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Neutronic comparison of liquid breeders for ARC-like reactor blankets

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Abstract

The proposed blanket for Affordable Robust Compact (ARC) reactor is one of the simplest blanket concepts. It is a bulk tank filled with a lithium and beryllium fluorides molten salt. The fluid effectively works as tritium breeder, vessel coolant and neutron moderator and shield. However, despite the simplicity of the concept, the compactness of the reactor constitutes a novelty in the fusion field. It is thus necessary to evaluate all the possible solutions for an effective blanket component. This work analyses different liquid blanket identifying the most suitable for a compact fusion reactor. More specifically, the study addresses the capability of breeding tritium in a compact solution, actively shielding the coils and reducing the radioactive waste. Findings are that FLiBe optimizes the most the system in terms of applicability, tritium breeding, compactness and activation. Nonetheless, there is no lack of backup choices. For instance, there are hints that lithium-zirconium fluoride salts could accomplish the blanket main tasks in a compact reactor too. Leaving PbLi as inefficient, but cheap and still virtually viable solution.

Keywords: ARC, blanket, TBR, OpenMC, neutronics, neutron induced activation

1. Introduction

Affordable Robust Compact (ARC) reactor is a tokamak with an innovative concept design [1][2][3]. The fundamental feature is the implementation of the new High Temperature Superconductors (HTS) for the main coil sets. Such magnets can be demounted allowing a vertical assembly of the machine and, ultimately, a bulk liquid tank as blanket. HTS virtually allow to design a powerful reactor reduced in size. However, a high power-density reactor featuring a high magnetic field leads designers to face complex engineering challenges. Most of the components must be effective for their purposes in a limited space. Components such as vacuum vessel, divertors, blanket and neutron shields need to be carefully designed in order to withstand thermal, neutron, chemical and magnetic loads without dramatically increasing the system complexity nor the reactor cost. In this instance, a thin vacuum vessel and a bulk liquid blanket seem a viable choice that simplifies the design. Still, addressing to blanket choice and design is not a trivial process. In fact, blanket accomplishes very particular tasks, such as breeding tritium, transporting heat and shielding the magnets. Its optimization in terms of compactness, costs, safety and effectiveness must be carefully carried out. Because of such different purposes of a blanket and the complexity of the reactor environment, preliminary reactor designs should set aside different viable solutions. Literature provides solid assessments regarding blanket and structure/blanket concepts [4]. This work aims to provide additional compounds and to address to the most size-effective fluids. In this work several materials are analyzed as possible blankets for ARC and compact reactors in general, with the aim of coming up with different possible solutions that could backup the baseline design material (namely, lithium-beryllium fluoride – FLiBe - molten salt [1][2][3]). More specifically, the main nuclear aspects are addressed, namely the capability of breeding tritium and shielding the magnets. In addition, for safety and handling purposes, a neutron activation analysis is carried out. Once most suitable compounds are identified as breeder future studies should address other core

aspects related to the blanket component. Most essential studies, for a complete blanket assessment include deeper studies on the tritium loop self-sufficiency [5], safety studies, corrosion aspects and thermo-fluid-dynamics and magneto-hydro-dynamics analysis [6].

2. Material options

Main solutions for tritium breeders focus on compounds with high content of lithium and the other elements having a good (n, 2n) cross section and an absorption rate as low as possible. Proposed configurations range from solid breeders to liquid breeders [1][2][3][7][8][9]. Depending on the reactor design, the size and the first wall and blanket approach, such breeding-multiplying components could become dramatically complex. Some of them are divided in cassettes, each with bundles of different pipes and even double fluids for breeding and cooling, all along with neutron multipliers for meeting the required Tritium Breeding Ratio (TBR). ARC-like reactors, pursuing the high-field and compact-reactor approach [1][2][3][10][11], need to seek for simplicity on every component. A component capable to accomplish different functions would be a great achievement. In this respect, the concept of liquid breeder, able to work also as coolant and neutron shield that furthermore can fit into a tank, dramatically reducing the structure complexity [12], would hardly get abandoned in a smart design. Among the liquid breeders, the most mentioned and studied by far are the lithium-lead (PbLi) liquid metal [7][9][13] and the 2LiF-BeF₂ (FLiBe) molten salt [1][2][3][7][14]. ARC designers addressed to FLiBe, and in general, to light-molten salts for two reasons. Firstly, since ARC is a high-magnetic field tokamak, a relatively low electric conductive fluid could turn out to be the only option for magneto-hydro-dynamics issues. Secondly, ARC vacuum vessel is comparable to a thin vacuum shell immersed in the liquid. Hence, heavy and magnetic susceptible fluids could unnecessarily raise mechanical and chemical loads on the chamber walls. FLiBe seemed then to be the most valuable option for an ARC-like reactor blanket [1][2][3], thanks also to the advanced state of nuclear research for this salt, which was in first instance proposed for the molten salt fission reactors (MSR) [15][16][17] that furthermore have all conventional components analyzed and developed, already [18][19][20]. This compound shows a fairly high content of lithium, fluorine, which has a low neutron capture rate, and beryllium that is an effective neutron multiplier [21]. For FLiBe, the most likely reactions are then the following ones:



In addition, FLiBe demonstrated to be much easier to handle after irradiation, because of its low-activation properties with respect other liquid breeders [22]. However, the presence of beryllium raises concerns in terms of chemical hazards and reactor cost-effectiveness. In this view, previous studies managed to remove a thick layer of pure beryllium from ARC vessel by applying a vanadium alloy as structure [23]. Although such element is supposed to hold unique nuclear properties [24], the application of different fluids without beryllium would be obviously welcomed. In the present work, several compounds are evaluated in order to assess their eligibility as alternative breeders. To do so, the capability of overcoming the TBR>1 requirement in an ARC-like system is the first aspect that must be checked. This study proposes compound on the basis of their lithium content, presence of elements that can multiply fast neutrons and melting temperature. Alongside with beryllium, starting from 8-10 MeV of neutron energy, many other elements show a significant (n, 2n) cross section (e.g. W, Zr, V, I etc.). However, unlike beryllium, such elements have a high capture cross section in the thermal-epithermal region. They can then work as energy filters and show a particularly spectrum-dependent multiplying effectiveness. As mentioned, ARC requires liquid breeders as they do not need additional structure nor coolant. Therefore, the melting point of the proposed compounds needs to be suitable with vessel and tank materials. According to previous studies, the vessel structure will be made of

Inconel718 [1][2][3], or some vanadium alloy (e.g. V-4Cr-4Ti) [22][23]. Thus, according to creep limits of such alloys [25][26][27][28], the operation temperature range is assumed to be 750 – 1000 K. In order to expand the range of possibility, other issues that may rise with different fluids (e.g. corrosion, MHD) will not be addressed in this study and will be discussed in future works. Under these assumptions, this study identifies and analyzes the following compounds: pure-lithium, PbLi, LiF-BeF₂ (FLiBe), LiF-NaF-BeF₂ (FLiNaBe), LiF-NaF-KF (FLiNaK), LiF-LiBr-NaBr, LiF-LiBr-NaF, LiF-LiI, LiF-NaF-ZrF₄. Still, there are several other compounds containing lithium that melt at lower temperatures than ARC operating one [1][2][3][29]. Exception made for lithium and PbLi that are liquid metals; all the others are classified as molten salts.

