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Evaluation of an alternative shielding materials for F-127 transport package

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ABSTRACT

Lead is used as radiation shielding material for the Nordion's F-127 source shipping container is used for transport and storage of the GammaBeam –127's cobalt-60 source of the Nuclear Technology Development Center (CDTN) located in Belo Horizonte, Brazil. As an alternative, Th, Tl and WC have been evaluated as radiation shielding material. The goal is to check their behavior regarding shielding and dosing. Monte Carlo MCNPX code is used for the simulations. In the MCNPX calculation was used one cylinder as exclusion surface instead one sphere. Validation of MCNPX gamma doses calculations was carried out through comparison with experimental measurements. The results show that tungsten carbide WC is better shielding material for γ-ray than lead shielding.

1. Introduction

Traditionally, lead has been used as radiation shielding materials, collimator, equipment and sources containers in the nuclear industry to help protect against gamma radiation exposure, dues to its high density, high atomic number and very cheap. Nevertheless, other materials such as tungsten, copper, bismuth, steel etc. are been used as shielding material (Abd El-Latif and Saeid Khalifa, 2010; McAlister, 2013; Soylu et al., 2015).

Recently, researchers in different countries have been investigated new shielding materials against gamma radiation, for example: amethyst investigated by Korkut et al. (2011); pure tungsten, tungsten carbide-cobalt (WC-Co) materials studied by Buyuk and Tugrul (2014); Gd_2O_3 according to Kaewjanga et al. (2014); nanocomposites types material investigated by Atta et al. (2015), and lead-free composite material tungsten carbide (50%, 60%, and 70%) (Soylu et al., 2015).

The goal of this work is to evaluate three types of material, Th, Tl and WC that can be used as good γ-shielding materials for F-127 transport packing model. The lead will be used for the comparison.

The Nordion's F-127 source shipping container, made of lead, is used for transport and storage of the GammaBeam –127's (GB-127) cobalt-60 source installed at the Nuclear Technology Development Center (CDTN).

The exchange of the cobalt-60 source is performed periodically and taking into account the optimization of the recharge costs and their benefits to the facility yield production. For the CDTN Gamma Irradiation Facility, the optimal source exchange period is 5 years. Once defined the optimized exchange frequency, the new source is ordered by the manufacturer and send to the facility in its B(U) packages containing radioactive material though the ground and maritime transport until the facility. The exchange of sources and its initial operational tests are performed by specialized technicians from the manufacturer under the supervision of the facility radiation safety service.

The lead F-127 container is classified as type IAEA B(U) (IAEA, 2012). As such, the equivalent dose rates on the surface of the shipping container should comply with the regulatory requirements of the Brazilian Nuclear Energy Commission (CNEN), and the IAEA Regulations for the Safe Transport of Radioactive Material (CNEN, 1988) and IAEA (2007). As stated in those regulations, the maximum dose rate at any point on the external surface of the shipping container may not exceed 2.0 mSv/h.

2. Materials and methods

To evaluate the radiation shielding performance of these materials, the GB-127 ^{60}Co were utilized. The photon energy levels for these sources were of 1.1732 MeV and 1.3325 MeV, respectively. The source of the gamma irradiation facility has a maximum activity of 60,000 Ci, which is composed of 16 double encapsulated radioactive pencils placed in a rack. The characterization of the irradiator source was made in the previous work (Gual et al., 2017). Fig. 1 shows a picture of the lead F-127 transport-packing model.

The lead F-127 was modeled with the Monte Carlo code MCNPX v. 2.6.0 (Hendricks et al., 2008). This model includes geometric and structural details of the F-127 container. Table 1 summarizes the main design parameters of the lead F-127, which were used in the MCNPX modeling.

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Fig. 1. Lead F-127 transport-packing model.

Table 1
Main design parameters of the lead F-127 used on the MCNPX modeling.

Parameters	
Height (cm)	102.2
Outer diameter (cm)	62.2
Cavity (cm)	16.3 × 34.8
Thickness of Ni (mm)	5.0
Material	Pb
Density (g/cm^3)	11.34
Height of top (cm)	22
Outer sheet diameter (cm)	80
Material	Steel
Density (g/cm^3)	7.87
Height of handle (cm)	6.8

The studied materials were selected based on the high atomic mass number and high density. Table 2 summarizes the element composition of materials which were used in the MCNPX modeling.

