

MATERIALS SELECTION FOR A TRANSPORT PACKAGING OF Mo-99

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ABSTRACT

The radiopharmaceuticals are radioactive isotopes used in nuclear medicine for more accurate diagnosis and treatment of diseases or dysfunctions. Currently, the most important radionuclide for the preparation of radiopharmaceuticals for diagnostic purposes is technetium-99m (Tc-99m), a product of the radioactive decay of molybdenum-99 (Mo-99). The aim of this work was the materials selection that can enable the manufacture of a package for Mo-99 transport with the aid of CES EduPack program and the methodology developed by Ashby. The ESTAR program was used to check the occurrence of *Bremsstrahlung* and the XCOM program was used to calculate the attenuation coefficient of gamma radiation from some of the selected materials for the shield; after, the thickness required for radiation shielding was calculated. From the results, the materials selected as potential candidates for the manufacture of the shielding were the tungsten alloys. Related to the thermal insulation and the impact protection, woods, plywoods and particleboards stand out. With regard to internal and external coatings, the selected materials focus on groups of steels and nickel alloys.

1. INTRODUCTION

The radiopharmaceuticals can be defined as substances that contain a radioactive isotope which are used in more accurate diagnosis and treatment of pathologies and dysfunction (radiotherapy). Currently, one of the most important radionuclides for the preparation of radiopharmaceuticals for diagnostic purpose is the technetium-99 metastable (Tc-99m) [1], a product of the radioactive decay of molybdenum-99 (Mo-99). In Brazil, there is no other way to obtain this radionuclide than by import, once there is no reactor capable to produce it largely.

In May 2009, the Canadian company *MDS Nordion*, responsible for nearly 40% of the global Mo-99 production, interrupted its activities after a failure in the reactor which supply the radionuclide [2]. The event caused a crisis on radiopharmaceutical market, affecting its application in nuclear medicine. In early 2014, the National Commission of Nuclear Energy (*Comissão Nacional de Energia Nuclear - CNEN*) and the Brazilian Society of Nuclear Medicine (*Sociedade Brasileira de Medicina Nuclear - SBMN*) discussed about the occurrence of another crisis in Mo-99 supply, expected to happen in 2016 [3,4,5].

In this scenario, Brazil is preparing itself to face the possible crisis in Mo-99 supply; for this, the country is investing in a project that aims to achieve independence in the production of this radionuclide: the Brazilian Multipurpose Reactor (*Reator Multipropósito Brasileiro - RMB*) project. The reactor will provide radioactive isotopes for medicine and research in areas like energy, agriculture and material sciences. The development of this project is being accomplished by *CNEN* and its construction is being made in an area granted in part by the Navy's Technological Center of São Paulo (*Centro Tecnológico da Marinha de São Paulo - CTMSP*) and in part by the state government of São Paulo [3].

In this context, the development of a transport packaging for Mo-99 is very important, once it has to be transported from the place where it is produced to the place where occurs its cracking. It is of interest that this transport packaging can be manufactured with national resources and technology.

In Brazil, the main responsible agency for the transport regulation of radioactive materials is the *CNEN*, that establishes the *CNEN-NE-5.01* regulation in order to control the activities related to the transport of radioactive materials. The regulation determines requirements about radiation protection and security to ensure an adequate level of radiation exposure for people, goods and the environment, by establishing the specifications about types of radioactive materials, transport packaging selection, design requirements and tests [6].

The aim of this work was the materials selection that can enable the manufacture of a package for Mo-99 transportation, which product of the radioactive decay is used for diagnose purpose in nuclear medicine. According to the *CNEN-NE-5.01* regulation, Mo-99 has to be transported in a Type B package, which stands normal and accidental conditions of transport.

2. THEORETICAL FUNDAMENTALS

The radiopharmaceuticals can be defined as substances which are used in diagnosis and therapy because of their pharmaceutical form, quality and quantity of the radiation emitted by a constituent radioisotope, regardless the route of administration (oral, intravenous, inhalation, among others) [1].