3. Neutronics Analysis

The main necessity for a breeding blanket is to guarantee a self-sustaining tritium loop for reactor fueling. This study analyzes the tritium production of each considered compound taking advantage of a Monte Carlo neutron transport code. More specifically, the code adopted is OpenMC, developed by the advanced computation and simulation group of the Nuclear Science and Engineering Department of Massachusetts Institute of Technology [30]. The code has been developed with the main goal of a good scaling with number of computer cores during parallel computation [30][31]. Furthermore, it features a Python API interface, which makes it particularly suitable for the integration of Python routines, like multiple simulations for parametrization and sensitivity analysis. For instance, this work cycles over the different fluids and the Li-6 enrichment ratio from 10 to 100%, for each compound. The expected necessity of high Li-6 abundance over Li-7 is due to the higher (n, t) cross section of Li-6 with respect Li-7 one. However, for reactor economics, alongside a high TBR, a low Li-6 enrichment would be preferable, seeing as how Li-6 natural abundance is about 7.6% and the enrichment costs are expected to rise with the enrichment ratio. In any case, the Li-6 enrichment would probably need a detailed fuel-reactor life cycle assessment and natural reserves analysis. In fact, despite the enrichment costs, a high Li-6 fraction enhances the reactor power output increasing the heat generation directly in the liquid (see Eq. 1). On the contrary, tritium production through Li-7, besides being less likely to occur, would decrease the reactor thermal power.

3.1 Neutronic model

The model has been built as simple as possible for two main reasons. Firstly, it needed fast building and fast running performances, in order to run parametrized simulations. Secondly, although this work focuses on ARC reactor, a simplified model results could help providing an idea of tritium production in different tank-like blankets, regardless of the blanket and vessel geometry. For this reason, a model with cylindrical geometry has been built, as such geometry has already been applied in other preliminary blanket studies [23][32]. The model is similar to the one described in [23]. The main differences in the present model and the expected actual geometry are related to a cylindrical shape instead of a D-shaped and toroidal symmetric vacuum vessel. The D shape is not expected to strongly affect the TBR. It could cause some neutrons to travel longer distances than in a circular shape. However, since there is vacuum in the vessel and it is thin, it seems sufficient to us that outside the vessel there is still enough blanket (50-60 cm) to breed tritium. Regarding the toroidal shape instead of cylindrical, we just expect that the high field side of the blanket will have less blanket while the low field will have much more blanket. Therefore, it is expected that the tritium concentration will be higher in the high field side. However, such difference is not expected to affect the overall TBR. In the present model, cylinder height is 100 cm that is about a slice of 1/18 of the whole toroidal component. Inner surface is equal to the main chamber first wall surface of ARC, excluding divertors region. Thicknesses are as follows, starting from the inner cylinder: 0.1 cm of first wall made of tungsten, 1 cm of structure, 3 cm of blanket cooling channel, 3 cm of structure and 100 cm of blanket region. The vessel configuration recalls the one described by Kuang et al.[3], with the only exception of having removed the solid beryllium layer from between the two walls of the vessel. Such decision has been made because the reduction of beryllium inventory is among the goals of this work. The two structure layers have been filled with V-4Cr-4Ti (composition in [33]) as previous works suggest it helps maximizing the TBR [23] and is

evaluated to be a good structural candidate because of its good mechanical, thermal and creep properties [27][28][34][35], superior radiation resistance [35], and low activation properties [22]. Furthermore, as mentioned, such alloy allows for the solid beryllium layer removal [23]. This study picks 1 m of blanket behind the vessel as generic thickness for comparing the tritium generation capability and the shielding performances over a given thickness. Still, 1 m is on the order of ARC tank thickness [1][2][3]. An example of the model as well as all the resulting data can be found in [36]. Figure 1 shows the OpenMC model geometry output.

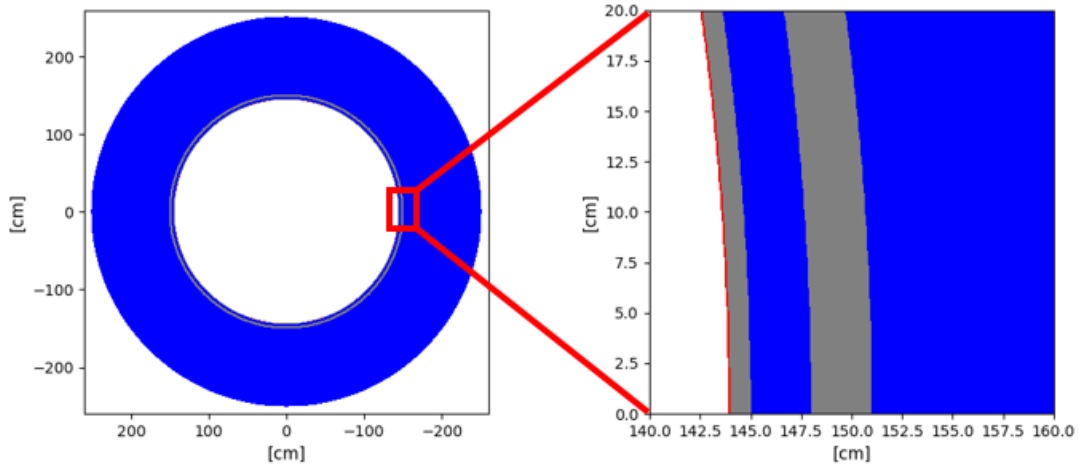


Figure 1: Geometry of the simplified model with a zoom of the vessel region (right). Vacuum vessel layers (gray) and channel and blanket region (blue) [36].

The neutron source has been set as an isotropic box source [30] 100 height and with 20 cm of edge. The model features a 14.1 MeV homogeneous energy source. Reflective boundary conditions have been imposed on both side of the cylinder, in order to simulate toroidal axisymmetry. Neutrons escaping from the blanket are considered lost, hence a vacuum boundary condition has been imposed on the outer side of the blanket cylinder. No hole has been considered, that is the blanket fully surrounds the vessel and, ultimately, the neutron source. Table 1 lists the fluids that will be analyzed in this work for evaluating their eligibility as blanket/breeders.

Table 1: Main properties of the compounds simulated in the neutron transport model.

	Composition (%mol)	Li content (%mol)	Density (g/cm ³)	Melting temperature (K)	References
Li	100	100	0.4720	454	[37]
Pb-Li	84-16	16	11	508	[38]
LiF-Bef2 (FLiBe)	67-33	28.57	1.960	732	[39][40][41]
LiF-NaF-BeF2 (FLiNaBe)	31-31-38	14.29	2.030	588	[41]
LiF-NaF-KF(FLiNaK)	46.5-11.5-42	23.25	2.020	727	[40][41]
LiF-LiBr-NaBr	20-73-7	46.5	3.160*	723	[42]
LiF-LiBr-NaF	14-79-7	46.5	3.200*	728	[42]
LiF-LiI	83.5-16.5	50	3.680*	684	[43]
LiF-NaF-ZrF4	55-22-23	20.36	2.720*	863	[44]

*According to apparent density formula

Table 1 also shows compositions and main properties that have been inserted in the OpenMC model. Because of the lack of data and validated experimental results, some of the fluid densities needed to be approximated according to the apparent density formula [42][45]:

$$\rho_{mix}^T = \sum_i X_i \cdot \rho_i^T \quad \text{Eq. 5}$$

where ρ is the density, T is the set temperature and X is the molar concentration. Model temperature has been set to 900 K for all the materials, as reference temperature. In order to achieve satisfactory statistics, the number of random walks generated for each simulation were $1E+5$ while the batches were set equal to 10. Such settings permitted to achieve relative standard deviations on the order of $1E-3 - 1E-4$ for each of the tallied quantities. Lastly, concerning the cross sections, ENDF/B-VII.1 library has been applied [46].

