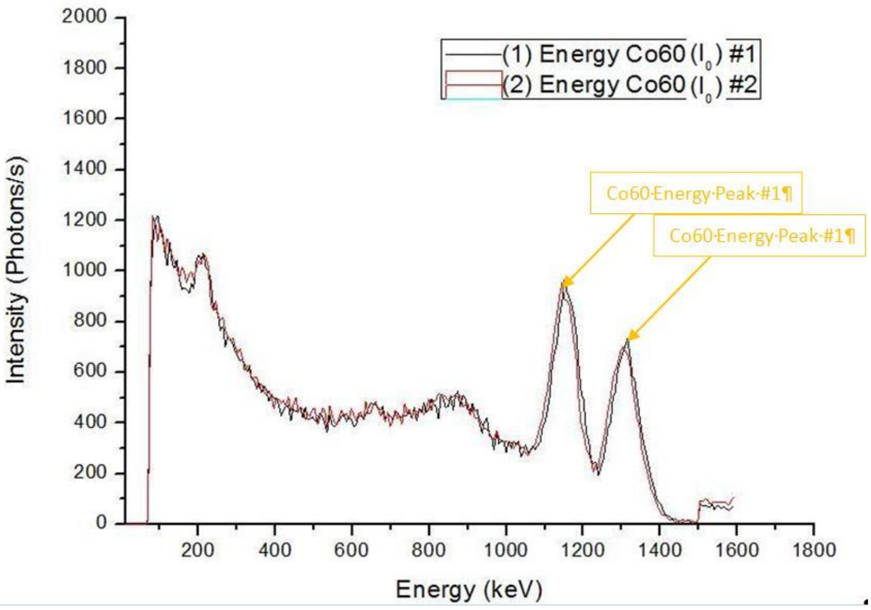


Figure 6. Free path for Co^{60} radiation emissions.

In the graph showed in Fig. 7 can visually compare the values of attenuation between the peaks of each sample containing 15%, 20% and 25% by mass of copper with respect to the peak of maximum intensity ($I_{(0)}$). Tab. 4 shows the relative intensity values ($I_{(x)}$) for each peak energy per proportion of copper in the sample.

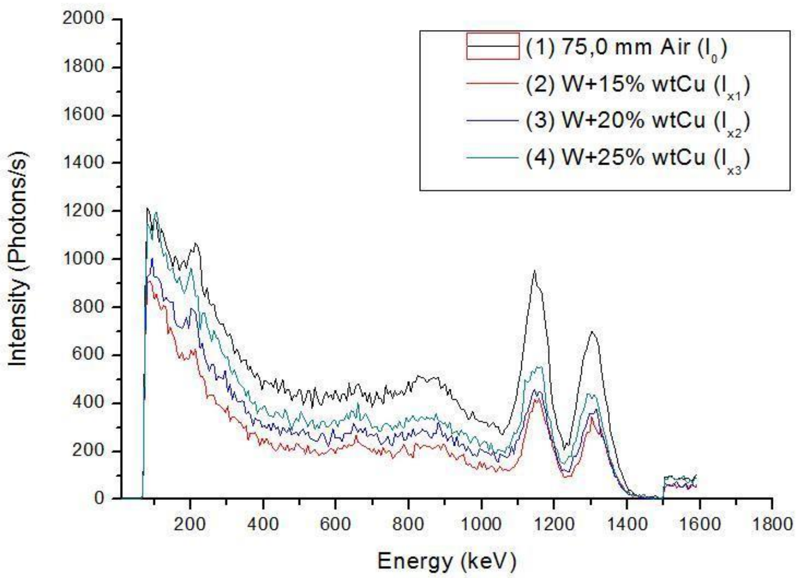


Figure 7. Intensity absorption by percent amount of tungsten in the samples.

Table 4. Average of linear absorption coefficient by amount of tungsten in the samples

Sample	I(0)	I(x)	u (cm^{-1})	Energy (keV)
W+15%Cu	957	358	9.701	1164
W+20%Cu	963	438	7.864	1145
W+25%Cu	948	555	6.341	1138
W+15%Cu	701	207	4.824	1305
W+20%Cu	712	324	4.107	1302
W+25%Cu	709	381	3.780	1319

For the source of Co^{60} the energy values are close to that will be imposed to shield against radiation for the pair of radionuclides ^{99}Mo-^{99m}Tc transportation. Thus one can use the experimental values to obtain more accurately the attenuation coefficient for Mo^{99}.
The compacting and sintering conditions should be improved as well powders particle relative size.
The median relative powder size of tungsten of 16.6 um to copper of 27.7 um, from the commercial suppliers, should be inversed, i.e., the copper powder should be finer then tungsten in order to coat the tungsten particle evenly
Finally the samples compacted at 250 MPa showed a poor mechanical strength in the green and after sintering state.

4. CONCLUSIONS

The attenuation data is conclusive that sample with the larger tungsten ratios imply higher attenuation of radiation through the sample than the samples with higher percentage of copper.

There is a necessity of changing the copper powder should to a 98% of purity adding 2% of others particles (Co and Ni), to allow a more wettability of the tungsten particles by the copper.

From the experiments with radiation attenuation it is possible to correlate the amount of tungsten with the intensity absorption by percent amount.

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