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ARC reactor: Activation analysis of realistic FLiBe compositions for the liquid blanket

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SOMMARIO

A causa della crisi ambientale che stiamo vivendo, l'energia nucleare sarà probabilmente una parte importante della soluzione per soddisfare il fabbisogno energetico mondiale. Al giorno d'oggi la produzione di energia nucleare avviene solo grazie ai reattori a fissione, ma si sta studiando un nuovo metodo che utilizzerebbe la fusione di due nuclei leggeri. La produzione di energia da fusione ha diversi vantaggi tra cui: il combustibile è praticamente inesauribile in quanto si basa su due isotopi di idrogeno, non c'è possibilità di incidenti dovuti a reazione di fuga, è un'energia carbon free, ha un rischio di proliferazione trascurabile e ha un basso livello di radioattività residua.

Sono diversi i progetti che si stanno concentrando sull'energia da fusione, uno dei più interessanti è sicuramente quello del Plasma Science and Fusion Center del Massachusetts Institute of Technology denominato ARC (Affordable, Robust and Compact). Questo reattore si basa sulla reazione del Deuterio con il Trizio ed è caratterizzato da importanti innovazioni sia dal punto di vista progettuale che di design. Tra le innovazioni più importanti ci sono la presenza di superconduttori ad alta temperatura (HTS), che permettono di diminuire le dimensioni del progetto e allo stesso tempo permettono di raggiungere un alto valore di campo magnetico, e un'innovativa coperta composta da un sale fuso chiamato FLiBe che è una miscela di Fluoro, Litio e Berillio.

Lo scopo del lavoro è l'analisi di attivazione del FLiBe. Per fare ciò le analisi sono state effettuate utilizzando il programma FISPACT-II che permette di valutare l'Attività Specifica, la Dose Rate di contatto e il Calore di Decadimento come funzioni del tempo. Diverse ipotesi sono state fatte sulla composizione del sale fuso, valutandolo prima puro e poi con impurità. Successivamente, l'analisi è passata a uno dei componenti principali di FLiBe, vale a dire il berillio. Questo materiale è in grado di produrre trizio quando viene irradiato per trasmutazione. Il problema è che durante l'estrazione c'è spesso la possibilità di trovare dell'uranio parassitario. L'uranio porta a varie complicazioni essendo materiale radioattivo. L'analisi è stata effettuata tenendo conto delle diverse percentuali di uranio all'interno del berillio. Successivamente, è stata effettuata un'analisi di attivazione di un nuovo materiale sperimentale chiamato "BP-1" contenente uranio tra le impurità.

L'ultima parte dello studio è un'analisi di attivazione di FLiBe con uranio all'interno. Gli aspetti fondamentali esaminati sono lo studio della trasmutazione del plutonio durante l'irradiazione e una valutazione degli attinidi e dei prodotti di fissione durante la fase di raffreddamento.

Il risultato più interessante è sicuramente l'influenza che anche una piccola percentuale di uranio ha sul sistema. Questo elemento una volta irradiato produce isotopi altamente radioattivi con un'emivita molto lunga, come il plutonio-239, che sono dannosi per il sistema.

ABSTRACT

Due to the environmental crisis, we are experiencing, nuclear energy will likely be an important part of the only solution to meet the world's energy needs. Nowadays nuclear energy production takes place only thanks to fission reactors, but a new method is being studied that would produce energy through nuclear fusion. The production of energy from fusion would bring several advantages including: the fuel is practically inexhaustible as it is based on two isotopes of hydrogen, there is no possibility of accidents due to runaway reaction, it is a carbon free energy, it has a negligible proliferation risk and it has a low level of residual radioactivity.

There are several projects that are focusing on fusion energy, one of the most interesting is certainly that of the Plasma Science and Fusion Center of the Massachusetts Institute of Technology called ARC (Affordable Robust and Compact). This reactor is based on the reaction of Deuterium with Tritium and is characterized by important innovations from the point of view of design and size. Among the most important innovations there are the presence of high temperature superconductors (HTS), that permit to decrease the dimension of the project and at the same time it can reach high value of magnetic field, and an innovative blanket composed of a molten salt called FLiBe which is a mixture of Fluorine, Lithium and Beryllium.

The aim of the work is the activation analysis of the FLiBe. To do that the analyses were made using the program FISPACT-II that permits to evaluate the Specific Activity, the contact Dose Rate and the Decay Heat as functions of the time. Several hypotheses have been made about the composition of the molten salt, evaluating it first pure and then with impurities. Subsequently, the analysis moved on to one of the main components of FLiBe, namely Beryllium. This material is able to produce tritium when it is irradiated by transmutation. The problem is that during the extraction there is often the possibility of finding parasitic uranium. Uranium leads to various complications as a radioactive material, in fact it produces a series of radionuclides, such as plutonium, with a very long half-life. The analysis was carried out taking into account different percentages of uranium within beryllium. Subsequently, an activation analysis was made of a new experimental material called "BP-1" containing uranium among the impurities.

The last part of the study is an activation analysis of FLiBe with uranium inside. The fundamental aspects examined are the study of the transmutation of plutonium during irradiation and an evaluation of actinides and fission products during the cooling phase.

The most interesting result is certainly the influence that even a small percentage of uranium has on the system. This element once irradiated produces highly radioactive isotopes with a very long half-life, like the plutonium-239, which are harmful to the system.

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1. Introduction

1.1. Fusion reaction

Nuclear fusion will be one of the most important innovations regarding energy production in the future: it is renewable, it does not release CO_2 into the environment and it does not generate nuclear waste, which are typical of fission reactors. Another difference with the fission's reactors is the different way to produce energy, conceptually the opposite, in fusion indeed the emission of energy comes from the union of two atoms, these, merging, produce one or more different atomic nuclei with the release of some subatomic particles like neutrons or protons.

From a physical point of view, it is very complicated to merge two atoms as there are several steps to overcome requiring huge amounts of energy. The main force to overcome is the Coulomb one. This causes two electrically charged objects to interact with each other, it is a repulsive force in case the charges have the same sign, attractive otherwise [1]. To overcome the repulsion, it is necessary to bring the nuclei close enough to intervene a new force, the one called "strong nuclear force". This force, however, is characterized by being short-range [2]. It is therefore necessary to evaluate the binding energy, this is given by the difference in the energy of the free components minus that of the same bound components. In the figure 1 it's possible to see the average value of the binding energy as a function of the number of nucleons.



Figure 1: Binding energy per number of nucleons [1].

On the x-axis we find the number of nucleons and, on the y-axis, the average binding energy. The maximum of the curve is reached with the isotope of iron ⁵⁶Fe, this isotope acts as a watershed between light nuclei and heavy nuclei. In the light ones we see how the binding energy grows very quickly as the atomic mass increases, while for the

isotopes after ⁵⁶Fe the curve tends to decrease slowly. This implies that the fusion of light nuclei leads us to climb the steepest part of the curve, releasing large amounts of energy.

Another advantage of light nuclei is that these are small and poor in protons making it easier to overcome repulsion.

The first step in making two nuclei merge is to create the ideal conditions for the nuclear force of attraction to act. Then we eliminate the electrons from the system by completely ionizing all the atoms. In doing so we create a new state of matter called plasma. The problem with plasma is that the atoms are no longer neutral and therefore the Coulomb force acts which causes the ions to repel each other. To overcome this force, we therefore need very high temperatures. A useful figure of merit in nuclear fusion is the triple product that allows us to evaluate the feasibility of self-sustaining fusion reactions through three parameters: temperature, density and confinement time. Since the last two are limited to a narrow range the only that can be easily modified is temperature.

Once we manage to bring the nuclei closer together the force of nuclear attraction grows rapidly and the nucleons can "fall" into each other [2], the result is fusion and the net energy produced. The fusion of lighter nuclei, which creates a heavier nucleus and often a neutron or free proton, generally releases more energy than is needed to force the nuclei together. The physics behind fusion as an energy source is based on exothermic reactions involving the isotopes of hydrogen and helium:

$$D + D \rightarrow He^{3} + n + 3.27 MeV$$

 $D + D \rightarrow T + p + 4.03 MeV$
 $D + He^{3} \rightarrow He^{4} + p + 18.3 MeV$
 $D + T \rightarrow He^{4} + n + 17.6 MeV$

The first two reactions could be the preferable choice in a hypothetical reactor given the great abundance of deuterium present in nature but as can be seen in Figure 2, D-D reactions are characterized by low reaction rates. The D-He³ relationship is characterized by low reaction rates, moreover, ³He is very rare in nature. Only the D-T relationship remains. This one has the largest cross section at low temperatures. The problem with this relationship is tritium, that is rare and radioactive, and is also characterized by a very low half-life. This feature forces us to produce it, to do this we use ⁶Li blanket which, surrounding the nuclear reactor, can provide tritium using the neutron output [3]. The relationship that describes this reaction is:

$$Li^6 + n \rightarrow He^4 + T + 4.8 MeV$$

Figure 2 shows the collision cross-section curves for some common fusion reactions.