Currently, one of the main radionuclides used in the radiopharmaceutical preparation for diagnostic purposes is Tc-99m, which presents ideal characteristics for nuclear medicine, once it is a low energy gamma mono-emitter (140 keV), it has a relatively short half-life (approximately 6 h) and it hasn't have a particle radiation emission (Tc-99m nuclei decay by the emission of γ radiation). Such characteristics allow an excellent image resolution, without a dosimetric commitment for the patient [1].

The Tc-99m is a product decay of Mo-99. Approximately 87,5% of the Mo-99 atoms of a sample decay by the emission of β radiation and produce Tc-99m nuclei which, in turn, decay by the emission of γ radiation to produce Tc-99.

There are three parameters used in order to control and reduce the radiation exposure to materials and people, emitted by a source: time, distance and shielding. The cumulative dose of a individual exposed to radiation is directly proportional to the exposure time, while the dose rate is inversely proportional to the square of the distance between a point source and

the individual. When time reduction or distance increase is not sufficient to ensure the maximum dose received by an individual, the shielding offers a reliable way to limit the dose, by using materials capable of stopping or attenuating the incident radiation [7].

2.1 β Radiation Shielding

Charged particles dissipate energy by colliding with the atoms particles of the shielding material [7]. The distance traveled until the kinetic energy comes into thermal equilibrium with the medium particles is named range. So, to shield particle radiation, the shielding material must to have a thickness greater than its range (related to this material).

β decay present two types of radiation emission: β particles and stopping power (also referred as *Bremsstrahlung*, from German). When β particles interact with the electric field of a high atomic number nucleus, they reduce their kinetic energy and suffer a change in initial direction, emitting the collision energy difference in a electromagnetic wave form, called stopping power.

In a β decay, the stopping power is the most important radiation to design a shielding, because it presents the greatest risk – while β particles can be shielded by a thickness greater than their range, stopping power needs to be analyzed in the same way that γ radiation, as described below.

2.1 γ Radiation Shielding

Because of its waviness, the γ radiation is capable of penetrate a material traveling large thicknesses until suffer the first interaction [7]. Once it is an electromagnetic wave, the γ radiation cannot be stopped; in this case, the right term to define the process is attenuation.

There is a probability of γ radiation interaction with the atoms of shielding material. Once it occurs depending on the atomic number of the shielding material and the energy of the incident radiation, three main effects are expected: Compton effect, photoelectric effect and pair formation, prevailing one to another, depending on the characteristics cited before. So, attenuation will occur because of the interaction with the atoms of shielding.

Due to this probabilistic characteristic of interaction, is not possible to fully stop the radiation; it means that this type of radiation cannot be completely eliminate, it can just be reduced to acceptable levels of dose (established by *CNEN-NE-3.01* regulation [8]).

The level of attenuation of γ radiation is dependent on four main factors: the energy of the incident radiation, atomic number and density of shielding material elements, and the thickness of the shielding [9]. The attenuation of γ radiation can be described by EQ.1:

$$I = I_0 \cdot e^{-\mu x} \quad (1)$$

I represents the radiation intensity after pass through the shielding, I_0 is the initial intensity of the radiation, μ is the total attenuation coefficient and x is the shielding thickness.

3. MATERIALS AND METHODS

The methodology used in this work is diagrammed in FIG.1 and is described below.

A design analysis of the transport packages found at Radioactive Material Packaging (RAMPAC) [10] was made do define the main parts of a transport packaging for Mo-99.