3.2 Neutronic results

3.2.1 Tritium Breeding Ratio

First outcomes recorded are about the tritium production. For this result a source intensity of one neutron per second has been set. Also, (n, X_t) , namely the tritium production cell tally, has been imposed on the fluid channel and the outer blanket cylinder cells. As previously mentioned, a Python routine cycled simulations over the Li-6 enrichment ratio for each of the analysed compound. Figure 2 shows the tritium production main results. On the left, there is the overall TBR for all the fluids as a function of the Li-6 enrichment ratio. On the right, the maximum TBR achievable for each fluid is displayed. On the x-axis the name of the compound is followed by the enrichment ratio in percentage that corresponds to the maximum. Both the graphs feature the error-bars, which however are not visible as standard deviations are 3-to-4 orders of magnitude lower than the mean value.

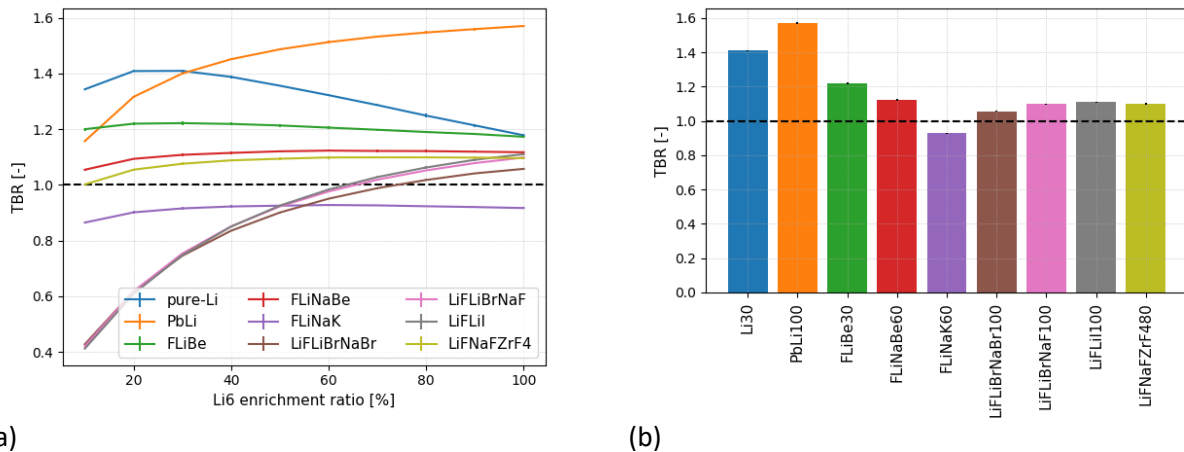
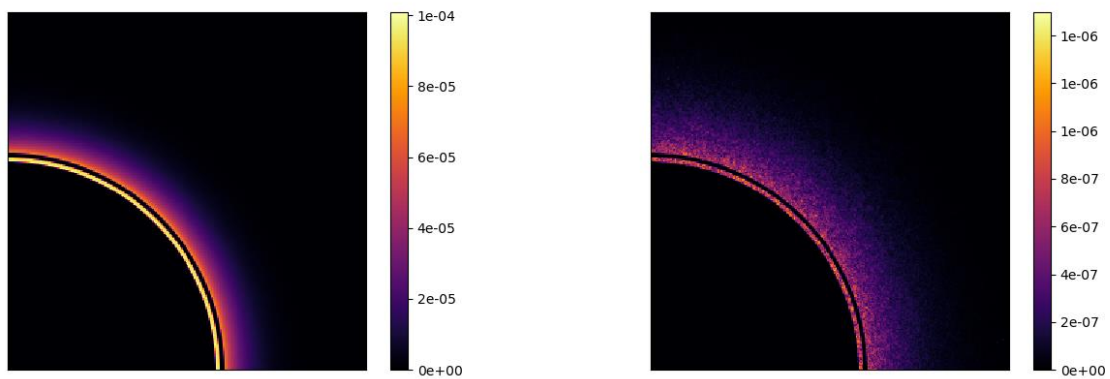


Figure 2: Tritium breeding ratio for the different fluids as a function of Li-6 enrichment ratio (a). Maximum value of TBR for each fluid (b) [36].

From Figure 2 it is possible to observe that there are many fluids able to achieve the minimum TBR requirement. Indeed, FLiNaK is the only material never reaching the unit. This is because sodium and potassium do not multiply fast neutrons and have a significant absorption cross section in the thermal region. Every other compound does have at least one multiplying element (i.e. Be, Pb, Zr Br, I, Zr) or have an overwhelming lithium concentration (i.e. pure lithium). LiF-LiI, LiF-LiBr-NaBr and LiF-LiBr-NaF exceed the unit only for extremely high lithium-6 enrichment ratio. Their applicability is then related to the economic viability of such enrichment ratio in commercial plants. LiF-NaF-ZrF4 and FLiNaBe show a good tritium production rate all over the enrichment spectrum. As their TBR is slightly higher than 1, their feasibility could be undermined by the differences among this simplified model and the reality. In addition, FLiNaBe has a

beryllium content that is similar to FLiBe one. Hence, its only advantage over FLiBe is its lower operating temperature. FLiBe and especially pure lithium and PbLi show a satisfactory TBR with a gap over the unit that can reliably balance the loop losses. Still, lithium has the flammability issue and lead is still toxic, heavy, highly magnetic susceptible and gets activated.

Most of the compounds here analysed are expected to be particularly expensive and hard to handle, especially the ones containing highly enriched lithium and beryllium. For this reason, it is here proposed a study on the minimum thickness needed for the blanket to guarantee a sufficient TBR. Figure 3 shows an example of mesh with reaction (n,Xt) tallied. In this case FLiBe with 90% of Li-6 enrichment ratio has been applied.

