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NEUTRONIC AND NUCLEAR CALCULATIONS FOR THE Pb/Au TARGET OPTION FOR ESS

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ABSTRACT

Lead-gold eutectic (85 w% Pb, 15 w% Au) is considered as a target material for the European Spallation Source (ESS). Neutronic and nuclear calculations have been performed using MCNPX 2.5.0. The neutron production was compared with two other proposed materials, mercury and Pb/Bi. Besides neutron production, particular care was devoted to investigate potential problems using this material, such as the possible decrease of the neutronic performance, the increase of the activity and decay heat, and the burn-up of the gold, with consequent mercury production, due to a long-term irradiation of many years.

The main conclusions are the following: there is very little difference in the neutron production of the three target materials, and the neutron fluxes inside the moderators are very similar: using Pb/Au or Hg, the integrated flux below 1 eV in an H_2 moderator is 5-6 % lower than using Pb/Bi. In the case of 40 years irradiation the activity from the Pb/Au target is significantly greater than for the Pb/Bi one (nearly 3 times higher at the end of irradiation) for about 10 days, then it becomes lower. The decay heat is also about 3 times larger for the first 10 days after shutdown and it is of 45 kW at the end of irradiation. This could be a problem for cooling in case of draining of the target material and it should be taken into account in the design phase. We also calculated the burn-up of the gold: after 40 years of irradiation, the amount of gold in the target is decreased to 97 % of the initial amount, while about 3% is converted to mercury. The impact on the possible change in the properties of the eutectic.

1. Introduction

Lead-gold eutectic (LGE) has been recently proposed as a target material for ESS [1]. LGE is a relatively unknown material, which appears to combine the advantages of mercury and lead-bismuth eutectic (LBE), without having their pitfalls, in particular safety issues related to Hg, and high polonium production in LBE. It is important to evaluate how valid is this option from the neutronic and nuclear points of view. The main advantage of the LGE target with respect to an LBE target is the fact that there is no production of Po isotopes, and in particular ^{210}Po, from proton and neutron reactions on the bismuth. There is still very limited production from alpha reactions on the lead, but this is orders of magnitude lower than on bismuth. However, there are some potential drawbacks, due to the fact that gold has a high thermal and epithermal neutron capture cross section.

The first possible problem is the influence on the neutron yield of the presence of the gold. ^{197}Au has a thermal neutron capture cross section of 98.7 b, and a resonance integral cross section of 1551 b, mainly due to the resonance at 4.9 eV. In comparison, also Hg isotopes have relatively large capture cross sections, especially ^{199}Hg (16.87 % of the natural composition) as shown in Fig. 1. It is therefore expected that there should be no negative effect from the neutron production point of view, but we need to verify the effect of an LGE target on the produced flux to the ESS users.

A second possible problem related to the neutron capture was suggested [2]. Following neutron capture from ^{197}Au, the ^{198}Au ground state decays with a half life of 2.7 days, with emission of γ-rays of several hundreds of keV (notably the 412 keV γ ray). This may have consequences on the decay heat in the target after irradiation, and in the high dose rate for up to 10 days after irradiation, which is expected to be very large compared to an LBE or mercury target.

A third possible problem is the burn-up of the gold following neutron capture, which, for the extended operation of ESS may be significant. This not only can lead to unacceptable losses of the gold, with consequent change of the chemical properties of the LGE, but may also give a high production of mercury, namely ^{198}Hg from the decay of ^{198}Au. At the operating temperature of the LGE, this would imply a possible safety problem in case this mercury is not retained in the target.