Figure 2: Fusion cross-sections of various fusion reactions as a function of kinetic energy of an incident D or p on a stationary target [2].

One of the biggest problems when it comes to plasma is confinement. To do this an exploitation of the electromagnetic properties of the plasma, which allows to confine it magnetically. The most effective way to trap plasma particles along magnetic field lines is through a machine called a tokamak. In a tokamak, the plasma is kept in a toroidal shape thanks to a specially designed magnetic field configuration. This configuration is composed of three magnetic fields: the toroidal one, the vertical one and the poloidal one. The first two are externally induced while the poloidal one is generated directly from the plasma through an induced current. Figure 3 shows a solenoidal set of ring-like coils, in the form of a torus, thus forming a toroidal magnetic field.



Figure 3: Schematic of a tokamak [2].

1.2. ARC

The ARC reactor is a project born within the "Plasma Science and Fusion Center of the Massachusetts Institute of Technology", the acronym ARC stands for: Affordable, Robust and Compact. The main objective of this ambitious project is to minimize the size of the reactor. This would entail a series of advantages, the most important however is certainly that of reducing the cost of the system [4]. Figure 4 shows a conceptual scheme of ARC reactor and power plant.



Figure 4: ARC reactor and power plant scheme [5].

ARC is a conceptual reactor, still under study. It is characterized by innovative details, such as the use of high temperature superconductors, which allow to obtain both high values of magnetic fields and to reduce the size of the tokamak. Another novelty is the liquid blanket, composed of a molten salt called FLiBe (Fluoride, Lithium, Beryllium), the blanket has a moderation and cooling function. Finally, one last big news is the design of a one-piece vacuum vessel. This feature makes replacement easier and is possible thanks to the presence of high temperature superconducting field coils that can be split to remove the top of the reactor and replace the vacuum vessel.

The expected fusion power is 525 MW, not far from the expected value for ITER of about 620 MW, but the big difference is in the radius: that of ARC is practically half compared to that of ITER, in fact the larger one measures 3.3 m while that of shorter 1.13 m, in ITER instead 6.2 m and 2 m. [4]. Another value influenced by the difference in size is the Plasma volume, in ARC it measures 141 m³ against the 837 m² of ITER.

The main design parameters of ARC are reported in the table 1.1.:

| DESIGN PARAMETER | VALUE |
|-----------------------------|-------------------------------------|
| Fusion Power | 525 MW |
| Total thermal power | 708 MW |
| Plant thermal efficiency | 0.40 |
| Total electric power | 283 MW |
| Net electric power | 190 MW |
| LHCD coupled power | 25 MW |
| ICRF coupled power | 13.6 MW |
| Power multiplication factor | 3.0 |
| Major radius | 3.3 m |
| Plasma semi-minor radius | 1.13 m |
| Plasma elongation | 1.84 |
| Plasma volume | 141 m ³ |
| Toroidal magnetic field | 9.2 T |
| Peak on-coil magnetic field | 23 T |
| Plasma current | 7.8 MA |
| Bootstrap fraction | 0.63 |
| Tritium breeding ratio | 1.1 |
| Avg. Temperature | 14 keV |
| Avg. density | $1.3 X 10^{20} m^{-3}$ |
| On-axis temperature | 27 keV |
| On-axis density | $1.8 \times 10^{20} \text{ m}^{-3}$ |
| Greenwald fraction | 0.67 |
| Toroidal beta | 1.9% |
| Internal inductance | 0.67 |
| Normalized beta | 2.59 |
| Safety factor at r/a=0.95 | 7.2 |
| Edge safety factor | 4.7 |
| Minimum safety factor | 3.5 |
| Fusion power wall loading | 25 MW/m ² |
| Energy confinement time | 0.64 s |
| H89 confinement factor | 2.8 |
| H98(y,2) confinement factor | 1.8 |
| G89 gain factor | 0.14 |

Table 1.1. List of ARC design parameters [4].

1.2.1. Main Components

The main components that characterize the design of ARC are:

- Vacuum Vessel: one-piece component with a toroidal shape, composed of Inconel-718. The presence of Nickel has an advantage from the point of view of corrosion resistance but makes the vessel subject to nuclear activation, creating many problems, especially during disposal. It is located between the first wall and the tank. There is also a layer of beryllium used as a neutron multiplier, in addition beryllium, once activated, produces tritium that can be collected and used as fuel.
- First-wall: located in the inner part of the vacuum vessel facing the plasma chamber. It is composed predominantly of tungsten. Due to the proximity to the plasma, it is subject to high temperatures and neutron flux. For these reasons it can release impurities into the system. The release of impurities is a phenomenon called

sputtering. This phenomenon occurs when a solid is bombarded with charged particles. If the energy of these particles is greater than the bond energy of the solid, there is an expulsion of atoms from the surface [6]. The problem of impurities is their neutrality, since, not being charged, they are not affected by the magnetic effect given by superconductors and therefore can cause a loss of energy, thus making the temperature inside the plasma decrease in consequence. To overcome this, a divertor is used.

- Divertor: is a component placed inside the vacuum vessel and is composed of plates that allow to concentrate the interactions in a limited region. In this way it is avoided that there are areas of the first wall are inadvertently affected by heat. The divertor is based on a particular X-shape magnetic configuration. This configuration modifies the diffusion of particles along the lines of the field.
- Blanket: a big difference compared to those used in other fusion reactors is the presence of a liquid blanket composed of a molten salt, in this case the salt used is the FLiBe. This compound is very useful both from a tritium production point of view and in carrying out the cooling task of the system. The production of tritium takes place because FLiBe is also composed of Beryllium and Lithium, materials that, by transmutation, produce H₃. The parameter that needs to be controlled is the tritium breeding ratio (TBR), which must be greater than 1 to sustain the entire reaction cycle, for ARC a TBR of 1.1 [4] was considered in its design.
- Cooling System: located inside the vacuum vessel is able to remove thermal energy from the system and transfer it to a turbine so as to be able to produce electricity. The ability to remove heat is useful during reactor operation as it avoids serious damage to components subjected to high thermal loads.
- Superconducting Magnets: there are three main magnets systems inside a fusion reactor, they are extremely important for the stability and confinement of the plasma. They are divided into central solenoid, toroidal field coils and poloidal field coils. Figure 5 shows the original model on the left and a simplified model in the centre.



Figure 5: Magnetic equilibrium, poloidal field (PF) and trim (TR)coil set, vacuum vessel and FLiBe tank geometry identified by the ARC-Divertor design study [7].

A difference compared to ITER consists in the use of high temperature magnets, which allow to reduce the size of the reactors without undermining the generation of magnetic fields. As previously mentioned, these magnets have the advantage of being able to be divided when you want to replace the vacuum vessel. For ARC the selected material is Rare Earth Barium Copper Oxide (REBCO).

1.2.2. The Blanket

Of particular interest in our study will be the blanket, this component is new in its genre, in fact, as we said previously, it is composed of a molten salt the FLiBe. This mixture is very useful both for cooling the system and to produce tritium. It is composed of LiF and BeF₂ and has several advantages from an engineering point of view: FLiBe acts as a high temperature heat transfer fluid that achieves high thermal efficiencies, is a good shield against radiations and has good fluid dynamic properties [8], also does not react with air and water. The large temperature range over which FLiBe is liquid permits an out-put blanket temperature of 900 K [4]. Figure 6 shows the production of tritium in the FLiBe cooling channel within the VV wall.



Figure 6: Production of tritium in ARC in the coolant channel [10].