The MCNP tally option utilized for this problem was the energy fluence *F5 (in MeV/cm^2) with DE and DF cards to introducing the (μ (E)/ρ) tabulated in Attix (2004), and the multiplication card FM (1.6021 × 10^{-07}) to convert (MeV/g) to mGy. It is used the equivalence that 1 J/kg (physical unit) of gamma absorbed dose corresponds to 1 Sv of equivalent dose. Dose rate was estimated at 10 points along the maze. These points were located at 162.5 cm above the floor level and 50 cm from the shielding wall.

MCNPX was run in photon and electron mode (mode p and e) and to improve the efficiency of electron and photon transport, two cards (PHYS:P and PHYS:E) are utilized. A detailed photon physics treatment, including photoelectric effect with fluorescence production, incoherent and coherent scattering and pair production, has been considered in the energy range between 0.001 and 2.6 MeV with “PHYS:P” card. For electron transport, MCNP addresses the sampling of bremsstrahlung photons at each electron sub step. The “PHYS:E” card is utilized in MCNP for biasing some physical parameters such as the production of secondary electrons by photons, coherent scattering, bremsstrahlung angular distribution and production of characteristic x-rays.

The isodose curves will be obtained from the data given by the “TMESH” mesh type 3 tally with the “total” option that superimposes a rectangular mesh grid over the simulation geometry. The “total” allows scoring the equivalent to an F6 heating tally for gammas rays in units of MeV/cm^3 normalized by source particle. A rectangular mesh is defined with 1 mm resolution. For visualizations, the software method MATLAB will be selected. In order to accelerate the calculations, the MCNP code has been parallelized in Intel's Core i7 CPU with 3.4 GHz and 8 GB RAM, using the MPI multiprocessing parallel protocol, in this case with 8 processors. This way, the problem speedup is achieved. It was taking into consideration the worst situation regarding maximum dose rate, the door has the full opening hole in the maze entrance with any additional shielding.

Table 2
Element compositions of shielding materials used in the MCNPX modeling.

Material	Composition (% w)	Density (g/cm^3)
Pb	100	11.34
Th	100	11.724
Tl	100	11.86
WC	Tungsten (93), Carbon (6.1), Iron (0.03)	15.63

2.1. Shielding material properties

Natural lead has four stable isotopes: Pb-204 (1.4%), Pb-206 (24.1%), Pb-207 (22.1%), and Pb-208 (52.4%). Lead is the most used material in nuclear technology as shielding material and collimator (Lansdown and Yule, 1986).

Natural tungsten consists of five isotopes whose half-lives are so long that they can be considered stable: W-180 (0.12%), W-182 (26.50%), W-183 (14.31%), W-184 (30.64%), and W-186 (28.43%). Tungsten is used as a target in a linear accelerator to produce X-rays.

Natural thorium has 6 naturally occurring isotopes: Th-232 (99.99%) and Th-230 (0.02%) are the most abundant and/or stable. Th-232 has a half-life of 14.05 billion years and Th-230 has a half-life of 75,400 years. Thorium is about as common as lead and is typically used as nuclear fuel and welding electrodes.

Thallium has 37 isotopes with atomic masses that range from 176 to 212. Tl-203 (29.5%) and Tl-205 (70.5%) are the only stable isotopes. Approximately 60–70% of thallium production is used in the electronics industry, and the remainder is used in the pharmaceutical industry and in glass manufacturing. Tl-205 is also used in nuclear magnetic resonance research.

Mechanical properties of the different materials studied including tensile strength, Poisson's ratio, modulus of elasticity, shear modulus and Brinell hardness are shown in Table 3 (Abd El-Latif and Saeid, 2010; McAlister, 2013; Soylu et al., 2015).

Lead has easiness of handling and machining as well as an affordable cost and it is often referred as a sort of ‘standard’ to which compare other shielding materials' effectiveness. The study and comparison of different mechanical properties and machinability of the studied materials can be carried out other paper.

The purpose of our study was to evaluate effectiveness respect the radiation quality parameters for decreasing operator's radiation dose without including a discussion of the structural design on the shielding performance. Other factors which were not analyzed in the study are the material and machining cost and geometrical shape.