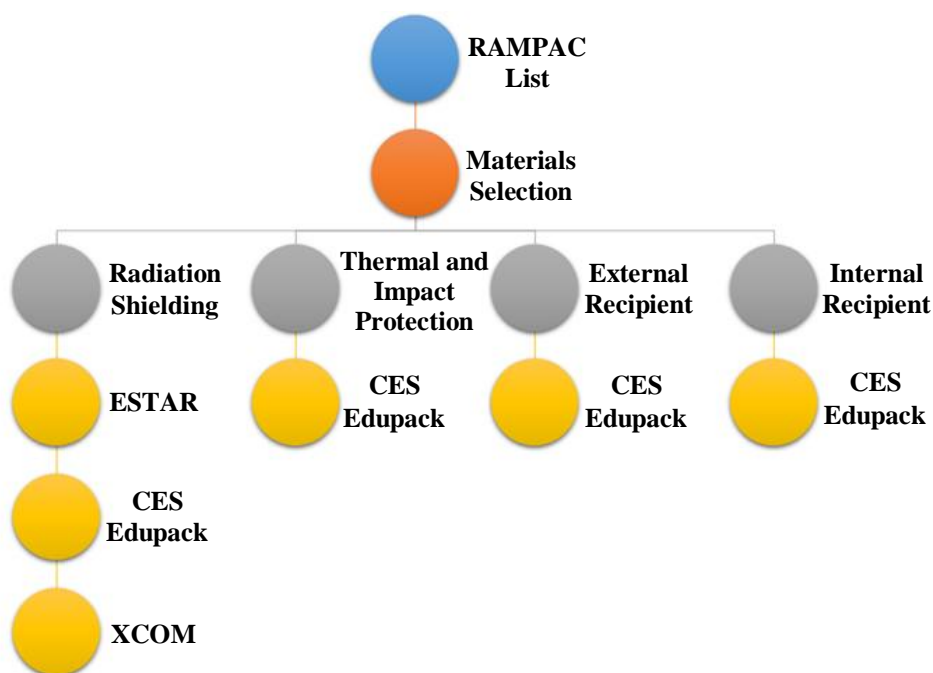


Figure 1: Procedure performed in this work.

The materials selection was performed to choose the best materials that can be used in the manufacture of the transport packaging, with the aid of CES Edupack software (2013 version) and with the methodology developed by Ashby [12], which consider the design requirements, as presented in FIG.2.



Figure 2: Flowchart of materials selection.

The ESTAR program was used to verify the occurrence of stopping power and the XCOM program was used to calculate the total attenuation coefficient from some of the materials

selected to the shielding part. With the total attenuation coefficient, the thickness needed to reduce the incident radiation to half (Half Value Layer - HVL) and to tenth (Tenth Value Layer - TVL) of the initial value was calculate with the aid of EQ.1.

4. RESULTS AND DISCUSSION

In the following, the results obtained in each stage described previously are presented and discussed.

4.1 RAMPAC List

An analysis was performed in order to define the main parts of a transport packaging for Mo-99; after the analysis, the main parts of this type of packaging was four: radiation shielding, thermal and impact protection, external recipient and internal recipient. The majority of the packages analyzed presented a material able to perform both, thermal and impact protection, that is the reason why it was treated like one part.

4.2 Radiation Shielding

First, the occurrence of stopping power emitted by Mo-99 had to be verified. Using ESTAR program, it was found that the stopping power produced in photons emission of Mo-99 decay presents order of magnitude ten times smaller than the photons produced by γ decay. So, the stopping power was not taken into account in the thickness calculation of the shielding.

After the verification, the materials selection was performed with the aid of CES Edupack software, as flowchart below (FIG.3).

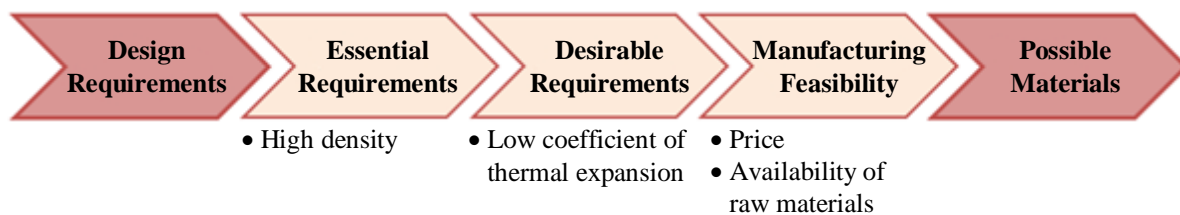


Figure 3: Materials selection flowchart with the requirements for the radiation shielding part.

The attenuation of gamma radiation is usually more effective with high density materials; at the beginning, the material used to start the analysis was lead (Pb) which is widely used in nuclear application as shielding. So, Pb is the material that set the lower limit of density ($1.13 \times 10^4 \text{ kg/m}^3$).