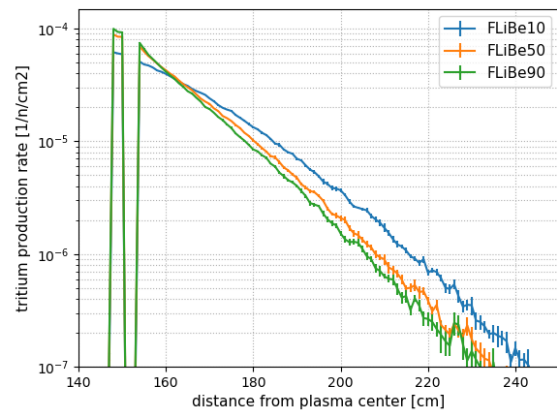
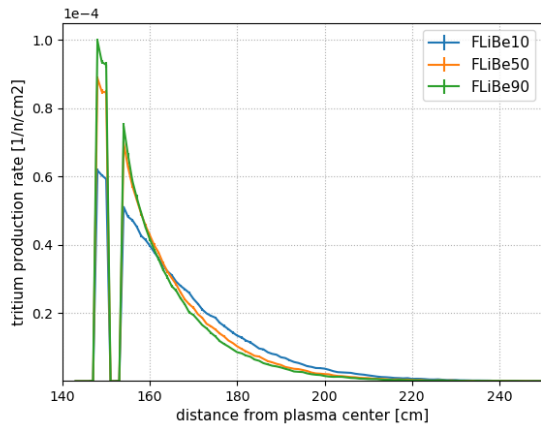


(a)

(b)

Figure 3: Tritium generation mesh tally $[1/n/cm^3]$ mean result (a) and standard deviation (b). Mesh elements are cubes of 1 cm of edge. Source intensity is equal to 1 neutron [36].

It is clear that almost all of the tritium is generated very close to the vessel, especially in the channel, confirming previous studies [3]. Such results suggest that the blanket is not required to be extremely thick for breeding tritium. Nevertheless, the tritium generation radial distribution could be affected by the lithium enrichment ratio. In fact, a compound with higher Li-7 concentration is expected to be more transparent to neutrons, according to the cross sections [21]. In this respect, Figure 4 depicts the radial distribution of the tritium production rate. Enrichment ratios of 10, 50 and 90% in FLiBe have been instanced. The first wall is at 144 cm from the center of the cylinder. The initial peak corresponds to the vessel channel while tritium production drops in the adjacent 3 cm of vessel structure. From 151 cm the continuous line introduces the bulk tank region. Figure 4 shows both a linear scale and a semilog one. Semilog scale has been added in order to easily show the difference in blanket thickness when the tritium generation rate changes the order of magnitude. For instance, a 90% enrichment requires 10 cm less thickness than the 10% enrichment to go under the $1e-6$ $1/n/cm^2$ generation. Although 10 cm could seem a small value with respect a tokamak size, it saves on the order of tens of cubic meters of blanket, assuming a vessel inner radius of 140 cm and 3.3 m of plasma major radius, that trace ARC parameters [1][2][3].

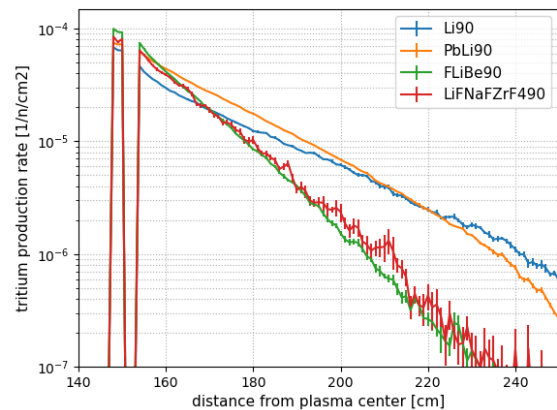
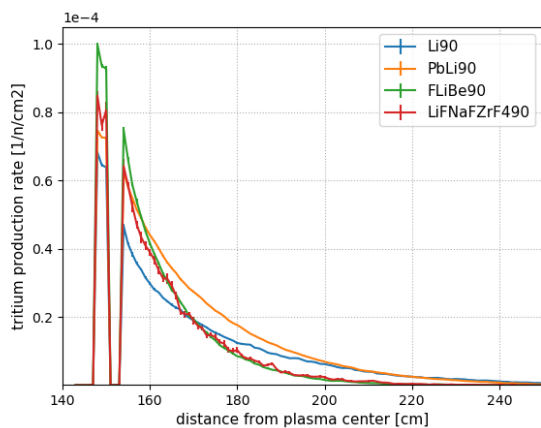


(a)

(b)

Figure 4: radial distribution of tritium generation for FLiBe with 10%, 50% and 90% enrichment ratios. Linear scale (a) and semilog scale (b) [36].

A similar analysis has been carried out for the different considered fluids. In particular, the most promising ones as breeders have been simulated (i.e. FLiBe, PbLi, pure lithium and LiFNaFZrF4) as it can be seen in Figure 5.



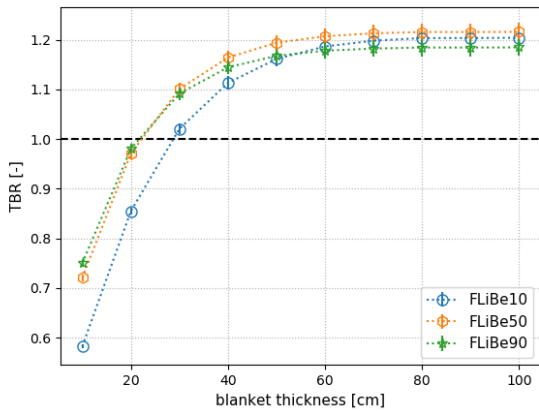
(a)

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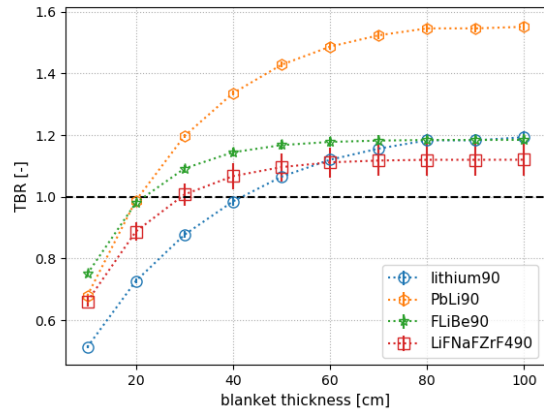
Figure 5: radial distribution of tritium generation for pure Lithium, PbLi, FLiBe and LiFNaFZrF4 with 90% Li-6 enrichment fractions. Linear scale (a) and semilog scale (b) [36].

FLiBe and LiFNaFZrF4 are the most thickness-independent breeders. Pure lithium and PbLi require additional blanket for considering the tritium production rate negligible. The first because of the low density, the latter because of the lower lithium concentration. PbLi indeed heavily relies on the multiplication effect of Pb over fast neutrons. Population increases and neutrons interact with lithium, eventually. Moreover, both PbLi and pure Li have a higher TBR than the two salts. Such additional TBR seems to be all because of the higher tritium production rate even at farther distances from the neutron source. Figure 6 displays the cumulative TBR at increasing blanket thickness. The computation has been made post-processing the (n, X_t) mesh tally results. Starting from the outer wall of the vessel and proceeding radially all the way through the blanket, the post-processing routine takes the tritium production value of each mesh element in the blanket and sums up the previous ones. In this case, the 3 cm of cooling channels are not considered blanket thickness, but rather part of the vessel. Nevertheless, their contribute in terms of tritium production is considered. From Figure 6 (a), which shows the TBR vs blanket thickness for FLiBe with three different Li-6 enrichments (10%, 50% and 90%), it is possible to notice that the higher the enrichment ratio, the thinner the minimum thickness required. High enrichment ratios require about 20-22 cm of blanket, lower enrichment ratios require 30-32 cm for reaching TBR=1. Furthermore, higher enrichment curves stabilize at about 60 cm while lower

enrichment stabilize at about 70-80 cm of thickness. In order to optimize the blanket by reaching the TBR peak, a lower enrichment ratio would need a thicker blanket.