Comparison of mass attenuation coefficient μ/ρ as a function of gamma radiation energy for proposed shielding materials are illustrated in Fig. 2.

As expected, the thorium, with the high atomic mass number and high density, has superior attenuation than lead. It is observed that the

Table 3
Mechanical properties of the different materials studied.

Mechanical properties	Lead	Thorium	Thallium	Tungsten
Tensile strength (MPa)	18.00	Not available	Not available	1725
Poisson's ratio	0.42	0.27	0.45	0.28
Modulus of elasticity (GPa)	14.00	79.00	8.00	400
Shear modulus (GPa)	4.90	31.00	2.80	156
Brinell Hardness, HB (MPa)	10	38–70	2	320–450

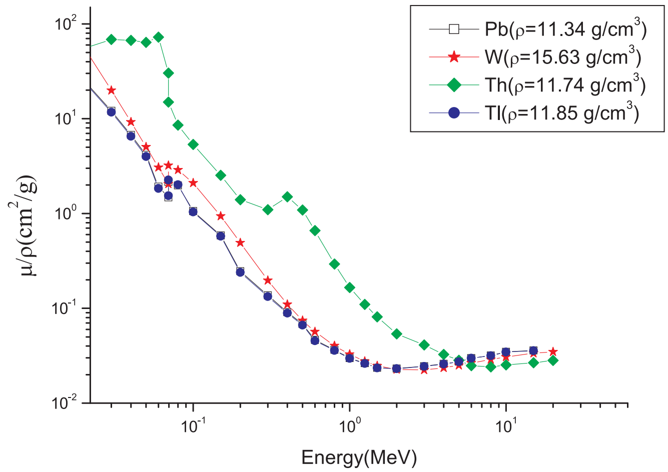

Fig. 2. Photon mass attenuation coefficient μ/ρ versus gamma radiation energy for proposed shielding materials. The units are cm^2/g.

photon mass attenuation coefficient μ/ρ for W and Pb are identical.

Cross section data for (G, X) and (G, XN) reactions of proposed shielding materials versus gamma radiation energy is plotted in Figs. 3 and 4, respectively.

All data to make the graphs are obtained from libraries available in the NIST (2016), National Institute of Standards and Technology, and IAEA (2016) Nuclear Data Services of International Agency Energy Atomic.

The (G, X) reaction sum of all reactions not given explicitly in another reaction. It is observed, that for (G, X) reactions on Pb-208, Th-232, Tl-203 and W-184 there are systematically differences between some libraries. The (G, X) reactions appear below 1 MeV.

There are good agreements with photonuclear reactions (G, N) between libraries, showing that neutrons appear only for photons up to 5 MeV. For this reason, it will not consider neutrons productions, but other reactions are necessary accompanying. Table 4 shows the threshold energy of incident gamma radiation for production of (G, N) and (G, X) reactions to proposed shielding materials versus gamma radiation. All data of the table were obtained from libraries available on the Internet (NNDC, 2016).

It is observed that the lead emits secondary x-rays, at around 78–80 keV when gamma strike the shield (γ, x). For this, the lead F-127 container has one steel layer around there.

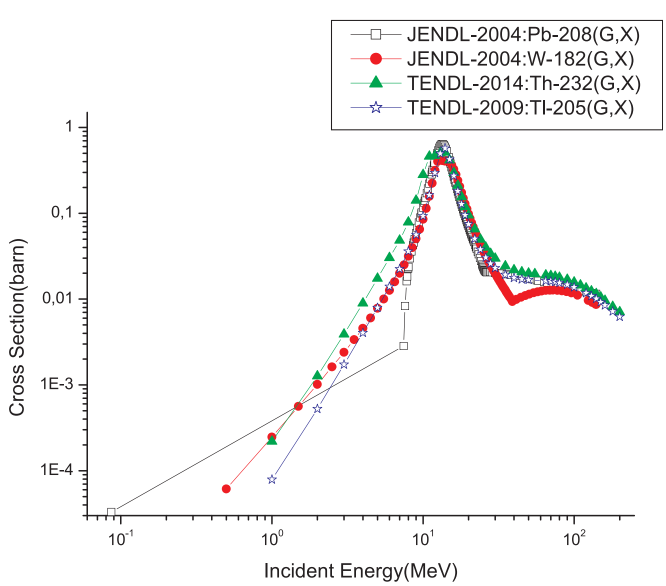

Fig. 3. Cross section data for (G, X) reaction of proposed shielding materials versus gamma radiation.