According to *CNEN-NE-5.01* regulation, for a Type B package, the integrity of the package should not be reduced with temperature variation (from - 40 °C to 70 °C) [6]. Thus, the

material has to have low coefficient of thermal expansion, so the package will not suffer a expansion or retraction that compromises its structure. Using data obtained from the RAMPAC list, among the materials used for the shielding part, Pb is the material that presents the highest value of coefficient of thermal expansion (among the materials selected before) [10], so the value was set at $29.2 \mu\text{strain}/^\circ\text{C}$.

Finally, the graphic was constructed using density relative to price (FIG.4). The gray spectrum refers to the excluded materials.

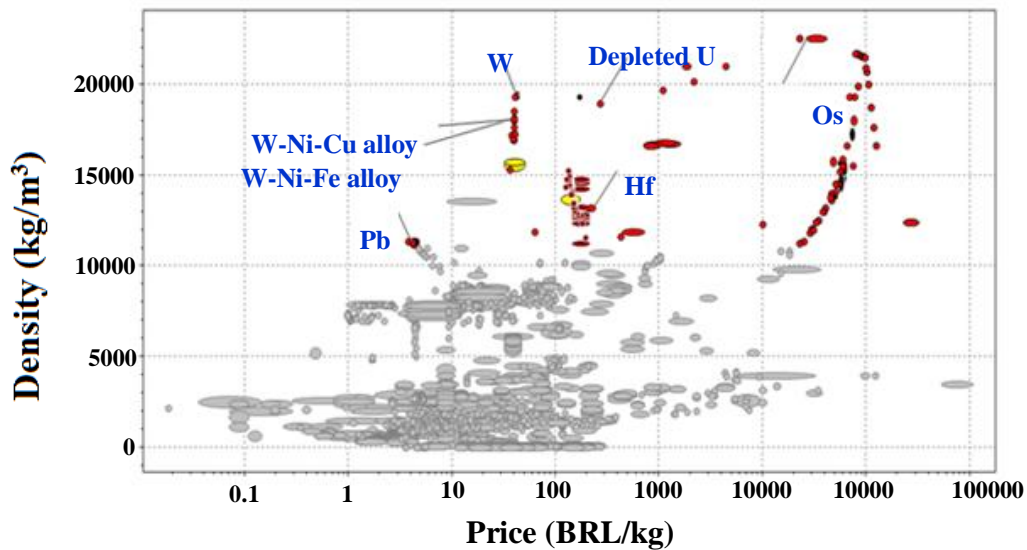


Figure 4: Density relative to price, presenting some of the selected materials that can be used for radiation shielding part of the packaging.

By fixing the maximum value of accepted price in 327 BRL/kg, the price of depleted uranium (U), the materials that still included were: lead (Pb) alloys, molybdenum (Mo) alloy, tungsten (W) alloys, hafnium (Hf) and depleted uranium (U). Analyzing the manufacture feasibility, the use of depleted U, Mo alloy and Hf are not practicable, because of the availability of raw materials and/or the processing form.

So, for the next step just Pb alloys and W alloys were considered. For these, the total attenuation coefficient of each one was calculated with the aid of XCOM, taking into account the emitted photons with the most significant relative intensity (I_R) [12]: 140.511 keV ($I_R = 37.3\%$); 181.063 keV ($I_R = 50.1\%$); 739.500 keV ($I_R = 100\%$) e 777.921 keV ($I_R = 35.3\%$). After, the thickness required to reduce the incident radiation to half (Half Value Layer - HVL) and to tenth (Tenth Value Layer - TVL) of the initial value was calculate with the aid of EQ.1. From the results, even the Pb alloys being an interesting material because of their properties and specially price, it would be necessary almost the double thickness (in TVL values) to reduce the same level of radiation, comparing with the W alloys, as showed in TAB.1. The largest thickness value of each material was considered.

Comparing pure W with two of its alloys (W-Ni-Cu alloy and W-Ni-Fe alloy), the thickness variation required for γ radiation is not significant, when applied for the same shielding ability. Then, any of them would serve the purpose of this work.