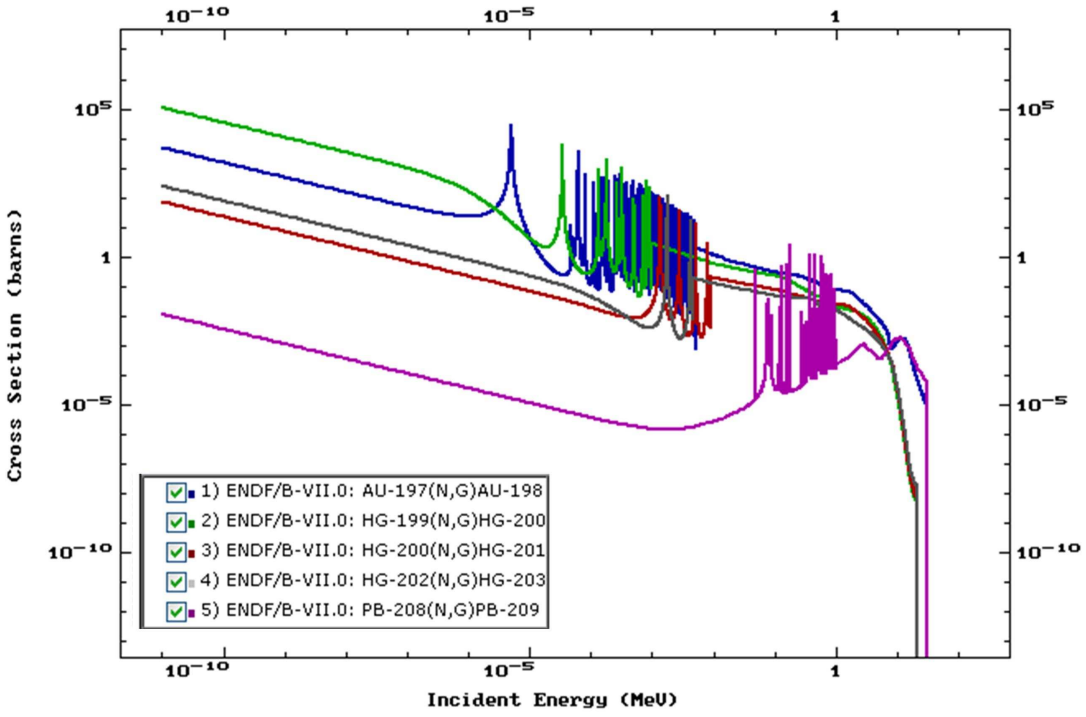

Figure 1. ENDF/B-VII neutron capture cross sections for ^{197}Au, ^{208}Pb and the three most abundant Hg isotopes (*A*=199, 200, 202).

2. Calculations

Calculations have been performed using MCNPX 2.5.0 and a model of the ESS target from C. Pohl was used [3]. The model of the target is represented in Fig. 2. The target volume in the model is of 37.6 liters, and the complete loop is not included. At present the reference volume for the liquid metal loop is of 1000 liters.

In our calculations the only change was for the target material, and calculations were performed using mercury, lead-bismuth (55.5 w% Bi, 44.5 w% Pb) or lead-gold (85 w% Pb, 15 w% Au). The densities for Hg, LBE and LGE were of 13.6 g/cm^3, 10.3 g/cm^3 and 12 g/cm^3, respectively. No impurities in the materials were considered. The energy of the incoming proton beam was of 1.334 GeV. Calculations were performed up to 40 years of

operation with the following irradiation conditions (see Table I): irradiation for 38 years with an average accelerator efficiency of 57% (equal to 5000 h of operation per year), followed by 2 years in which the beam is partially at full power, but with a shut down period in between. Although this irradiation table is still rather coarse, it should give the correct total charge on target, and give at the end of irradiation maximum values of quantities such as activation, dose rates and decay heat.

Table I. Irradiation parameters used in the calculations (proton beam energy 1.334 GeV).

Time	Proton current (mA)	Power (MW)
38 years	2.14	2.85
5000 hours	3.75	5
3760 hours	0	0
5000 hours	3.75	5