An important parameter in the study of the blanket is the Tritium Breeding Ratio (TBR), this value tells us the ratio of the rate of tritium production in the system to the rate of tritium burned in plasma [8]. It is vital for a fusion reactor that more than one tritium atom per fusion neutron is produced in order to maintain the fuel cycle. To do this, one of a 1 cm non-structural beryllium layer was added as a neutron multiplier. At the end of all the calculation of the TBR of the final project is $1.08 (\pm, 004)$. This value is slightly lower than the estimated 1.1. About 26% of the tritium produced inside the reactor is generated in the cooling channel FLiBe inside the VV wall [10].

1.3. FISPACT-II

The program used to carry out our analyzes is FISPACT-II. This program is an inventory code capable of performing modelling of activation, transmutations and depletion induced by neutron, proton, alpha, deuteron or gamma particles incident on matter [11]. that ENDF is the library that was used for the analysis, and the input data, like cross sections, were considered at the operational temperature of 900 K. The main inputs for the inventory file for FISPACT-II were the characteristics of the material, such as density, mass percentage composition, the total mass and the neutron flux. Fispact-II possesses many features but the most common simulations are performed with time-dependent inventories, observables and emitted particle data. The code operates in four stages:

- 1. process the library data
 - a. collapse cross-section data with incident particle spectra
 - b. condense decay and fission yield data
 - c. print summary of library and simulation-specific data
- 2. set initial conditions
- 3. run irradiation (heating) phases
- 4. run cooling phases

2. FLiBe activation analysis

2.1. Introduction

The use of molten FLiBe as a coolant for fusion reactors has several engineering and safety advantages. FLiBe serves as a high temperature heat transfer fluid that achieves high thermal efficiencies, acts as a good radiation shield, and has modest to good fluid flow properties. One of the inducements for FLiBe in fusion reactors is its inherent ability to produce the fusion fuel tritium from the reaction of fusion neutrons with the lithium in the FLiBe as we see in the paragraph 1.1. Lithium and beryllium are good moderators and form a eutectic mixture. Beryllium also does neutron splitting, improving neutron economy [12]. Another important aspect is the possibility to maximize the probability of tritium breeding thanks the Fluorine and the Beryllium, indeed these two materials absorb few neutrons. In figure 8 is possible to see the generation of tritium neutron transmutations of the beryllium.



Figure 7: Activation and decay paths for lithium and beryllium in FLiBe [9].

The radioisotopes created from neutron activation of pure FLiBe are ¹⁰Be, ¹⁴C, and ¹⁸F. The dominant activation product is ¹⁸F.

2.2. Pure FLiBe

Pure FLiBe was studied with different periods of irradiation: the first corresponds to one year, the second instead to ten years. A flux of $10^{15} \frac{neutrons}{cm^2 s}$ closer to expected typical values for ARC vacuum vessel was initially used.

Through FISPACT-II I was able to evaluate the irradiation phase first and then the cooling phase. In the latter, the trends in different were viewed. The interval taken into consideration ranges from one second up to a thousand years. In the table 2.1. there is the composition by elements of the material provided to us by the program including the number of atoms of the element, number of gram-atoms, number of grams, power output (Curie-MeV and kW), power output (Curie-MeV and kW) and power output (Curie-MeV and kW):

| | ATOMS | GRAM- ATOMS | GRAMS | BETA CURIES- MeV | kW | GAMMA CURIES- MeV | kW | ALPHA CURIES- MeV | kW |
|----|----------|----------------|----------|------------------------|----------|-------------------------|----------|-------------------------|----------|
| Li | 9.83E+28 | 1.63E+05 | 9.98E+05 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Be | 4.91E+28 | 8.15E+04 | 7.35E+05 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| F | 1.97E+29 | 3.27E+05 | 6.21E+06 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |

Table 2.1. Composition by elements of the Pure FLiBe

FLiBe is a molten salt made from a mixture of lithium fluoride (LiF) and beryllium fluoride (BeF₂). In our study we used stable isotopes, including ⁷Li, ⁹Be and ¹⁹F. Furthermore, the Lithium has been enriched to 90% with ⁶Li to have a TBR value > 1 [13].

2.2.1. Formation of nuclides and percentage contribution of them to activity, dose rate and Heat during the irradiation phase

With a flux of 10¹⁵ for a year forty-two nuclides were formed, of these four were already present and are stable, ⁶Li, ⁷Li, ⁹Be and ¹⁹F. Among the different isotopes formed, some are stable, others are not, the following table shows:

| STABLE: | H1-H2-He3-He4-B10-B11-C12-C13-N14-N15-016-017-018-Ne20- Ne21-Ne22-Ne23 |
|-----------|--|
| UNSTABLE: | H3-HE6-BE10-BE11-B12-B13-C14-C15-N13-N16-N17-N18-N19- O19-O20-F17-F18-F20-F21 |

Table 2.2. Different isotopes formed during the radiation

The production of tritium (³H) is certainly very important, 1.043E+04 grams are produced. This nuclide is particularly important because it is the major contributor to the activity produced, respectively 39.07%. Total activity is 9,4982E+18 Bq. ¹⁶N is certainly also relevant, in addition to the contribution to the activity of 2.2611E + 18 Bq (23.81%) it is the largest emitter of radiation, 5.1335E+05 Sv/hr i.e., 85.48% of total radiation and is also the major heat producer with 2.6449E + 03 kW i.e., 71.33% of all heat produced.

In the figure 9 is possible to see the contribution of each nuclide to heat production, the total one produced is 3.7079E+03 kW.



Figure 8: Percentage contribution of each nuclide to the Decay Heat produced.

In the figure 10 it's possible analyse the contribution of each nuclide to the Dose Rate, the total one is 6.0054E+05 Sv/hr.



Figure 9: Percentage contribution of each nuclide to the Dose Rate.

In the last figure it's been analysed the contribution of each nuclide to the Activity, the total one is 9.4982E+18 Bq.



Figure 10: Percentage contribution of each nuclide to the Activity.

A final aspect analysed is the composition of the material by elements, useful to understand all the new elements that have formed during irradiation:

| | | GRAM- | | BETA CURIES- | | GAMMA CURIES- | | ALPHA CURIES- | |
|----|----------|----------|----------|-----------------|----------|------------------|----------|------------------|----------|
| | ATOMS | ATOMS | GRAMS | MeV | kW | MeV | kW | MeV | kW |
| | | | | | | | | | |
| Н | 2.28E+27 | 3.79E+03 | 1.08E+04 | 5.73E+05 | 3.39E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| He | 3.28E+27 | 5.44E+03 | 2.17E+04 | 3.01E+07 | 1.79E+02 | 1.09E+05 | 6.46E-01 | 0.00E+00 | 0.00E+00 |
| Li | 9.62E+28 | 1.60E+05 | 9.77E+05 | 5.24E+04 | 3.11E-01 | 2.79E+02 | 1.65E-03 | 2.64E+04 | 1.56E-01 |
| Be | 4.89E+28 | 8.11E+04 | 7.31E+05 | 3.93E+01 | 2.33E-04 | 1.20E+01 | 7.12E-05 | 3.07E-01 | 1.82E-06 |
| В | 2.08E+22 | 3.46E-02 | 3.81E-01 | 1.35E+06 | 7.99E+00 | 1.93E+04 | 1.15E-01 | 1.42E+03 | 8.41E-03 |
| С | 3.97E+23 | 6.59E-01 | 8.57E+00 | 3.43E+05 | 2.03E+00 | 4.35E+05 | 2.58E+00 | 1.18E-02 | 6.99E-08 |
| Ν | 5.42E+26 | 9.00E+02 | 1.35E+04 | 1.64E+08 | 9.71E+02 | 2.82E+08 | 1.67E+03 | 2.76E+03 | 1.64E-02 |
| 0 | 2.48E+26 | 4.12E+02 | 7.14E+03 | 4.52E+07 | 2.68E+02 | 2.65E+07 | 1.57E+02 | 0.00E+00 | 0.00E+00 |
| F | 1.96E+29 | 3.26E+05 | 6.19E+06 | 2.27E+07 | 1.34E+02 | 5.19E+07 | 3.08E+02 | 0.00E+00 | 0.00E+00 |
| Ne | 5.64E+24 | 9.37E+00 | 1.87E+02 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Na | 1.12E+08 | 1.86E-16 | 4.28E-15 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |

Table 2.3. Composition by elements of the pure FLiBe after a year of irradiation

2.3. Cooling phase analysis for different radiation times

As for the cooling discourse, different time periods have been analysed since the flow was zeroed: 1 Seconds, 3 Seconds, 5 Seconds, 25 Seconds, 40 Seconds, 1 Minutes, 7 Minutes, 12 Minutes, 30 Minutes, 45 Minute s, 45 Minutes, 1 hours, 5 hours, 9 hours, 17 hours, 1 days, 2 days, 5 days, 12 days, 20 days, 50 days, 120 days, 230 days, 1 years, 3 years, 8 years, 15 years, 50 years, 100 years, 200 years, 500 years, 1000 years.