Table 1: Thickness required to reduce the incident radiation to half and to tenth of the initial value from some of the selected materials.

Material	Density (g/cm ³)	Photon Energy (keV)	μ (cm ² /g)	HVL (cm)	TVL (cm)
Pb	11.3	777.921	0.0865	0.71	2.35
Pb-Sb alloy	11.2	140.511	0.1132	0.63	2.09
W	19.3	777.921	0.0788	0.45	1.51
W-Ni-Cu alloy	17.9	777.921	0.0783	0.49	1.64
W-Ni-Fe alloy	18.4	777.921	0.0785	0.48	1.59

4.3 Thermal and Impact Protection

Considering the design requirements for the thermal and impact protection part of the packaging, the materials selection was performed as flowchart below (FIG.5).

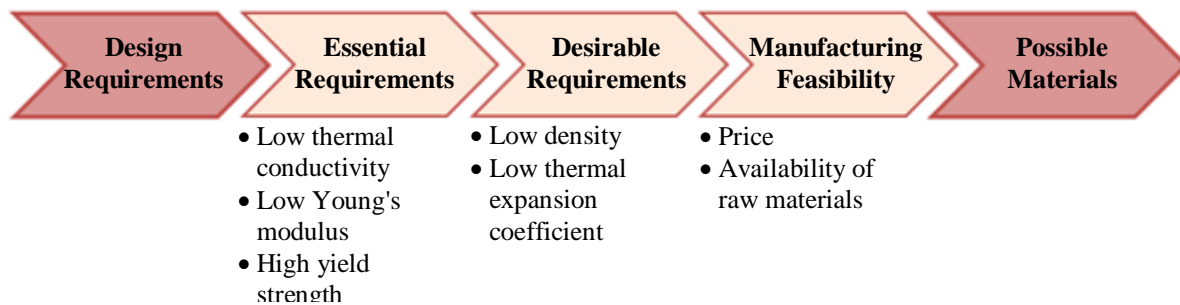


Figure 5: Materials selection flowchart with the requirements for the thermal and impact protection part.

The first property to be evaluated was the thermal conductivity. Using data obtained from the RAMPAC list, among the materials used for the thermal and impact protection part, steels are the materials that presents the highest value of thermal conductivity [10], so, the graphic was constructed using density relative by fixing its value at 55 W/m.°C as upper limit.

The next step was analyzing the resilience property, once the thermal protection must to stand the tests established in *CNEN* regulation, like drop tests for example [6]. The CES Edupack software do not provide this parameter, so the Young's modulus (upper limit fixed in 12.1 GPa) and the yield strength (lower limit fixed in 9.44 MPa) was used instead, because they have relation with the resilience modulus [13].

Once the radiation is shielded, it is interesting that the material presents a low density in order to do not increase the final weight of the packaging. Then, the maximum value set for density

was 1140 kg/m³.

In order to ensure consistency between the shielding material with the thermal and impact protection part, the thermal expansion coefficient of Pb was used (upper limit fixed at 29.2 μ strain/^oC).

Finally, the density related to price graphic was analyzed (FIG.6), with maximum price fixed at 6.54 BRL/kg.