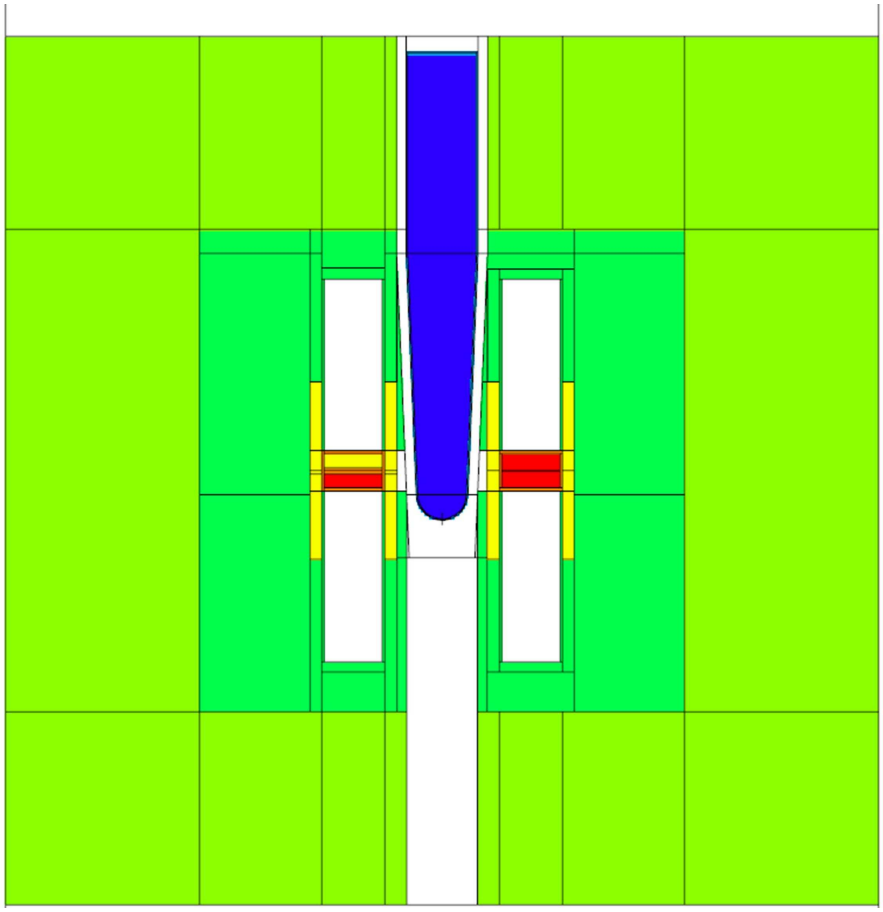

Figure 2. The ESS MCNPX model, after Ref. [3].

All calculations were performed using the default Bertini-Dresner option for spallation and evaporation models. For flux and activity calculations two different procedures were applied. For flux calculations the model of Fig. 2 was used, with the real target volume of 37.6 liters. Fluxes were calculated using f4 tallies in the target or

moderators. For activity calculations, the procedure was the following: *i*) the same model of Fig. 2 was used, but the volume of the target was set to 1000 liters. MCNPX was used to determine the production rates of the isotopes for proton reactions and for neutrons above 20 MeV. Moreover, the neutron flux in the target was calculated. *ii*) The MCNPX results were coupled with the FISPACT evolution code using the activation script from Ref. [4]. These production rates and neutron flux were then used by FISPACT, that calculates also the production for neutrons below 20 MeV. Then the calculation at any given irradiation or cooling time is performed. It is important to give as input the real volume of the loop, so that the correct evolution of the composition of the target is calculated by FISPACT.

2.1 Neutron spectra and neutron fluxes

We calculated the neutron fluxes inside the spallation target, for the three different materials used. Additionally, neutron spectra were calculated in the four moderators. Only absolute production has been examined, without any time dependent studies for a pulsed beam. Fig. 3 shows the neutron spectra inside the spallation target for three different materials. This figure shows the effect of a neutron absorbing material (gold in the LGE, or mercury) on the neutron spectrum inside the target. In particular, in the case of the LGE the thermal peak, which is quite strong with the LBE target, is almost completely absorbed. Additionally, the epithermal component up to the 5 eV resonance is strongly reduced.