2.3.1. Activity

One of the predictable things is the decrease of many of the unstable nuclides present at the beginning, at the end of the study period only ¹⁰Be and ¹⁴C present a still appreciable activity,

- ¹⁴C 5.3587E+11 Bq
- ¹⁰Be 1.4901E+08 Bq

Indeed, their half-life is equal to five thousand, seven hundred and thirty years for ${}^{14}C$ and is equal at about a million years for ${}^{10}Be$.

The trend of tritium activity remains approximately constant for the first five days, after which it begins to decrease until it is zero (in our analysis the zeroing comes shortly after five hundred years).

Most of the most unstable nuclides disappear after a few minutes. An example is ¹⁶N, this radionuclide is the second major contributor to the activity after the radiation, as it shown in figure 11, but it has a very short half-life, equal to 7.13 seconds. After five minutes only more ¹⁴C, ¹⁰Be and ³H have an appreciable activity. In figure 12 is shown the trend of the activity.



Figure 11: Total Activity and Tritium's activity during cooling time after a radiation of 1 year.

The curve is approximately constant for the first two hundred and thirty days, then the value decreases very quickly until it returns to about constant after three hundred and seventy-eight years. This happens because, as can be seen in the graph, tritium no longer contributes to activity, this implies that the only nuclides that contribute are ¹⁴C and ¹⁰B.

Although the irradiation time has increased, the results obtained show several peculiarities with the previous case, but also some differences. The analysed period is the same with the same intervals. The biggest differences are caused by the greater number of unstable nuclides that have produced, especially in the first phase of cooling, higher values for activity, dose rate and heat. Figure 13 shows the comparison between the activity after ten tears of irradiation and the one after 1 year.



Figure 12: Total Activity after 10 years compared with the one of 1 year during cooling time.

Also here at the end of the analysed period the unstable nuclides still present in our system that present an appreciable activity are:

- ¹⁴C 4.7094E+13 Bq
- ${}^{10}\text{Be}$ 1.5633E+09 Bq

As expected, the activity issued is much higher than that of the previous case, as the radiation time was ten times higher.

As you can see in the figure 13 the trend of the activity, both total and tritium, is the same with only an order of magnitude higher due to the greater radiation time. Interesting is the fact that tritium takes much longer to reset than in the previous case.

2.3.2. Dose Rate

As for the Dose Rate discourse, we have different results since. The graph is possible to see in figure 13, the values go to zero quite quickly, about after 1.20470E-02 years.



Figure 13: Dose Rate during cooling time after a radiation of 1 year.

Surely the Dose Rate has an interesting trend, very similar to the one seen in figure 11 with the graph of the activity. Initially the curve decreases due to the disappearance of the most unstable nuclides, especially the disappearance of radionuclide ¹⁶N, which as seen in section 2.3.1. has a half-life of 7.13 seconds, has a big impact on the System as it is the largest contributor to the dose rate just finished the irradiation phase, as can be seen in Figure 9. Subsequently it is quite constant, at about one hour it goes to peak until you get to zero. The peak is caused by the disappearance of tritium from our system.

The figure 14 is the one relating to the Dose Rate after a radiation of ten years, indeed it shows the most important news, as the time before zeroing increases considerably compared to the previous case. Another interesting peculiarity is the fact that for the first nine hours the value is identical to that of the previous case. Unlike the previous case we see how for the first five years, in addition to ¹⁰Be, ¹⁴C and ³H, there is also ²²Na, which has a half-life of about two and a half years, that contributes considerably. Once this nuclide decays the trend remains more constant.



Figure 14: Dose Rate after 10 years compared with the one of 1 year during cooling time.

2.3.3. Decay Heat

The last graph that is analyzed is the one related to the Heat Output shown in figure 15.



Figure 15: Decay Heat output during cooling time.

Unlike the analysis of the activity and the dose rate in the first phase of cooling the trend is immediately decreasing, consistent with the other two instead there is a constant central portion due to the disappearance of the most unstable nuclides and finally there is a very rapid decrease caused by the decay of tritium. Finally, in figure 16 there is the heat output after ten years of radiation. It does not show major variations from that of the previous case except for a different order of magnitude relative to the greater exposure to the flow.



Figure 16: Decay Heat output after 10 years compared with the one of 1 year during cooling time.

A peculiarity lies in the similarity of the values in the first stroke. Subsequently, however, the values deviate, until they have the greatest difference in the last period analysed.

2.4. FLiBe with impurities

The second part of the analysis focuses on FLiBe with present impurities, in the literature there are several examples of impurities related to FLiBe, the difference is made by the use that is made of it. As for the use in the ARC reactor I found the following composition:

| | F | Li | Be | Fe | Cr | Ni | Na | Mg | Al |
|-------|-------|------|-----|--------|--------|--------|-------|-------|--------|
| FLiBe | 76.79 | 14.1 | 9.0 | 0.0004 | 0.0003 | 0.0001 | 0.089 | 0.018 | 0.0033 |

Table 2.4. Mass percentage contribution of each element to the FLiBe

Through the FISPACT-II program I analyzed the behavior of the material following a flux of $10^{15} \frac{neutrons}{cm^2 s}$ for one year and for ten years, like the previous case with pure FLiBe. The section on irradiation was first viewed and then the one on cooling. Below is the composition of FLiBe at the beginning of radiation:

| | ATOMS | GRAM- | CRAMS | BETA CURIES- MeV | LW. | GAMMA CURIES- MeV | LW | ALPHA CURIES- MeV | ĿW |
|----|----------|----------|----------|------------------------|----------|-------------------------|----------|-------------------------|----------|
| | ATOMS | ATOMS | URAMS | NIC V | KVV | WIC V | KV | NIC V | K VV |
| | | | | | | | | | |
| Li | 2.03E+29 | 3.37E+05 | 2.34E+06 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Be | 9.98E+28 | 1.66E+05 | 1.49E+06 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| F | 4.04E+29 | 6.71E+05 | 1.27E+07 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Na | 3.87E+26 | 6.42E+02 | 1.48E+04 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Mg | 7.40E+25 | 1.23E+02 | 2.99E+03 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Al | 1.22E+25 | 2.03E+01 | 5.48E+02 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Cr | 5.52E+23 | 9.16E-01 | 4.77E+01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Fe | 7.16E+23 | 1.19E+00 | 6.64E+01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |

Table 2.5. Composition by elements of the FLiBe with impurities

Now we will show the new nuclides formed divided, as in the previous paragraphs, between stable and non-stable, since there are impurities in our compound also the number of new nuclides is more than in the previous case, there are in fact one hundred and fifty-one nuclides compared to forty-two of pure FLiBe.

| | Table 2.6. Different isotopes formed during the radiation | | | | | | | | | |
|-----------|--|--|--|--|--|--|--|--|--|--|
| STABLE: | H1-O17-P31-V51-H2-O18-Ar38-Cr52-He3-F19-Ar40-Cr53-H4- Ne20-K41-Cr54-Li6-Ne21-Ca42-Mn55-Li7-Ne22-Ca43-Fe54-Be9- Na23-Ca44-Fe56-B10-Mg24-Ca46-Fe57-B11-Mg25-Sc45-F58-C12- Mg 26-Ti46-Co59-C13-Al27-Ti47-Ni60-N14-Si28-Ti48-Ni61-N15- Si29-Ti49-Ni62-O16-Si30-Ti50 | | | | | | | | | |
| UNSTABLE: | H3-He6-Li8-Be10-Be11-B12-B13-C14-C15-C16-N13-N16-N17- N18-N19-O19-O20-F17-F18-F20-F21-F22-F23-Ne 23-Ne24-Na21- Na22-Na24-Na25-Na26-Na 27-Mg27-Mg28-Al25-Al26-Al26m- Al28-Al29-Ar39-Ar42-Ar43-K42-K43-K45-K46-Ca41-Ca45-Ca47- Ca48-Sc44-Sc44m-Sc46-Sc46m-Sc47-Sc48-Sc49-Sc50-Sc50m- Ti51-Ti52-V48-V49-V50-V52-V53-V54-Cr49-Cr50-Cr51-Cr55- Cr56-Mn51-Mn52-Mn52m-Mn53-Mn54-Mn56-Mn 57-Mn58- Mn58m-Mn59-Fe52-Fe53-Fe55-Fe59-Fe60-Fe61-Co56-Co57-Co58- Co58m-Co60-Co60m-Co61-Ni58-Ni59 | | | | | | | | | |

2.4.1. Formation of nuclides and percentage contribution of them to activity, dose rate and Heat during the irradiation phase

Compared to the previous case, the percentages of nuclides will also change, which will affect the activity, the dose rate and the heat output. As can be seen from the following graph, the greatest contribution is made by ²⁶N and no longer by ³H as in the previous case. ³H which contributes about 3% compared to 39% of pure FLiBe. Total activity produced amounted to 1.2561E+19 Bq.