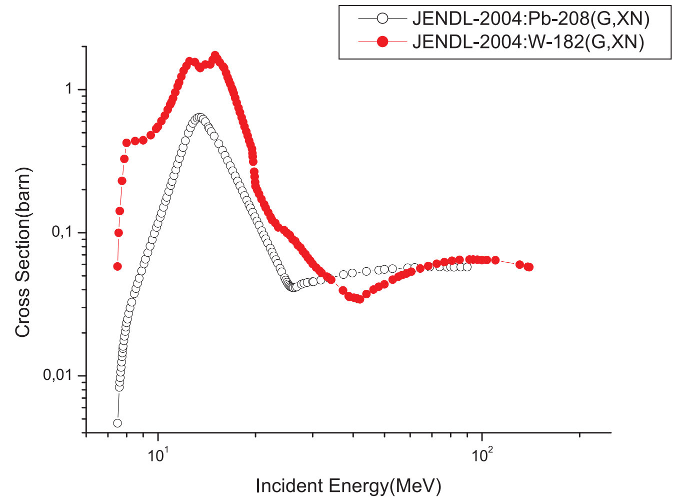

Fig. 4. Cross section data for (G, XN) reaction of proposed shielding materials versus gamma radiation.

Table 4
Threshold energy of incident gamma radiation for the reaction (G, N) and (G, X) of proposed shielding materials versus gamma radiation.

Reactions	Threshold energy (MeV)
^{208}Pb(G,N)^{207}Pb	7.368
^{208}Pb(G, X)	0.086 (from JENDL-2004
^{232}Th(G, N)^{231}Th	6.440
^{232}Th(G, X)	1.000 (from TENDL-2014)
^{203}Tl(G, N) ^{201}Tl	7.845
^{203}Tl(G, X)	1.000 (from TENDL-2009)
^{184}W(G, N) ^{183}W	7.411
^{184}W(G, X)	0.500 (from JENDL-2004)

The tungsten emits secondary x-rays, at around 0.5 MeV when gamma strikes the shield (γ, x).

Thorium does not have neutrons from (γ, x) reactions, but has x-rays to 1 MeV energy. It can be overcome by lining the shield with layers of lower Z material on the inside as steel.

Thallium emits secondary x-rays around 1 MeV when gamma strikes the shield (γ, x). It can be overcome by lining the shield with layers of lower Z material on the inside as steel.

In all materials, the neutrons are produced through photonuclear reaction (γ, n) if the energy of the incident gamma radiation is high than 5 MeV. So, it is not a problem for the lead replacement as material shielding in F-127 container. Furthermore, the proposed materials should satisfy radiation shielding needs and it is necessary taking into account the competitive price compared with lead.

3. Validation of model and numerical method

Fig. 5 illustrates the XY MCNPX view (in the left part) and the XZ view (in the right part) of the F-127 model. A visualization of the MCNP model was reproduced with Vised 22S (Schwarz et al., 2008). The normalized isodose curve representation was made using MATLAB.

The sources rack consists of 24 holes with 16 double encapsulated radioactive cylindrical pencils, model C-198. The total initial activity of the sources was 28,882 Ci (14 Feb. 2007).

For validation of MCNPX model, the gamma doses rate results are compared with the measured dose from the source transfer record of MDS Nordion-supplied on the surface of transport package F-127 located at center on 2007 February 14th. The comparison is presented in Table 5.