(a)



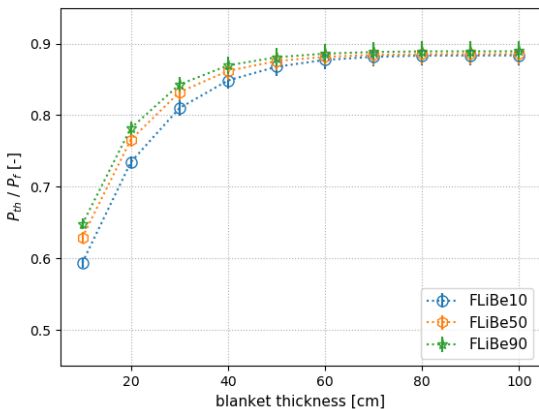
(b)

Figure 6: Cumulative TBR values as a function of different blanket thickness. For three different Li-6 enrichment in FLiBe (a) and for different fluids with 90% of Li-6 enrichment ratio (b) [36].

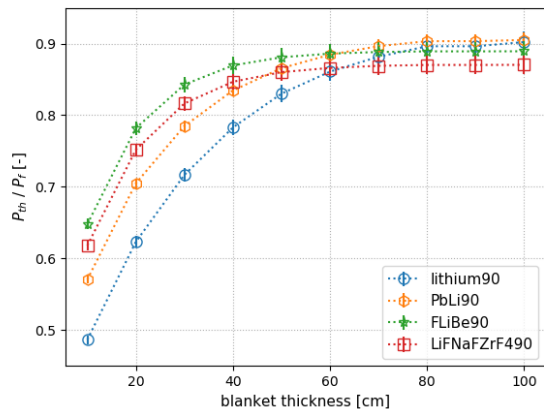
Figure 6 (b) compares the blanket thickness effect for pure lithium, PbLi, FLiBe and LiFNaFZrF4. All of them with an enrichment ratio of 90%, as reference value for this study. FLiBe and PbLi overcome the unit at about 20 cm, the Zr salt requires 30 cm and lithium 40 cm for reaching the same value. In such figure PbLi reaches the highest TBR values by far. Nevertheless, it does not seem to stabilize its cumulative tritium production rate before 80 cm of blanket thickness, and so does the pure lithium. In this sense, it is possible to state that compact blankets with PbLi are virtually doable, but they would not be efficient in terms of the tritium inventory potentially reachable.

3.2.2 Power Deposition

Although breeding tritium is probably the critical aspect of a blanket system, especially for the materials and technology choice, blankets are required to accomplish other functionalities, such as convert neutron kinetic energy to thermal energy and shielding the magnets. Concerning the heat converted it is clear that it is necessary to achieve the highest value possible, or reasonably bearable by surrounding structures, as it is directly related to the power output. With this premise, the blanket thickness is therefore defined by such necessity, once the TBR requirement is satisfied. Figure 7 shows the power deposited (P_{th}) in the reactor core (vessel, channels and blanket) normalized on the reactor fusion power ($P_f = 525$ MW), as a function of different blanket thickness. The ratio never reaches the unit because a fraction of the fusion power remains with the alpha particles still confined in the plasma.



(a)



(b)

Figure 7: Cumulative thermal power deposition (P_{th}) to plasma fusion power ratio (P_f) values as a function of different blanket thickness. For three different Li-6 enrichment in FLiBe (a) and for different fluids with 90% of Li-6 enrichment ratio (b) [36].

From Figure 7 (a) it is possible to observe that there are little differences in the power deposition of FLiBe at different enrichment ratios. Once again, a highly enriched FLiBe would need little less blanket thickness to get the power deposition to an almost constant value. Still, a 60-70 cm thick blanket seems enough for not wasting neutron energy at about every enrichment ratio. Power deposition for different fluids (Figure 7 (b)) shows a similar behavior to the tritium breeding ratio. Indeed, while fluoride salts reach their maximum at about 50-60 cm of thickness, lithium and lithium-lead require additional blanket, still on the order of 10-15 cm. Finally, TBR value slightly affects the power output. Equations 1 and 2 in section 2 show that lithium could have either an exothermic or endothermic reaction while breeding tritium. Li-6 and Li-7 cross sections show that the exothermic reaction (Eq. 1) is much more likely. Thus, a higher TBR results in a slightly higher power deposition and, ultimately, power output. Figure 7 (b) shows indeed that Li and PbLi, which showed higher TBRs experience a little higher overall power deposition, while LiFNaZrF4 show a lower power. Anyway, the difference is on the order of 10 MJ per second.

3.2.3 Shielding Effectiveness

A blanket also needs to effectively shield the structure and the magnets from neutron. In the case of ARC, in order to guarantee a coil lifetime of about 10 years[3], a 90% Li-6 enriched FLiBe needed 25 cm of ZrH2 shields in front of the most exposed magnets. Therefore, compounds able to reduce the neutron load on the magnets would surely relax the shielding component requirements. This work records the cell flux on the outer wall of the vessel and the surface current exiting the 1-meter thick blanket. Here, cell flux is defined as the number of neutrons interacting with the cell in terms of volume (namely, $[n \cdot \text{cm} / \text{cm}^3 / \text{s}]$). It is the flux integrated in the volume and then divided by the volume itself in order to obtain an average value on unit volume. Such results allow to evaluate the shielding capability of the cooling channel and the bulk tank. Figure 8 displays the flux on the vessel structure, normalized over the neutron source intensity. Namely, in order to obtain the actual flux ($n/\text{cm}^2/\text{s}$) it is necessary to multiply by the source intensity in terms of neutrons per second. It is noticeable that PbLi causes a huge load on the vessel structure. This is because of the high content of lead, it is an effective neutron multiplier and, since it is a heavy element, it causes back-scattering on the vessel. The flux is indeed more than two times higher than the flux caused by any other analysed fluid. On the contrary, pure lithium causes the lowest fluxes on the structure, especially for high enrichment ratios. This is because lithium has an extremely low cross section for (n, Xn) reactions in the fusion spectrum and it does not cause significant backscattering.

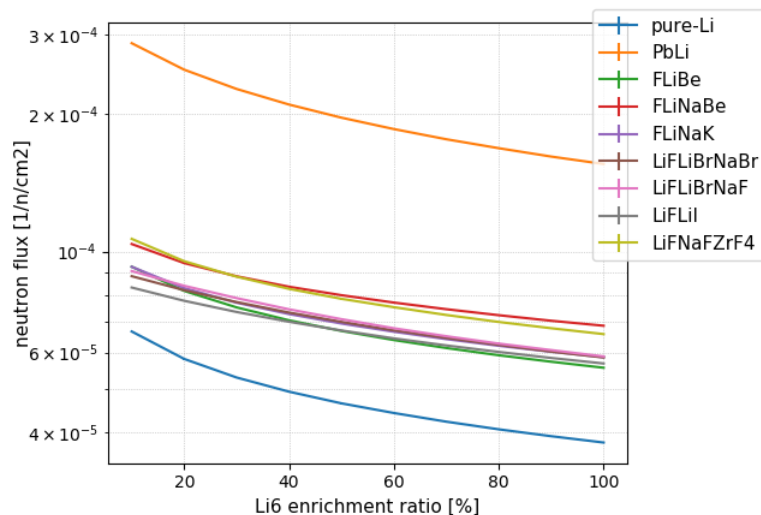


Figure 8: Cell flux on the outer wall of the vessel vs Li-6 enrichment ratio Source intensity equal to 1 [36].