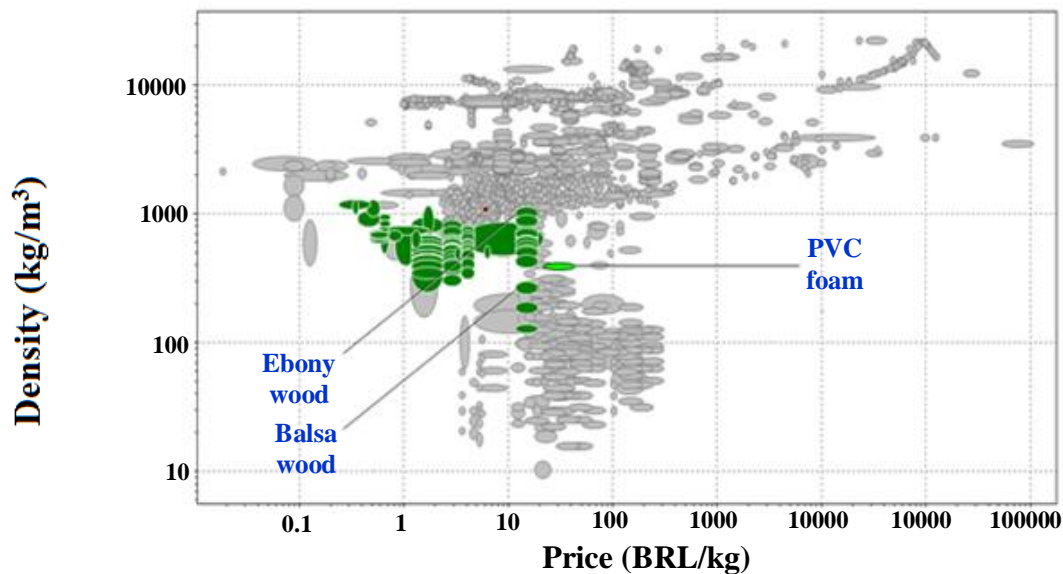


Figure 6: Density relative to price, presenting some of the selected materials that can be used for thermal and impact protection part of the packaging.

The last requirement to be discussed is the availability of raw materials in the country. From the results, it was verified that the woods figured as the better materials for thermal and impact purpose, and the species found in Brazil were: *Cariniana spp.*, *Juglans spp.*, *Ceiba pentandra*, *Hura crepitans*, *Terminalia amazonia*, *Cocos nucifera*, *Araucaria augustifolia*, *Aspidosperma spp.*, *Pinus spp.*, *Sequoia sempervirens*, *Cedrela spp.* and *Cedrelinga catrenaeformis*.

Beyond the woods, agglomerate and plywood materials were included in the possible materials, presenting advantages with respect to price and ease of forming.

4.4 External Recipient

Considering the design requirements for the external recipient part of the packaging, the materials selection was performed as flowchart below (FIG.7).

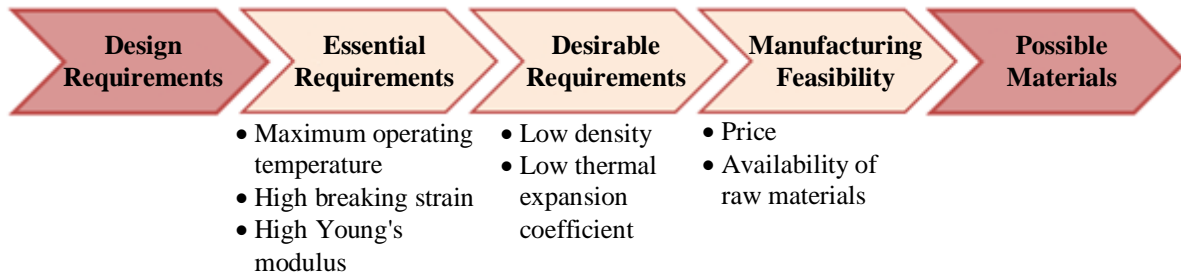


Figure 7: Materials selection flowchart with the requirements for the external recipient part.

One of the tests established in *CNEN* regulation is the thermal test, which consists in fully enclose the sample in a fire presenting average flame temperature of 800 °C, for Type B packages [6]. Then, the maximum operating temperature was set at 800 °C.

The second refinement considered the breaking strain and the Young's modulus by setting them at 345 MPa, for the breaking strain, and at 189 GPa, for Young's modulus (minimum values accepted).

Another tests established in *CNEN* regulation are drop tests [6]. Considering that normally ceramic materials present brittle fracture, they were excluded from selection.

As discussed for the thermal and impact protection, it is interesting that the material used in the external recipient part presents a low density. So, the upper limit of density was fixed at 8100 kg/m³. About the thermal expansion coefficient requirement, this property did not change the results and the graphic was the same (FIG.8).