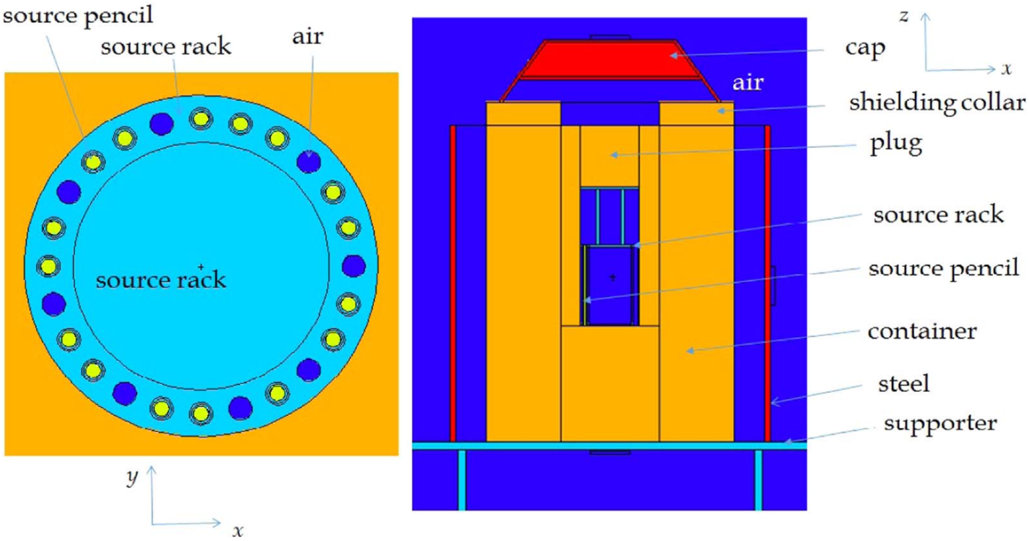

Fig. 5. F-127 transport packing MCNPX model.

Table 5
Comparison between measured and calculated gamma doses rate at the surface of transport package F-127 located at center with the *F5 tally.

Tally	MCNPX D (mSv/h)	MDS Nordion D(mSv/h)	% Difference
Top	1.196	Not reported	–
Side	0.717	1	28.33
Bottom	0.005	Not reported	–

The difference with respect to experimental result is less than 30%, but not exceeds the limit established for the safe transport of radioactive material that is equal to 2.0 mSv/h. The uncertainties in the geometrical and physical details of the F-127 container are factors that causing this difference.

In order to obtain a statistical error lower than 5% about 130 × 10^6 photon histories were calculated. The CPU processing time was approximately 1.10 days with MPI multiprocessing parallel protocol with 8 processors. In order to reduce the variance and speed up the calculations, it was employed the cell importance technique.

All simulations were obtained using the MCNPX *F5 tallies and 130 × 10^6 particles histories.

The numerical method has been validated because of the tally did pass all 10 statistical checks and the recommended relative errors have been reached.

Fig. 6 illustrate the part of the F-127 container that was changed for studying the shielding performance of the different materials and the comparison of the densities of them with respect to the Pb.

4. Results

Fig. 7 shows the normalized isodose distribution in the lead F-127 transport packing. The dose rate at the side is higher than at the top and the side of the lead F-127 transport package due to the extra shielding in the pathway of the radiation.

The isodose curves were obtained for all the materials, but since the densities are very similar, it was not possible show much differences between them.

Table 6 shows the comparisons between the uses of the different surface of exclusion. It is observed that with the cylinder as exclusion surface the computation time is lower than if is used sphere.