Increasing the Li-6 enrichment the flux on the vessel structure visibly decreases. This is because Li-6 has higher (n, Xt) and absorption cross section than Li-7 and, also, Li-7 does not influence the neutron population while breeding tritium. Its tritium generation reaction is indeed ($n, t+n'$).

Recent studies on material damage in the case of a FLiBe-Vanadium system [47] highlighted a lattice damage rate of about 20 dpa/y on the structure, from a wall loading of about 8 MW/m², as it is expected in ARC. Assuming the damage rate goes with flux almost linearly (that is, neglecting the different spectra effect), a lithium channel could decrease the damage rate by about 30-40% with respect a FLiBe one. Similarly, lithium-lead causes a damage rate that is about three times higher the one caused by FLiBe.

Figure 9 shows the current exiting the 1 m thick blanket in logarithmic scale. Once again, normalized over the neutron source intensity. Pure lithium and lithium-lead show poor shielding capability with respect the salts. Lithium has a density that is four times lower than the FLiBe density, which rises the neutron mean free path and reduces the shielding effectiveness in terms of thickness. On the other hand, this is not the case of PbLi, which is denser than FLiBe. However, Pb has a higher ($n, 2n$) cross section for 8-14 MeV than Be. Also, Pb is much more abundant in Pb than be in FLiBe. In this case, the high content of lead and its density cause much more collisions and each collision is much more likely to get a ($n, 2n$) reaction with Pb, with respect other fluids. Even with 100% of Li-6 enrichment, current exiting from 1 m of FLiBe is 2 orders of magnitude lower than that of PbLi and pure lithium.

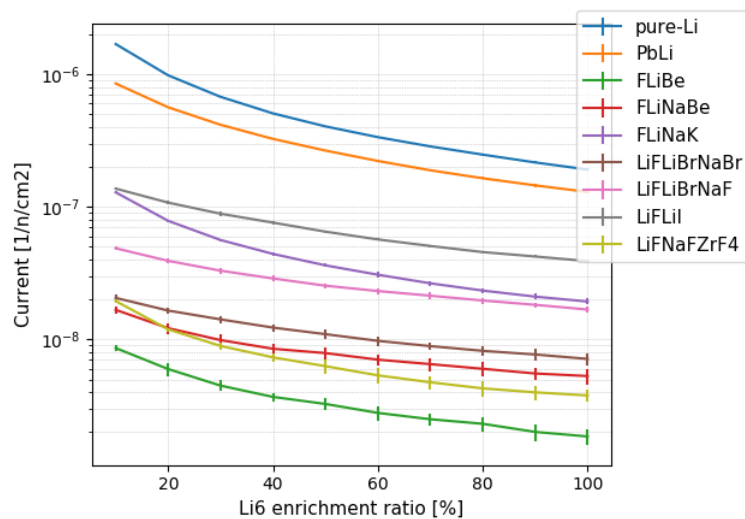


Figure 9: current exiting the 1 m thick blanket vs Li-6 enrichment ratio Source intensity equal to 1 [36].

Although currents exiting from FLiBe, FLiNaBe, LiFLiBrNaBr and LiFNaFZrF4 are on the same order of magnitude, shows that FLiBe holds the best shielding capability in this configuration. This is because, despite having a density that is similar or little lower than the other molten salts, it has a considerably lower average atomic weight, which dramatically enhances the moderation capability of the material. Therefore, it is likely that blankets other than FLiBe would need a more effective neutron shield for the magnets, in order to provide a similar lifetime of the superconductors. Once again, it is possible to notice that, increasing the Li-6 fraction, fluids become less transparent to neutrons, ultimately enhancing the compound shielding effectiveness.

4. Induced Activation Analysis

As previously mentioned, the application of FLiBe or FLiNaBe could raise some concerns for the presence of beryllium, which is considered a chemical hazard and expensive element. On the other hand, lithium-lead has a high concentration of Pb that is toxic as well. Nevertheless, chemical hazards are probably the most

concerning issues for handling such compounds only during plant loading phase. In normal operation and during unload and decommissioning phases, radiological hazards are expected to raise concerns over chemical ones. In this instance, a neutron-induced activation analysis integrates the information needed for a safety-oriented blanket material choice. This study takes advantage of the Fispact-II inventory code [48]. A reference model of 1 m³ of material has been built. Fispact-II requires the neutron flux of the studied cell, the relative energy spectrum and the irradiation time. From the OpenMC model presented in previous sections it was possible to tally the neutron flux and the energy spectrum, in particular from the channel cell, as it is the most exposed element. One year of continuous irradiation has been applied as reference setting. Although the conditions here described do not match the exact ones expected in ARC, they have been considered a good starting point for comparing the behavior of different fluids under neutron irradiation. This study carries out a preliminary analysis comparing the main elements of the compound. Thus, the compounds here simulated are considered pure, leaving an attempt impurity analysis for further works. Table 1 lists the neutron fluxes resulting from OpenMC for the different compounds considered, resulting from a 8 MW/m² of neutron wall loading, as it is expected in ARC.

Table 2: channel cell flux for the compounds selected as possible blanket. Results correspond to a neutron wall loading of 8 MW/m².

Analysed blankets	pure Li	PbLi	FLiBe	FLiNaBe	FLiNaK	LiFLiBrNaBr	LiFLiBrNaF	LiFLiLi	LiFNaFZrF4
Flux (n/cm ² /s)	7.07E+14	2.80E+15	9.94E+14	1.08E+15	9.55E+14	9.06E+14	9.22E+14	8.54E+14	1.10E+15

Figure 10 displays the Fispact-II results under the conditions previously described. Specific activity plot (Figure 10 (a)) shows a behavior that is similar to all the compounds. FLiNaK molten salt is the only one that generates a high concentration of long-lived nuclides (i.e. Ar-39, generated by a (n, p) reaction in K-39). Indeed, its specific activity decreases at a slower rate than the other compounds in the medium-long-term period. However, activity includes the tritium contribute, which is three orders of magnitude lower than the total activity at the beginning of the cooling and usually becomes dominant in the 10-30 years period. Although tritium is expected to be regularly removed from the system, this study includes its contribute as very worst-case scenario in terms of safety.