Figure 17: Percentage contribution of each nuclide to the Activity.

In the case of heat, on the other hand, 26 N remains the largest contributor but the percentage is lowered from about 80% to about 71%. The total heat produced is equal to 7.6033E+03 kW.



Figure 18: Percentage contribution of each nuclide to the Decay Heat produced.

Also in the last graph we can see how the greatest contribution during irradiation is given by the N16, as in the case of pure FLiBe. In this case the percentage of the contribution is about the same. The total amount of the Dose Rate is 5.9282E+05 Sv/hr.



Figure 19: Percentage contribution of each nuclide to the Dose Rate.

A final aspect analysed is the composition of the material by elements, useful to understand all the new elements that have formed during irradiation, the results are shown in the table 2.6.

| | | GRAM- | | BETA CURIES- | | GAMMA CURIES- | | ALPHA CURIES- | |
|----|----------|----------|----------|-----------------|----------|------------------|----------|------------------|----------|
| | ATOMS | ATOMS | GRAMS | MeV | kW | MeV | kW | MeV | kW |
| | | | | | | | | | |
| Н | 7.39E+26 | 1.23E+03 | 2.75E+03 | 1.12E+05 | 6.61E-01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| He | 2.56E+27 | 4.26E+03 | 1.70E+04 | 5.95E+07 | 3.53E+02 | 2.15E+05 | 1.28E+00 | 0.00E+00 | 0.00E+00 |
| Li | 2.03E+29 | 3.37E+05 | 2.34E+06 | 9.99E+05 | 5.92E+00 | 5.31E+03 | 3.15E-02 | 5.03E+05 | 2.98E+00 |
| Be | 9.93E+28 | 1.65E+05 | 1.49E+06 | 8.06E+01 | 4.78E-04 | 2.46E+01 | 1.46E-04 | 6.29E-01 | 3.73E-06 |
| В | 4.27E+22 | 7.10E-02 | 7.81E-01 | 2.76E+06 | 1.64E+01 | 3.97E+04 | 2.35E-01 | 2.91E+03 | 1.72E-02 |
| С | 8.13E+23 | 1.35E+00 | 1.76E+01 | 7.03E+05 | 4.17E+00 | 8.92E+05 | 5.29E+00 | 2.42E-02 | 1.43E-07 |
| Ν | 1.11E+27 | 1.85E+03 | 2.77E+04 | 3.36E+08 | 1.99E+03 | 5.79E+08 | 3.43E+03 | 5.67E+03 | 3.36E-02 |
| 0 | 5.08E+26 | 8.44E+02 | 1.46E+04 | 9.26E+07 | 5.49E+02 | 5.44E+07 | 3.23E+02 | 0.00E+00 | 0.00E+00 |
| Ne | 1.30E+25 | 2.16E+01 | 4.35E+02 | 2.15E+05 | 1.27E+00 | 1.97E+04 | 1.17E-01 | 0.00E+00 | 0.00E+00 |
| F | 4.02E+29 | 6.68E+05 | 1.27E+07 | 4.72E+07 | 2.80E+02 | 1.07E+08 | 6.34E+02 | 6.60E-02 | 3.91E-07 |
| Na | 3.86E+26 | 6.40E+02 | 1.47E+04 | 6.96E+04 | 4.13E-01 | 4.81E+05 | 2.85E+00 | 1.94E-08 | 1.15E-13 |
| Mg | 7.39E+25 | 1.23E+02 | 2.98E+03 | 4.32E+03 | 2.56E-02 | 5.52E+03 | 3.27E-02 | 0.00E+00 | 0.00E+00 |
| Al | 1.22E+25 | 2.02E+01 | 5.46E+02 | 7.06E+02 | 4.19E-03 | 1.01E+03 | 6.00E-03 | 0.00E+00 | 0.00E+00 |
| Si | 6.58E+20 | 1.09E-03 | 3.06E-02 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Р | 2.44E+06 | 4.05E-18 | 1.26E-16 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Ar | 1.02E+09 | 1.69E-15 | 7.10E-14 | 9.89E-08 | 5.86E-13 | 1.07E-07 | 6.34E-13 | 0.00E+00 | 0.00E+00 |
| Κ | 3.70E+08 | 6.14E-16 | 2.62E-14 | 5.67E-07 | 3.36E-12 | 6.96E-07 | 4.13E-12 | 0.00E+00 | 0.00E+00 |
| Ca | 7.83E+15 | 1.30E-08 | 5.98E-07 | 1.45E-04 | 8.62E-10 | 4.34E-04 | 2.57E-09 | 0.00E+00 | 0.00E+00 |
| Sc | 7.21E+14 | 1.20E-09 | 5.73E-08 | 4.32E-02 | 2.56E-07 | 2.76E-01 | 1.64E-06 | 0.00E+00 | 0.00E+00 |
| Ti | 1.46E+20 | 2.42E-04 | 1.19E-02 | 9.44E-01 | 5.59E-06 | 3.96E-01 | 2.35E-06 | 0.00E+00 | 0.00E+00 |
| v | 9.41E+20 | 1.56E-03 | 7.96E-02 | 3.14E+02 | 1.86E-03 | 4.24E+02 | 2.52E-03 | 0.00E+00 | 0.00E+00 |
| Cr | 5.51E+23 | 9.15E-01 | 4.76E+01 | 4.11E+00 | 2.44E-05 | 2.32E+01 | 1.37E-04 | 0.00E+00 | 0.00E+00 |
| Mn | 7.21E+20 | 1.20E-03 | 6.51E-02 | 3.73E+02 | 2.21E-03 | 8.16E+02 | 4.84E-03 | 0.00E+00 | 0.00E+00 |
| Fe | 7.15E+23 | 1.19E+00 | 6.63E+01 | 1.94E+00 | 1.15E-05 | 1.79E+00 | 1.06E-05 | 0.00E+00 | 0.00E+00 |
| Co | 5.90E+17 | 9.80E-07 | 5.77E-05 | 1.20E-04 | 7.09E-10 | 1.62E-03 | 9.61E-09 | 0.00E+00 | 0.00E+00 |
| Ni | 1.80E+13 | 3.00E-11 | 1.80E-09 | 3.30E-16 | 1.96E-21 | 1.82E-16 | 1.08E-21 | 0.00E+00 | 0.00E+00 |

Table 2.6. Composition by elements of the FLiBe with impurities after a year of irradiation

2.5. Cooling phase analysis for different radiation times

The analysis again focuses on different time intervals, starting from 1 second up to 1000 years, below are the different time steps analyzed: 1 Seconds, 3 Seconds, 5 Seconds, 25 Seconds, 40 Seconds, 1 Minutes, 7 Minutes, 12 Minutes, 30 Minutes, 45 Minutes, 1 Hours, 5 Hours, 9 Hours, 17 Hours, 1 Days, 2 Days, 5 Days, 12 Days, 20 Days, 50 Days, 120 Days, 230 Days, 1 Years, 3 Years, 8 Years, 15 Years, 50 Years, 100 Years, 200 Years, 500 Years, 1000 Years.

2.5.1. Activity

A first big difference lies precisely in the impurities, these in fact, generate nuclides that remain in the system for over a thousand years. Nuclides that contribute appreciably to the activity at the dose rate and heat. Below are the nuclides that after a thousand years have not yet decayed and contribute to the activity:

- ¹⁴C, 9.75E+11 Bq
- ¹⁰Be, 3.03E+08 Bq
- ²⁶Al, 1.99E+07 Bq
- ⁵³Mn, 9.55E+05 Bq
- ⁶⁰Fe, 1.81E+00 Bq
- ⁶⁰Co, 1.81E+00 Bq
- ⁵⁹Ni, 2.60E-03 Bq
- ⁵⁰V, 2.59E-07 Bq
- ⁴¹Ca, 8.83E-08 Bq
- ⁵⁰Cr, 3.42E-08 Bq
- ⁴⁸Ca, 9.85E-17 Bq
- ⁵⁸Ni, 1.04E-22 Bq

Although the contribution of many of them is relatively small, it is still a big difference compared to the case of pure FLiBe where only ¹⁴C and ¹⁰B did not decay in the period analyzed.