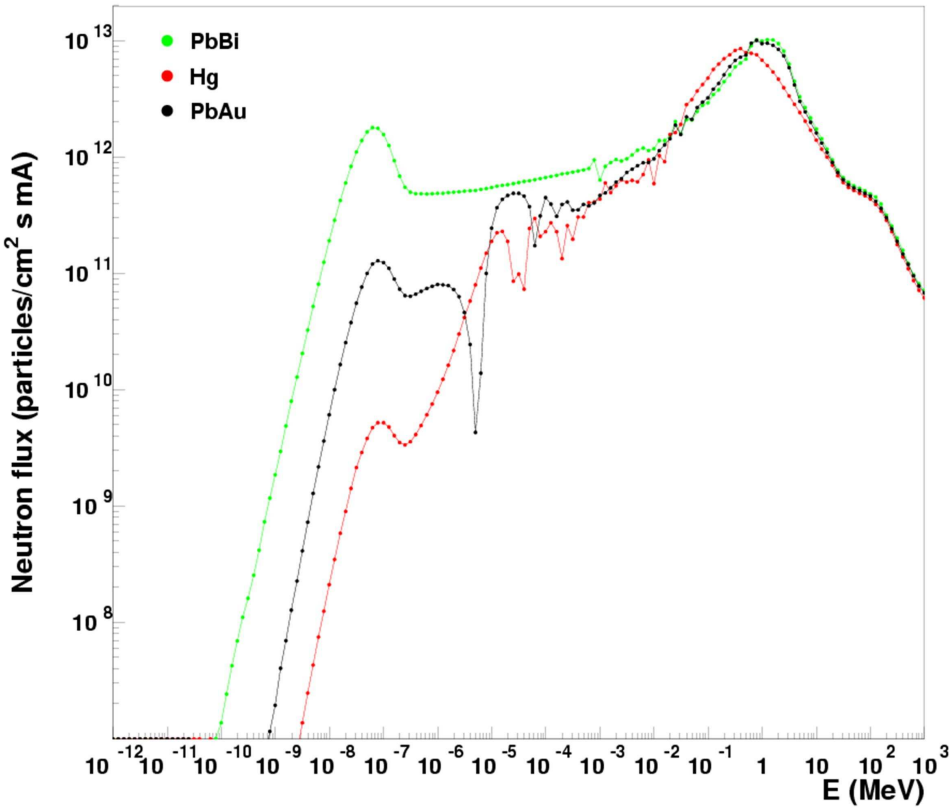

Figure 3. Neutron spectra inside the spallation target for three different materials.

The fluxes in the four moderators were also calculated. Fig. 4 (left) shows the spectra in the four moderators with the LGE target. In Fig. 4 (right) the spectra in one of the liquid hydrogen moderators, calculated with the three different target materials, is shown. One

can see that there is some effect, but minimal, on the peak at low energies. Integral values of the fluxes below 1 eV are indicated in Table II. From the point of view of the neutron flux in the moderators, there are no drawbacks in using LGE instead of LBE. In the third column of Table II the integral flux averaged over a surface of 5 m radius surrounding the target, in a calculations where all the structure surrounding the target has been put to void, is indicated. These numbers give the absolute neutron production of the targets without moderators or structure. As can be seen, neutron production is very similar.

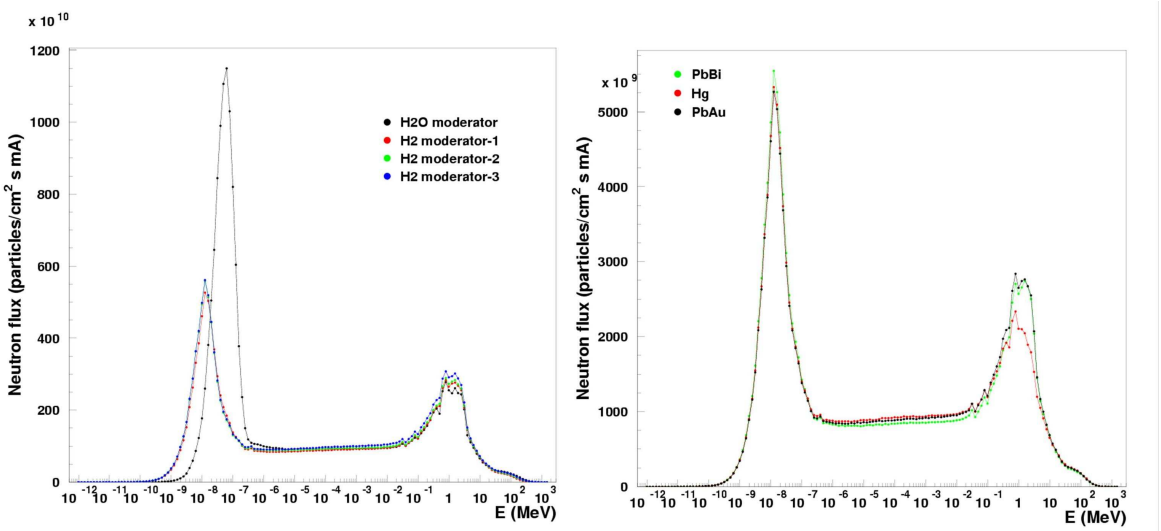

Figure 4. *Left*: Neutron spectra in four moderators with the LGE target. *Right*: comparison between the neutron spectra in one liquid hydrogen moderator with three different target materials.