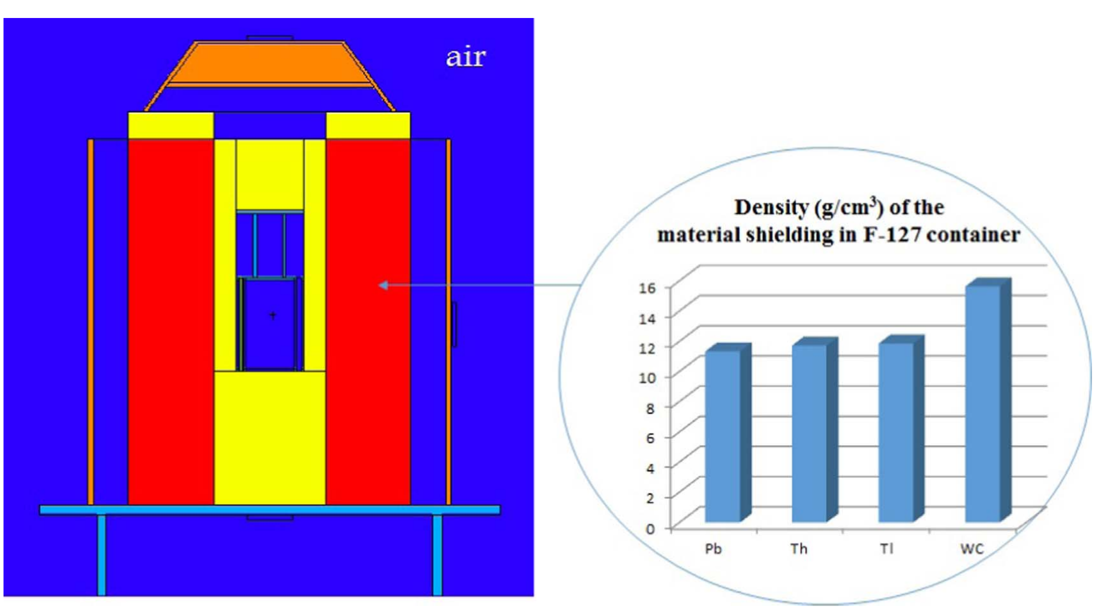

Fig. 6. Part of the F-127 container materials that were changed and the comparison of the densities of them with respect to the Pb.

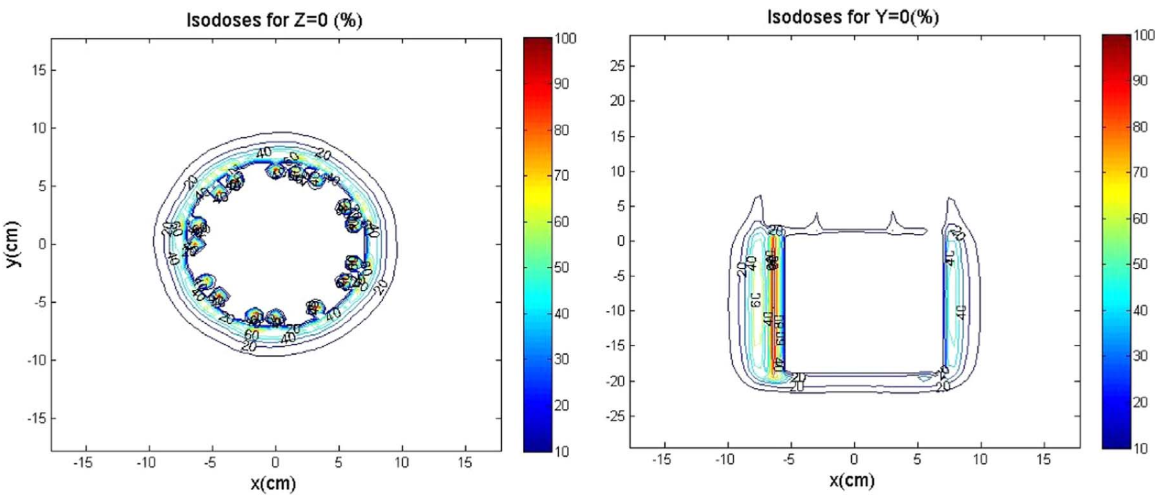

Fig. 7. Normalized isodose curve representation of XY and XZ view of the gamma dose rate distribution using MATLAB in the lead F-127 transport packing.

Table 6
Comparisons between the uses of the different surface of exclusion in MCNPX calculation.

Exclusion surface	Sphere	Cylinder
t (day)	8.69	1.10

Table 7
Comparison of dose rate (in mSv/h) obtained with MCNPX code for the studied shielding material for γ-ray in F-127 transport packing.

	MCNPX D (mSv/h)			
Position	Pb	Th	Tl	WC
Top	1.196	1.283	0.962	1.993
Side	0.717	0.126	0.350	0.014
Bottom	0.005	0.004	0.004	0.004

Table 7 summarizes the dose rate comparisons considering the proposed shielding materials.

The results demonstrate that the dose equivalent rate on the surface of the F-127 source shipping container fulfills the radiation protection requirements of being less than 2.0 mSv/h for all proposed shielding materials. All materials are good as shielding materials. As shown, the tested material's shielding efficiency is higher than lead. The WC is the better. In future work, should calculate the doses produced for other reactions that are necessary accompanying not studied in this work.

5. Conclusions

The obtained dose equivalent rate on the surface of the F-127 source shipping container is below the established dose limits by the national Brazilian regulations and the international recommendations for Safe Transport of Radioactive Material for all proposed shielding materials. Those results are promising for application of Th, Tl and WC as a shielding material for γ-ray in F-127 transport packing model without compromising on radiation protection.

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