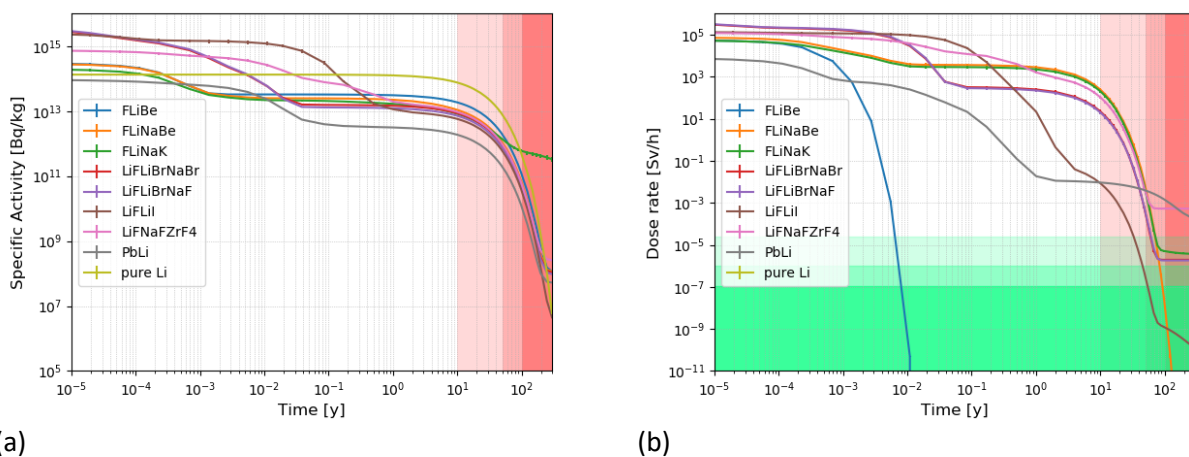


Figure 10: Specific activity vs cooling time (a) and contact dose rate vs cooling time (b).

Presence of tritium does not affect the contact dose rate (Figure 10 (b)) as such quantity does not consider alpha particles nor low energy beta radiation, which is the tritium decay-mode. Contact dose rate is mainly affected by high energy gamma radiation. In Figure 10 (b) green areas correspond to most common recycling limits [22] and it is possible to notice that few salts are able to meet such limits in a relatively short period (e.g. Pure-Li, FLiBe, FLiNaBe, LiFLiLi). Nevertheless, such results could be modified by the presence of

impurities [22]. In any case, it is clear that pure lithium and FLiBe are particularly promising from the activation viewpoint. Lithium main products are hydrogen, helium and lithium. Of these, tritium and Li-8 are the only ones to be radioactive. Tritium must be extracted as fuel, while Li-8 has a half-life of about 800 ms, which is why pure lithium does not even appear on the contact dose diagram. Fluorine and beryllium generate only short-lived nuclides as well. More specifically, N-16, F-18 and O-19 are the most abundantly generated and they have half-lives of about 7 s, 1.8 h and 27 s, respectively. The effect of adding sodium and potassium to the mentioned elements can be seen in FLiNaBe and FLiNaK curves. Potassium gives birth to Ar-39 that has a half-life of 269 years. Sodium main product is Na-22 with a half-life of 2.6 years. On the other hand, iodine generates nuclides that decay fairly quickly (e.g. I and Te isotopes). Br, Zr and Pb give birth to nuclides that decay-time is not even comparable with the timescale here selected. Bromine and lead in particular have just two natural isotopes each, suggesting that an isotopic tailoring that selects just the lowest activation isotopes would not be particularly effective. In addition, Pb leads to the generation of Po-209 and Po-210. This type of nuclides has a high energy alpha decay, which is particularly dangerous for body mucous membranes. In addition to its relatively high volatility, polonium is considered particularly hazardous, especially in case of inhalation [49]. Zirconium, with its 5 natural isotopes, could take advantage of isotopic tailoring for reducing the induced activity. Nonetheless, such process would be an additional technological step with respect a pure lithium or FLiBe blanket. It is clear that, despite the hazardous content of beryllium that could make it hard to handle in the reactor loading and startup phase, FLiBe behaves considerably better than most of the other fluids in terms of radiological hazard.

Another safety issue caused by transmutation is the heat generated by decay. This aspect must be studied both for normal operations (shut down and maintenance) and in case of accidents such as loss of coolant, loss of flow and loss of heat sink. Decay heat could cause de failure and melting of reactor components and piping if the sink fails to remove the heat. On the other hand, in case of molten salts and liquid metals, decay heat could be also used for preventing a premature freezing of the fluid.

5. Discussion

This study highlights that there are several different fluids able to overcome the requirement of $TBR > 1$. They usually are compounds made out of lithium and a neutron multiplier, often other elements that should have a low absorption cross section. Be, Pb and Zr have been found to be the most effective neutron multipliers, being beryllium the only one that could be classified as low activation. Still, beryllium raises some concerns for its high toxicity and raw material costs. This is why a compound that is able to meet all the blanket requirements without implementing beryllium would be surely well accepted in a tokamak design.

It has been shown how the Li-6 enrichment ratio heavily affects the TBR of a material. In particular salts like LiFLiBrNaBr, LiFLiBrNaF and LiFLiI require a very high enrichment ratio in order to guarantee a sufficient TBR. Hence, their applicability is related to the technologic and economic viability of a high enrichment ratio. However, such materials that show a TBR close to the unit (i.e. FLiNaK, FLiNaBe, LiFLiBrNaBr, LiFLiBrNaF and LiFNaFZrF4) do not guarantee the self-sustainability of the tritium loop. Indeed, this study does not account for tritium losses in the blanket loop and does not feature a toroidal geometry nor the actual vessel design. On the other hand, the system here considered does not implement neutron multiplying devices either. For instance, the implementation of layers or pebbles of W or PbO could help increasing the neutron population in the blanket and the TBR, ultimately. Still, these fluids could be suitable for one-of-a-kind reactor (e.g. a FNSF) that does not need a completely self-sustained fuel loop.

Pure lithium, FLiBe and especially lithium-lead show a higher TBR that could safely overcome the possible losses. For achieving the tritium production peak lithium and FLiBe require a 30% enrichment, while TBR of PbLi monotonically increases with the enrichment. This is because F and Be absorb very few neutrons. Therefore there is room for a combined effect of Li-6 (n, t) reaction and the less likely Li-7 ($n, t+n'$), optimizing the amount of tritium produced. Contrarily, as lead has a non-negligible absorption cross section, maximize

the tritium generation probability at each neutron-lithium interaction by using solely Li-6 enhances the TBR. In any case, the three fluids seem to achieve a satisfactory breeding rate at any enrichment ratio. This suggests that the decision of the enrichment ratio will be driven by other aspects than TBR requirements, for instance magnets-shielding effectiveness or economic necessities. . Both lithium and lithium-lead do not need beryllium as multiplier, which is considered a good achievement for both safety and economics. Nevertheless, lithium has flammability issues and lead is still toxic, heavy, highly magnetic susceptible and gets activated. Despite PbLi drawbacks, it is probably the cheapest raw material, thanks to the low enrichment needed and the dominant presence of lead. However, if the choice is to discard beryllium, while Li and PbLi issues turn out to be not viable in a compact reactor, a multiplier of fast neutrons such as Zr is likely to satisfy the TBR requirement. Once such condition is met, the fluid choice should be driven by safety aspects due to tritium solubility [50], thermofluid, chemical, MHD and induced activation results.