Table II. Integrated neutron fluxes below 1 eV in an H_2 moderator for three different target materials. In the last column, Φ_{TOT} is the total flux exiting the target, in a calculation where all the materials surrounding the target have been put to void.

Material	Φ (n/cm^2/s/mA) E< 1 eV moderator	Φ_{TOT} (n/cm^2/s/mA)[1]
LGE	6.51 10^{13}	6.20 10^{10}
LBE	6.93 10^{13}	5.99 10^{10}
Hg	6.56 10^{13}	6.06 10^{10}

[1]Integral flux averaged over a spherical surface with R=500 cm surrounding the target.

2.2 Activation and burn-up effects

In Fig. 5 the buildup and decay of the total activity in the spallation target for three different target materials is shown. The peak in the activity observed in the last point of the buildup curve is due to the chosen irradiation history, which gives higher current for the last 5000 h of irradiation.

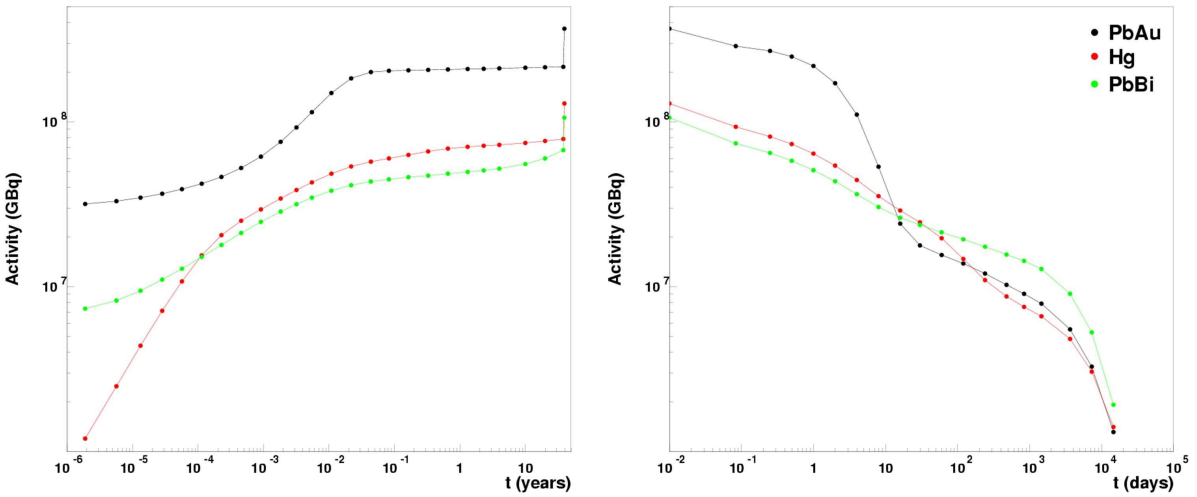

Figure 5. Buildup and decay of the total activity in the spallation target for three different materials.

We see from the above figures that the activity in the LGE target is higher during irradiation: at the end of irradiation the activity of the LGE is about 3 times the one of the LBE target, and remains higher for the first 10 days of cooling, because of the decay of the ^{198}Au isotope ($t_{1/2}$=2.7 d). After 10 days the total activity (as well as the decay heat and the dose rate, as shown below) is lower than the LBE, and between 10 and 100 days of cooling is also lower than mercury.

In Fig. 6 the LGE target composition at the beginning of irradiation and at four times, after 1, 10 and 40 years of irradiation is plotted. The amount of gold decreases, in favor of creation of mercury. A close up in the high mass region is also shown. Results are summarized in Table III and compared with LBE and Hg targets. We note that with the LGE target, after 40 years of irradiation, 4 liters of mercury are created in the LGE target, coming mainly from decay of gold (50 kg of gold are burned, about 3 % of the initial amount). Fortunately, most of the created Hg is stable ^{198}Hg.