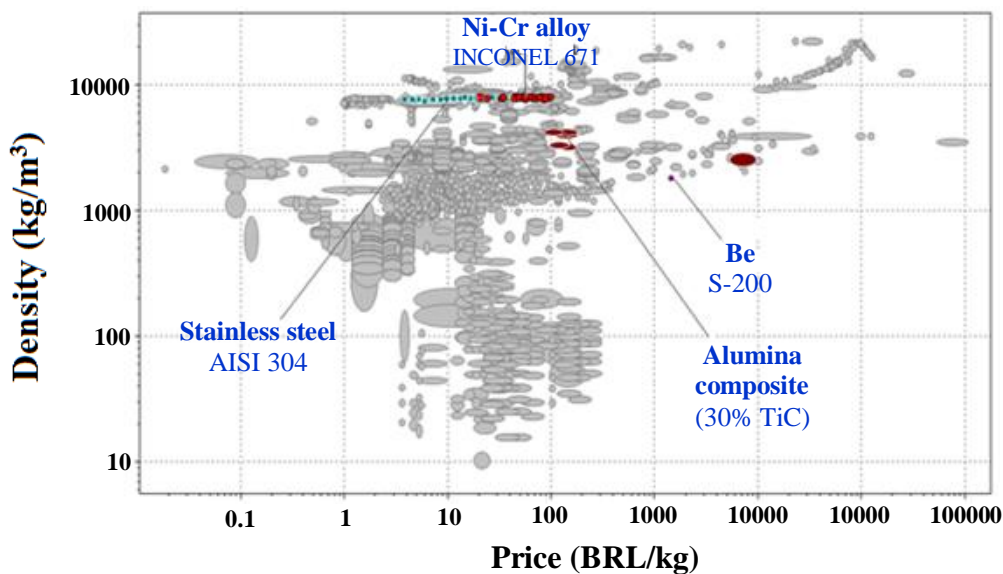


Figure 8: Density relative to price, presenting some of the selected materials that can be used for external recipient part of the packaging.

Analyzing the graphic in FIG.8, presenting density related to price, materials like beryllium (Be) and alumina composites were excluded, because of their high value of price, which would increase the total cost of the packaging.

Therefore, for the external recipient, the selected materials were in the family of steels and nickel (Ni) alloys. However, the stainless steels present a significantly reduced price when compared to Ni alloys, showing advantage in their choice.

4.4 Internal Recipient

Finally, considering the design requirements for the internal recipient part of the packaging, the materials selection was performed as flowchart below (FIG.9).

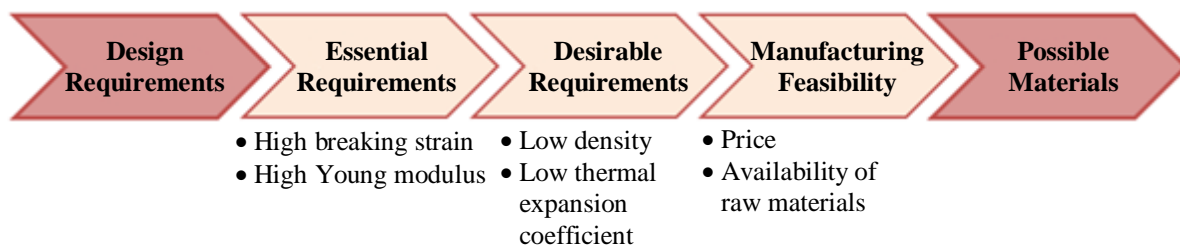


Figure 9: Materials selection flowchart with the requirements for the inner recipient part.

Once the design requirements of the internal recipient part consists of almost the same requirements of the external recipient, the results obtained for this part was equal to that one.

3. CONCLUSIONS

This chapter presents the conclusions made after the analysis of obtained results.

The essential purpose of a packaging is the radiation shielding; then, it was the first part analyzed. From materials selection, those which met the design requirements were Pb alloys and W alloys; however, the W alloys are the most suitable for presenting a thickness approximately two times lower than that found for Pb alloys.

Regarding the thermal and impact protection part, the woods presented the best results. Beyond them, the agglomerates and plywoods are included and present advantages with respect to price and ease of forming.

Related to the external recipient part, the steels and Ni alloys were indicated, most preferably steels because of their lower cost when compared to Ni alloys.

Finally, once the requirements of the internal recipient met the ones for external recipients, the results obtained for both were equal.

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