A very important aspect to analyze is the trend of tritium within the system, this remains about constant for the first one hundred and twenty days after which it falls very quickly until it resets shortly after two hundred years. The trend can be seen in figure 21.



Figure 20: Total Activity and Tritium's activity during cooling time after a radiation of 1 year.

The graph gives us the opportunity to immediately notice that, in a first period up to about 10⁻³ years, the trend of activity is higher than that of tritium, this due to the presence of the many instable radionuclides that, with an irradiation period longer, contribute more to the activity like: ¹⁸F, with a half-life of one hundred and ten minutes, ¹⁹O, with a half-life of 26.470 seconds and ⁶He that decay in eight hundred and ten milliseconds. Taking any figure representing the trend of the activity over time with pure FLiBe is possible to see that this had the same tendency as that of tritium. Once the contribution of tritium begins to decrease, the trend of the curve also plummets but does not reset as there are several nuclides that show activity, the main ones being ¹⁰Be and ¹⁴C. Figure 21 shows the comparison between the activity after ten years of irradiation and the one after one year.



Figure 21: Total Activity after 10 years compared with the one of 1 year during cooling time.

From Figure 21 it is possible to see how impurities most influence the first periods following the end of irradiation where the contribution of the most unstable radionuclides is greater.

2.5.2. Dose Rate

The figure 22 represents the Dose Rate. It has a very interesting trend compared to that of the case of pure FLiBe. The first important difference can be observed is that the FLiBe with impurities affects the Dose Rate until the end of the period analyzed. In the graph, moreover, it's possible to see three different decreases, the first is due to the disappearance of the radionuclide plus ¹⁶N which, at the end of the irradiation phase is the largest contributor to the Dose Rate as can be seen in figure 20 but has a very low half-life of about seven seconds. The second decrease is due to the disappearance of other very unstable radionuclides such as ¹⁸F which has a half-life of a hundred minutes and, once ¹⁶N is gone, has become the largest contributor to the dose rate. The last decrease is due to the disappearance of tritium from the system.



Figure 22: Dose Rate during cooling time after a radiation of 1 year.

Figure 23 shows us the trend of the dose rate after ten years of irradiation compared to that of only one year. In a first period they have the same trend that then changes when the most unstable radionuclides have stopped affecting the system. Finally, it precipitates when tritium stops contributing.



Figure 23: Dose Rate after 10 years compared with the one of 1 year during cooling time.

2.5.3. Decay Heat

The graph of the decay heat represented in figure 24 describes a trend very similar to that seen in figures 23 with regard to the Dose Rate. Here too we see three decreases due to the disappearance of the most unstable radionuclides. In the first decrease we come a greater inclination than that of the Dose Rate as the ¹⁹O affects more the heat of decay than the dose rate, more influenced instead by the ¹⁸F, the isotope of the oxygen, however, is characterized by a very small half-life of 26.470 seconds instead of the isotope of fluorine which has a half-life of one hundred and ten minutes.



Figure 24: Decay Heat output during cooling time after a radiation of 1 year.

The graph of the decay heat after a ten-year irradiation is shown in figure 25. Again, the trend is very similar to that of the dose rate where we have a first period strongly influenced by the most unstable radionuclides in which the curves overlap the one of one year of radiation. Subsequently, the decay heat curve after ten years remains higher as it is influenced by the longer period of irradiation.



Figure 25: Decay Heat output after 10 years compared with the one of 1 year during cooling time.

3. Beryllium

3.1. Introduction

The purpose of this section is to analyze more one of the constituent elements of the FLiBe: the Beryllium. This element when activated produces tritium by permutation. The tritium can be recovered and used as fuel for the fusion reactor. The main Beproducing countries in 2012 were the USA (88%), China (9%) and Mozambique (1%) [14]. The problem of Beryllium is that at the time of extraction it can possess parasitic uranium, a value that affects, as we will see later, the performance of the reactor. For example, the one extracted in the US is not admissible because it has a concentration of about 150 wppm of Uranium. Much lower percentages are required, for example for ITER a concentration of up to 30 wppm is allowed [16]. In figure 26 is possible to see the problems related to the presence of uranium consist in the formation of new nuclides once it is irradiated, the most harmful that is produced is Plutonium.



Figure 26: Activation processes for Uranium as impurity in the Beryllium

The production of this is well regulated by resuming in fact some of the French regulations indicates the following [15][15][16]:

- substances with a radioactivity lower than 100 kBq/kg should not enter into consideration for the classification of the installation on environment protection grounds (ICPE).
- for the radioactive waste storage in the Centre de Stockage de l'Aube (CSA), that is an intermediate level short-lived waste (ILW) disposal site, the French

radioactive waste management agency, ANDRA, has a limit of total alpha emitting radio nuclides at 300 years cooling time of 3700 kBq/kg.

- the limit for the FMA-VC (low and intermediate level waste, short-lived isotopes) fissile materials concentration in conditioned wastes is of 0.1 g/l.
- the limits for the MAVL (ILW, long-lived) are not yet available.

In the first part of the analysis, 1 kg of pure beryllium will be irradiated with different percentages of uranium present. The irradiation has been done with a flux of $10^{15} \frac{neutrons}{cm^2 s}$ for a year and then for ten years, resuming the procedure seen in the previous paragraphs. The different percentages of Uranium analyzed are: 0.5 wppm, 1 wppm, 2.5 wppm, 5 wppm, 10 wppm, 30 wppm, 50 wppm and 100 wppm. Subsequently, several commercial Beryllium will be analyzed, containing different impurities, including Uranium, with the same irradiation methods previously described.

3.2. Beryllium with only uranium as impurity, activation analysis

In this section I analyses one kilogram of beryllium with only uranium as impurities. The material will be subjected to a flux of $10^{15} \frac{neutrons}{cm^2 s}$ for different time of radiation, initially one year later for ten. Different percentages of uranium will be studied. Of interest is the comparison between beryllium containing percentages below 30 wppm with that with higher percentages. It was made also an evaluation of the production of Plutonium in our system.

| | ATOMS | GRAM- ATOMS | GRAMS | BETA CURIES- MeV | kW | GAMMA CURIES- MeV | kW | ALPHA CURIES- MeV | kW |
|----|----------|----------------|----------|------------------------|----------|-------------------------|----------|-------------------------|----------|
| | | | | | | | | | |
| Be | 6.68E+25 | 1.11E+02 | 1.00E+03 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| U | 1.27E+17 | 2.10E-07 | 5.00E-05 | 4.43E-13 | 2.62E-18 | 1.68E-13 | 9.96E-19 | 1.54E-10 | 9.15E-16 |

Table 3.1. Primary configuration of the Beryllium analysis

In the table 3.1. is possible to see the configuration before the irradiation of the case with 0.05 wppm of uranium inside the system, as can be noted uranium is not stable, so they are already emitters of radiation.

3.2.1. One year of irradiation

The first example analysed is the case with 0.05 wppm of uranium. Although the irradiation period is relatively small, the presence of uranium has allowed us to develop a very high number of nuclides within our system, in fact we have gone from 4 starting to 70 during the irradiation phase. Most of them are unstable, in fact only 5.71% of them are stable. Of relevance are the nuclides formed directly from Uranium such as ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am, ²⁴²Am, ^{242m} Am, ²⁴³Am, ²⁴⁴Am and ²⁴²Cm. The pattern of generation of these isotopes particularly harmful to our system is reported in the previous paragraph. Consistent with table 3.2. is possible to see how most of these isotopes decay into beta, the FISPACT-II program confirms this trend by giving us the values of the heat produced by each type of decay and the total one:

- Total alpha heat production: 1.40657E-07 kW
- Total beta heat production: 1.64100E-01 kW
- Total gamma heat production: 5.93162E-04kW

The heat produced by beta is the most impactful in our system.