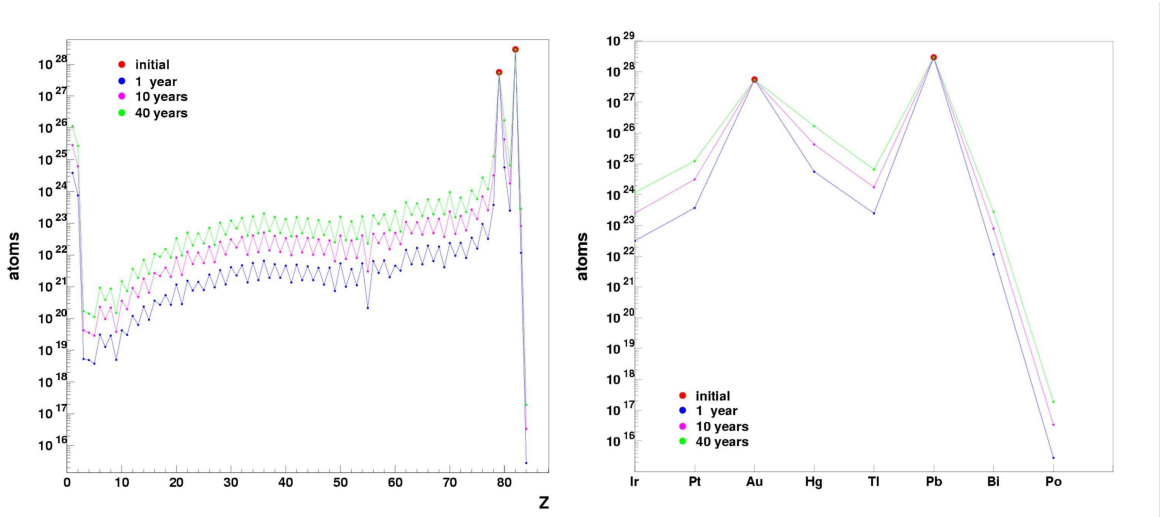

Figure 6. *Left*: LGE target composition (number of atoms per element) at different irradiation times. *Right*: composition for the isotopes with higher mass.

Table III. Mass (grams) for the heaviest elements at the beginning of irradiation and after 40 years of irradiation for the three different targets.

Target	Au	Hg	Tl	Pb	Bi	Po
			INITIAL			
LGE	$1.83\ 10^{6}$			$1.02\ 10^{7}$		
LBE				$4.58\ 10^{6}$	$5.72\ 10^{6}$	
Hg		$1.35\ 10^{7}$				
			40 years			
LGE	$1.78\ 10^{6}$	$5.62\ 10^{4}$	$2.25\ 10^{3}$	$1.01\ 10^{7}$	$9.80\ 10^{1}$	$6.58\ 10^{-5}$
LBE	$5.36\ 10^{2}$	$2.80\ 10^{3}$	$1.62\ 10^{3}$	$4.60\ 10^{6}$	$5.64\ 10^{6}$	$1.07\ 10^{1}$
Hg	$3.74\ 10^{3}$	$1.35\ 10^{7}$	$3.53\ 10^{3}$	8.7	0	0

2.3 Decay heat and dose rate

The decay heat curve, during and after irradiation, is shown in Fig. 7. As expected, the heat is much higher in the case of the LGE. The dose rate has the same behaviour (Fig. 8). A possible operation plan is irradiation for 45 days, followed by shutdown for liquid drainage from the circuit and target exchange [5]. Since the time for target exchange is estimated to be 1-2 weeks, in principle it is convenient to have a target material with low specific after-heat. It needs to be seen if the high specific decay heat of LGE would be a problem.