Considering other aspects that this work investigates, such as the tritium breeding distribution and the shielding capability, FLiBe seems to be the most efficient choice. Lithium is too low density for both breeding tritium and stopping neutrons in a thin blanket. Lead is a low-effective moderator, it keeps neutrons kinetic energy high along the blanket thickness increasing the lead multiplication rate and moving the interactions with lithium farther from the source. Such strategy requires more thickness than FLiBe, which has a higher Li content and moderates more effectively neutrons down to energies suitable for the (n, Xt) reaction. In this sense, FLiBe is a low-population and high-moderator breeder, while lithium-lead is a high-population and low-moderator breeder. FLiBe is then more efficient, provided that a perfectly efficient blanket would multiply as many neutrons as the minimum needed for achieving the sufficient TBR. Additional neutrons are just damaging load on the structure. On the other hand, PbLi has a TBR high enough that could breed enough tritium in the same thickness of FLiBe, in that thickness it would just not reach its maximum TBR achievable. Nonetheless, in this case, the flux exiting from the blanket is expected to be considerably higher, which would make it less effective as neutron shield. All the other salts have a good stopping and moderating capability, not as effective as FLiBe but still better than pure-Li and PbLi.

From the activation viewpoint, pure lithium and FLiBe showed the best results by far. Among the elements that do not have beryllium, LiFLiI seems to be the only compound able to get its products decayed in a reasonable time. In order to reduce the long-term radioactivity, an isotopic tailoring could be theorized for the other fluids. LiFNaZrF₄, especially, has zirconium as only element different than FLiBe and FLiNaBe and it has 5 natural isotopes. Of these, it is likely that at least one generates radionuclides with lower half-life than the others, raising the chances of an effective tailoring for reducing the long-term radioactivity. However, it would be an additional technological step, which pure lithium and FLiBe do not require.

Overall, for a compact reactor, FLiBe optimizes the system. It has a relatively low density; it breeds enough tritium in about 60-70 cm of thickness without the necessity of additional multipliers and is so far the best shield for magnets. In this respect, although FLiBe could be more expensive than other compounds as raw material, it relaxes other cost-related constraints, such as the enrichment technology, the neutron multipliers, the neutron shield, the essential volume of fluid needed and the costs related to the activated waste. Furthermore, it does not excessively multiply neutrons, unloading the vessel with respect PbLi. In ARC design viewpoint, a properly enriched FLiBe blanket could help reducing the blanket size itself. That would help relaxing ARC design limits. If 70 cm of thickness are considered enough for breeding tritium, the tank could be reduced in size and that would save more than 50 m³ of molten salt with respect the present design[3]. Such reduction in blanket thickness could be translated in a better magnet shielding components or in a bigger vessel, depending whether the problem is the coils survivability or the vessel wall loading.

More widely, from the results of this analysis it is possible to express a prevision of future high-field and compact magnetic fusion reactor concepts. According to recent studies, thanks to HTS [51] a reactor power and/or size is no more limited by the magnetic field itself. Rather it is limited by the capability of the structure to withstand the coils load, intense neutron fluxes and huge thermal loads. Nevertheless, there are hints that

new materials have the potentiality to solve such problems. High entropy alloys (HEA) [52] could relax the structural limits, including material's ductility, at both cryogenic temperatures [53][54] for the magnets, and high temperatures [55][56] for core components. Also, some of HEAs have shown superior resistance to neutron radiation [57][58]. Implementation such materials could enhance reactor's performance and/or further reduce the size. In addition to this, new materials for neutron shielding are being proposed, in particular tungsten borides and tungsten carbides [59][60][61] could be more effective and compact than boron carbides and zirconium hydrides, thanks to the high density and combined tungsten-boron absorption cross sections. Once the mentioned technologies become available on industrial scale, high-field and compact reactors should pursue HTS magnets, HEA structures, enriched FLiBe blankets and tungsten borides shields. The reactor would probably become more expensive in terms of \$/kg, but would be much more compact, reducing the volumes of the reactor itself, of the auxiliary and safety systems, inventories and facilities.

For what concerns ARC reactor, in future works other issues related to the here identified compounds will be addressed. For instance, cooling capacity, MHD and corrosion are the main concerning issues. Moreover, because of the concerns about beryllium, a life-cycle assessment should be carried out, alongside with a safety assessment addressing its actual volatility once it is in the BeF form. Positive results of such studies, parallel to this work outcomes would definitely identify FLiBe as most effective blanket for a compact fusion machine.

6. Conclusions

Although the designers of ARC already proposed FLiBe as liquid blanket for the reactor, some concerns about the presence of beryllium encouraged additional research for the identification of possible substitutes to the salt. In fact, it has been found that several compounds can virtually succeed as tritium breeders. Pure lithium, lithium-lead, FLiNaBe and LiFNaZrF₄ seem the most promising. Even other molten salts could overcome the TBR>1 barrier at high Li-6 enrichment ratios.

FLiNaBe contains beryllium and its eutectic has similar concentrations of such element as FLiBe. In addition, sodium increases the induced radioactivity of the salt. It is clear that, all other things being equal, FLiBe would be a better choice than FLiNaBe, as it also holds a safely higher TBR value. Pure lithium, although having a high TBR and being chemically compatible with vanadium, is flammable and does not shield the coils. It is not particularly suitable for a compact tokamak, then. PbLi shows the highest TBR but, despite its high density, it causes huge neutron loads on the structure. It does indeed multiply too many neutrons than needed for breeding tritium. Furthermore, it is very susceptible to MHD and its high density suggests that it would cause additional pressure stresses on the immersed vacuum vessel. Lead is also a toxic element and its induced activation lead to the formation of long-lived radionuclides. Nonetheless, PbLi is one of the cheapest liquids here considered, in terms of raw material. Lithium-lead should be considered in the case of prohibitive reactor costs caused by FLiBe or other fluids. In this view, a life cycle assessment on the combined vessel-blanket-shield system should give the most economic-viable solution.

It is clear, from the results, that FLiBe has unique features able to fit very well in a compact reactor. FLiBe seems to be the most efficient choice in terms of TBR, moderation, shielding and activation. TBR is safely similar to 1.2 at any enrichment ratio. It is relatively low-density but still shields neutrons better than any other fluid here studied. It is able to breed tritium in a relatively low thickness tank, helping to reduce the reactor size or leaving room for additional shields. Finally, it could probably be classified as low-activation material, depending on the actual impurities effect. FLiBe only problem seems then to be the beryllium presence. Regardless, it is most likely the best choice for a high-field compact reactor like ARC.

The only other option here found is replacing beryllium with zirconium as neutron multiplier. Namely, replacing BeF with ZrF₄ or NaF-ZrF₄, like in LiFNaZrF₄. Such process could satisfy the TBR requirement

without adding more stress to the structure or needing for additional neutron shields, with respect FLiBe option. Nonetheless, Zr generates longer-lived nuclides with respect beryllium.

Therefore, being most likely FLiBe the best fitting choice for an ARC-like reactor, lithium fluoride salts containing Zr in spite of Be, as possible substitutes, could worth additional research programs.

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Data Availability Statement

The raw data required to reproduce these findings are available to download from and the processed data required to reproduce these findings are available to download from [<http://dx.doi.org/10.17632/2bncdpftxm.1>].

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