A big difference compared to the studies for FLiBe consists in the long permanence of the isotopes formed by Uranium, these in fact have very long decay times, an example the ²³⁹Pu has a half-life of 24.100 years. This is also confirmed by the program where we can see how after a thousand years from the end of irradiation there forty-one isotopes are still, many of which are very radioactive.

Thanks to the FISPACT-II program it was possible to see another big difference compared to the activation of FLiBe, in fact, due to the presence of Uranium, there are fissions within the system, a phenomenon that previously did not happen. This is very important as it generates new elements and heat that can be harmful to the system.

A final aspect to be analysed is the composition of the material by elements, useful to understand all the new elements that have formed during irradiation:

| | | GRAM- | | BETA CURIES- | | GAMMA CURIES- | | ALPHA CURIES- | |
|----|----------|----------|----------|-----------------|----------|------------------|----------|------------------|----------|
| | ATOMS | ATOMS | GRAMS | MeV | kW | MeV | kW | MeV | kW |
| | | | | | | | | | |
| Н | 8.87E+21 | 1.47E-02 | 4.44E-02 | 2.43E+00 | 1.44E-05 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| He | 5.96E+23 | 9.90E-01 | 3.96E+00 | 2.77E+04 | 1.64E-01 | 1.00E+02 | 5.93E-04 | 0.00E+00 | 0.00E+00 |
| Li | 2.93E+22 | 4.87E-02 | 3.08E-01 | 4.71E-02 | 2.79E-07 | 2.50E-04 | 1.48E-09 | 2.37E-02 | 1.41E-07 |
| Be | 6.65E+25 | 1.10E+02 | 9.95E+02 | 1.38E-06 | 8.19E-12 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| В | 3.20E+12 | 5.31E-12 | 5.33E-11 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Pb | 2.37E+07 | 3.94E-17 | 8.19E-15 | 1.00E-11 | 5.96E-17 | 8.62E-12 | 5.11E-17 | 1.34E-21 | 7.92E-27 |
| Ra | 1.50E+06 | 2.49E-18 | 5.59E-16 | 2.98E-13 | 1.76E-18 | 9.05E-13 | 5.37E-18 | 3.57E-10 | 2.12E-15 |
| Th | 1.96E+11 | 3.26E-13 | 7.54E-11 | 7.91E-08 | 4.69E-13 | 7.79E-08 | 4.62E-13 | 3.56E-10 | 2.11E-15 |
| Ра | 1.76E+09 | 2.92E-15 | 6.75E-13 | 1.73E-07 | 1.03E-12 | 3.16E-07 | 1.88E-12 | 1.40E-13 | 8.28E-19 |
| U | 1.25E+17 | 2.07E-07 | 4.92E-05 | 5.47E-04 | 3.24E-09 | 1.55E-04 | 9.22E-10 | 9.30E-10 | 5.51E-15 |
| Np | 8.30E+14 | 1.38E-09 | 3.27E-07 | 2.65E-04 | 1.57E-09 | 1.99E-04 | 1.18E-09 | 1.11E-09 | 6.56E-15 |
| Pu | 1.16E+15 | 1.93E-09 | 4.61E-07 | 3.68E-09 | 2.18E-14 | 5.13E-09 | 3.04E-14 | 1.07E-06 | 6.36E-12 |
| Am | 4.91E+08 | 8.16E-16 | 1.97E-13 | 2.64E-14 | 1.57E-19 | 1.85E-14 | 1.10E-19 | 3.74E-12 | 2.22E-17 |
| Cm | 2.72E+06 | 4.51E-18 | 1.09E-15 | 3.68E-14 | 2.18E-19 | 4.97E-15 | 2.94E-20 | 2.24E-11 | 1.33E-16 |

Table 3.2. Composition by elements of the Be with U after a year of irradiation

As can be seen from table 3.2. during irradiation, several elements were formed that now make up the system, passing from the original two to twenty-one. An interesting aspect is the great presence of He within the system, the isotope ⁶He is also the largest contributor to the activity producing 97.65%, to the dose rate with 100% and to the heat with 99.9%.

The percentage value of the contribution to the activity is lower because the Beryllium, as mentioned in paragraph 1, when it is activated produces Tritium which has a high activity, in fact 3 H contributes 2.35% to the total activity.

3.2.2. Comparison of different percentages of uranium

In this paragraph different aspects have been compared of the samples examined, the only difference between the different components is the percentage of uranium within 1 kg of beryllium. Among the aspects that we will be compare are:

- The number of new nuclides
- The Activity
- The Dose Rate
- The Heat, divided for decay
- The Tritium's production
- The Number of fissions

| | | 0.5 wppm | 1 wppm | 2.5 wppm | 5 wppm | 10 wppm | 30 wppm | 50 wppm | 100 wppm |
|-----------------------|-------------|----------|----------|----------|----------|----------|----------|----------|----------|
| NUMBER OF NUCLIDES | | 70 | 71 | 73 | 75 | 77 | 83 | 85 | 86 |
| ACTIVITY [Bq] | | 6.72E+14 |
| TRITIUM [Grams] | | 4.43E-02 |
| DOSE I [Sv/l | RATE hr] | 8.00E+02 | 8.01E+02 |
| | A | 1.41E-07 | 1.42E-07 |
| HEAT [Kw] | В | 1.64E-01 |
| | G | 5.93E-04 | 5.94E-04 |
| NUMBER OF FISSIONS | | 3.71E+11 | 7.41E+11 | 1.85E+12 | 3.71E+12 | 7.41E+12 | 2.22E+13 | 3.71E+13 | 7.41E+13 |

Table 3.3. Comparison of data for different percentage of uranium

Thanks to the values shown in the table 3.3. is possible to make some deductions, first of all the values remain almost constant. There are very small variations in the values of the Dose Rate and the decay heat, especially those due to alpha and gamma decays tend to increase as the percentage of Uranium in the system increases. The production of tritium remains constant also because it is due to the activation of beryllium and therefore is not affected by the presence of the U. Also the activity, which, as it is specified previously is conditioned by tritium, remains constant. The data that have the most significant variations are the number of nuclides and the number of fissions. This last value is a count of the change of the number of nuclides that may undergo fission from the number in the initial inventory.

In conclusion of this paragraph, an irradiation of one year is not enough to get appreciable results to quantify the effect of parasitic uranium inside the beryllium.

3.2.3. Radiation with a flux equal to 1e+15 for ten years

In this section the analysis was made on thebehaviour of beryllium with small percentages of Uranium inside during a stationary irradiation for ten years. Since the period of irradiation is longer, it is expected an impact of the Uranium greater on the system than in the previous case. As a general example has been examined the case with 0.05 wppm of Uranium, although it is the case with the lowest percentage of U its effects are already appreciable. The first significant amount is the number of nuclides that are formed during irradiation, i.e., 101. Of these, as in the previous case, most are unstable and only 13.86% are stable. Impacting certainly are the isotopes of plutonium of americium and curium which are very toxic, moreover those of plutonium and curium have a very long half-life, therefore, they are difficult to dispose of. Other isotopes such as ^{242m}Am, a metastate from Americium, also has relatively long decay times, about one hundred and forty-one years.

As for the heat of decay produced, the values are like those with a year of irradiation:

- Total alpha heat production: 1.37473E-06 kW
- Total beta heat production: 1.57501E-01 kW
- Total gamma heat production: 5.68834E-04 kW

Again, the heat produced by the decay in beta is the most impactful.