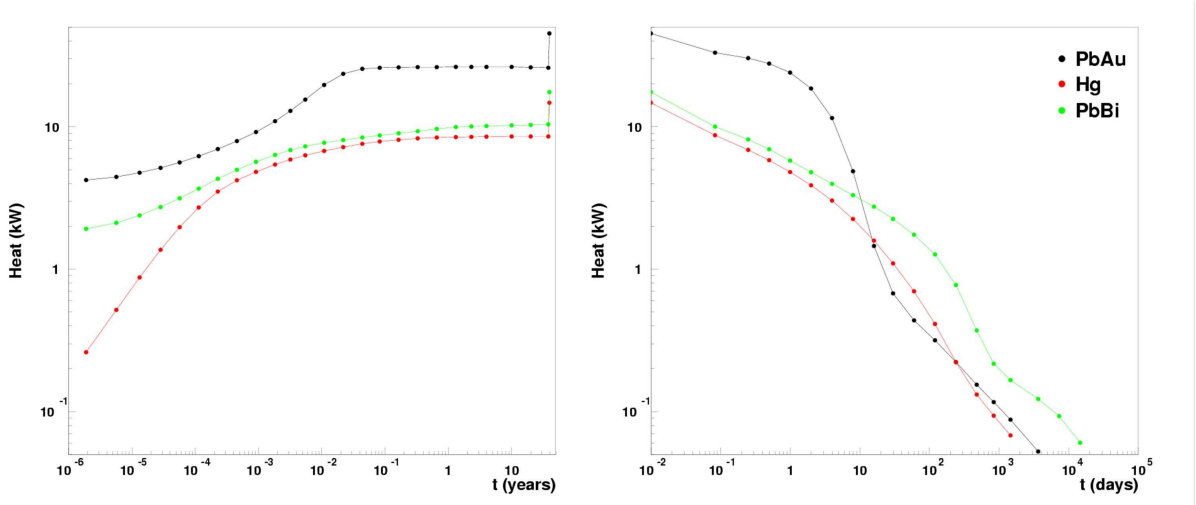

Figure 7. Decay heat in the 1000-liters liquid metal loop for three different materials during irradiation (left) and after irradiation (right).

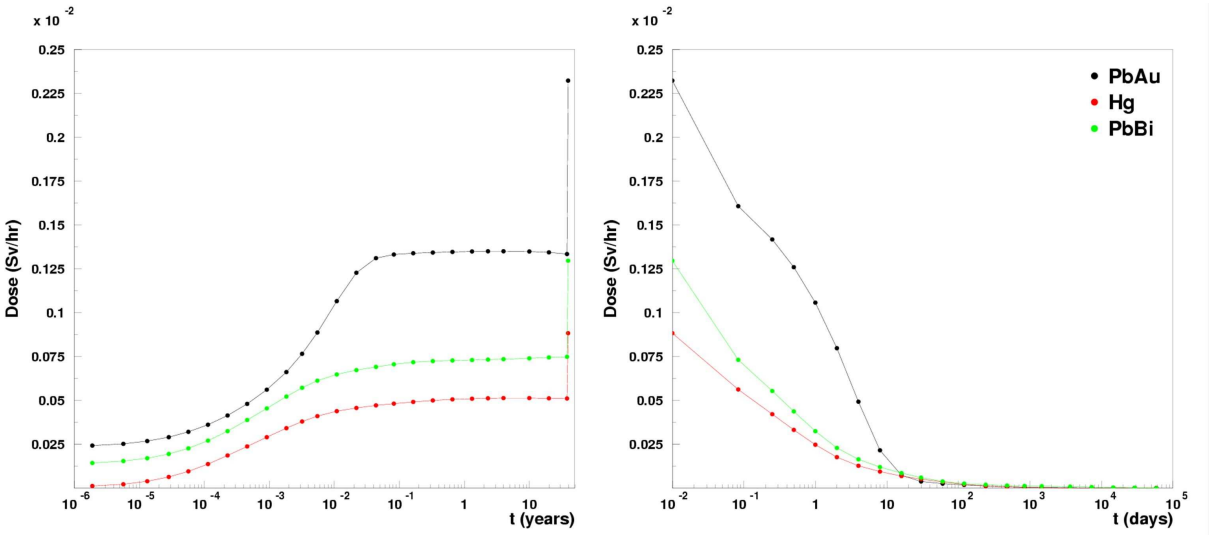

Figure 8. Dose rate from 1 g of target material at 1 m distance for three different materials during irradiation (left) and after irradiation (right).

3. Conclusions

We investigated the neutronic behaviour of an ESS target with lead-gold eutectic as spallation material. From the neutronic point of view, this material is excellent, giving practically the same neutron fluxes at the moderators as the other two materials examined.

From the nuclear point of view, we identified two potential issues with using LGE, one related to the higher decay heat and activity for a few days after shutdown, and another related to the burn-up of the target material. For the first 10 days after the end of irradiation the activity, dose rate and decay heat are considerably higher than for an LBE or mercury target (while at longer times they are lower than for the LBE), and the impact of this on the possible design of a target using this material should be studied. Additionally, the burn-up of the Au is not negligible, leading to a 3 % consumption after 40 years of operation.

4. Acknowledgments

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5. References

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