Another much higher value than in the previous case is the number of fissions within the system we went from 3.71E+11 to 3.55089E+12. In table 3.4. all the new elements are reported formed during irradiation:

| | ATOMS | GRAM- ATOMS | GRAMS | BETA CURIES- MeV | kW | GAMMA CURIES- MeV | kW | ALPHA CURIES- MeV | kW |
|----|----------|----------------|----------|------------------------|----------|-------------------------|----------|-------------------------|----------|
| | | | | | | | | | |
| Н | 9.74E+22 | 1.62E-01 | 4.65E-01 | 2.48E+01 | 1.47E-04 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| He | 5.87E+24 | 9.75E+00 | 3.90E+01 | 2.65E+04 | 1.57E-01 | 9.60E+01 | 5.69E-04 | 0.00E+00 | 0.00E+00 |
| Li | 2.66E+23 | 4.42E-01 | 2.80E+00 | 4.60E-01 | 2.73E-06 | 2.45E-03 | 1.45E-08 | 2.32E-01 | 1.37E-06 |
| Be | 6.37E+25 | 1.06E+02 | 9.54E+02 | 1.94E-05 | 1.15E-10 | 1.82E-06 | 1.08E-11 | 4.65E-08 | 2.75E-13 |
| В | 2.58E+14 | 4.28E-10 | 4.29E-09 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| С | 1.76E+07 | 2.92E-17 | 3.51E-16 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Hg | 2.44E+05 | 4.06E-19 | 8.28E-17 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Tl | 1.21E+05 | 2.02E-19 | 4.19E-17 | 7.26E-09 | 4.30E-14 | 4.20E-08 | 2.49E-13 | 0.00E+00 | 0.00E+00 |
| Pb | 1.09E+11 | 1.81E-13 | 3.76E-11 | 6.40E-09 | 3.79E-14 | 5.09E-09 | 3.01E-14 | 4.98E-18 | 2.95E-23 |
| Bi | 1.07E+08 | 1.78E-16 | 3.72E-14 | 1.74E-08 | 1.03E-13 | 3.70E-09 | 2.20E-14 | 7.66E-08 | 4.54E-13 |
| Ро | 3.72E+07 | 6.18E-17 | 1.30E-14 | 8.08E-18 | 4.79E-23 | 5.83E-16 | 3.45E-21 | 3.06E-10 | 1.81E-15 |
| Rn | 1.03E+05 | 1.71E-19 | 3.76E-17 | 5.02E-13 | 2.97E-18 | 2.17E-11 | 1.29E-16 | 2.21E-07 | 1.31E-12 |
| Ra | 6.80E+08 | 1.13E-15 | 2.53E-13 | 1.76E-10 | 1.04E-15 | 5.29E-10 | 3.14E-15 | 2.07E-07 | 1.23E-12 |
| Ac | 1.60E+07 | 2.66E-17 | 6.05E-15 | 2.69E-13 | 1.59E-18 | 1.63E-13 | 9.69E-19 | 5.59E-11 | 3.31E-16 |
| Th | 1.79E+12 | 2.97E-12 | 6.85E-10 | 7.39E-08 | 4.38E-13 | 6.67E-08 | 3.95E-13 | 1.98E-07 | 1.18E-12 |
| Ра | 2.45E+11 | 4.08E-13 | 9.42E-11 | 1.62E-07 | 9.58E-13 | 3.02E-07 | 1.79E-12 | 2.41E-11 | 1.43E-16 |
| U | 1.08E+17 | 1.79E-07 | 4.26E-05 | 4.67E-04 | 2.77E-09 | 1.35E-04 | 8.02E-10 | 3.41E-07 | 2.02E-12 |
| Np | 6.41E+15 | 1.06E-08 | 2.52E-06 | 2.76E-04 | 1.64E-09 | 3.15E-04 | 1.87E-09 | 8.63E-09 | 5.12E-14 |
| Pu | 1.22E+16 | 2.02E-08 | 4.83E-06 | 1.88E-07 | 1.11E-12 | 1.30E-07 | 7.68E-13 | 7.11E-05 | 4.22E-10 |
| Am | 3.57E+12 | 5.93E-12 | 1.43E-09 | 2.50E-08 | 1.48E-13 | 8.19E-09 | 4.85E-14 | 2.68E-08 | 1.59E-13 |
| Cm | 6.86E+10 | 1.14E-13 | 2.76E-11 | 9.48E-10 | 5.62E-15 | 3.23E-10 | 1.92E-15 | 5.41E-07 | 3.21E-12 |

Table 3.4. Composition by elements of the Be with U after ten years of irradiation

From the table 3.4. there is a big difference compared to the previous case: ⁶He has a much lower impact on activity going from 97.65% to 79.63%, this is since with a greater irradiation time the amount of tritium produced following the activation of Beryllium has increased, influencing this parameter more.

3.2.4. Comparison of different percentages of uranium

Also, in this paragraph the purpose is the comparison between the different percentages of uranium to be able to compare the following parameters during an irradiation of ten years:

- The number of new nuclides
- The Activity
- The Dose Rate
- The Heat, divided for decay
- The Tritium's production
- The Number of fissions

| | | 0.5 wppm | 1 wppm | 2.5 wppm | 5 wppm | 10 wppm | 30 wppm | 50 wppm | 100 wppm |
|-----------------------|-------------|----------|----------|----------|----------|----------|----------|----------|----------|
| NUMBER OF NUCLIDES | | 101 | 105 | 109 | 112 | 115 | 118 | 123 | 125 |
| ACTIVITY [Bq] | | 7.90E+14 |
| TRITIUM [Grams] | | 4.52E-01 |
| DOSE F [Sv/h | RATE hr] | 7.68E+02 |
| | A | 1.37E-06 | 1.38E-06 | 1.38E-06 | 1.38E-06 | 1.38E-06 | 1.40E-06 | 1.42E-06 | 1.46E-06 |
| HEAT [Kw] | В | 1.58E-01 |
| | G | 5.69E-04 |
| NUMBER OF FISSIONS | | 3.55E+12 | 7.10E+12 | 1.78E+13 | 3.55E+13 | 7.10E+13 | 2.13E+14 | 3.55E+14 | 7.10E+14 |

Table 3.5. Comparison of data for different percentage of uranium

Consistent table 3.3. of the previous case, the values that are most influenced by the presence of uranium are the number of fissions and the number of nuclides within the system. The production of tritium remains almost constant this implies that the activity also remains more or less on the same values. As far as heat is concerned, the only one that has an appreciable increase is that linked to decay into alpha.

3.2.5. Cooling phase

Consistent with the study done in the FLiBe different time periods have been analysed from the moment the irradiation ends which are: 1 Seconds, 3 Seconds, 5 Seconds, 25 Seconds, 40 Seconds, 1 Minutes, 7 Minutes, 12 Minutes, 30 Minutes, 45 Minute s, 45 Minutes, 1 hours, 5 hours, 9 hours, 17 hours, 1 days, 2 days, 5 days, 12 days, 20 days, 50 days, 120 days, 230 days, 1 years, 3 years, 8 years, 15 years, 50 years, 100 years, 200 years, 500 years, 1000 years.

The first aspect evaluated is the variation of the number of nuclides within the system. The next graph shows the case of irradiation for one year with 0.5 wppm of uranium.

The presence of uranium involves the production of isotopes with a very long half-life that then remain in the system influencing its characteristics. In the FLiBe missing this element at the end of the cooling phase there were only two nuclides here instead there are over fifty. Figure 27 shows the change of the number of nuclides inside the system.



Figure 27: Number of nuclides in the system during the cooling phase

There are several interesting factors in this graph, the first is certainly the high number of nuclides after a thousand years, in fact there are still forty-one elements, of which many of them radioactive. Figure 28 shows the dose trends for the different percentages of uranium following irradiation for one year.



Figure 28: Dose rate of the Be with different % of U during the cooling phase after an irradiation of one year

Since activity and heat are not affected by the effect of impurities, as can be seen in Tables 3.3. and 3.5., the study focused more on the Dose Rate graph. This graph is certainly the most interesting one in this part of the Beryllium analysis; in fact, it is evident that the contribution of uranium has consequences. The higher the amount of U corresponds to a higher value of the Dose Rate. It is interesting to note that there is a clear detachment of value for periods longer than a year: when the amount of U is greater than 30 wppm it's possible to see how the value of the Dose Rate has a further increase compared to cases with lower percentages.

3.3. BP-1 activation analysis

The second part of the beryllium analyses was done taking into account the impurities that can be found in Beryllium. As material to be analysed was taken BP-1, 'BP' stay for beryllium pebble. The specifications of "BP-1" shall contain a minimum beryllium content of 99.0 %. The chemical composition of "BP-1" is shown in the table 3.6., and it should be a sphere made by REP (Rotation Electrode Process). This material is one of the major candidates as a neutron multiplier in ITER [15].

| | Be | BeO | Al | Co | Fe | Mg | Si | U |
|------|----|-----|------|-------|-----|------|------|-------|
| BP-1 | 99 | 0.5 | 0.09 | 0.001 | 0.1 | 0.08 | 0.06 | 0.003 |

Table 3.6. Mass percentage contribution of each element to the BP-1

In this analysis the material will be irradiated in the same way as in the previous cases, therefore with a stationary flux of $10^{15} \frac{neutrons}{cm^2 s}$ first for a year and then for ten years. In the cooling phase, different time periods will be evaluated. The FISPACT-II program will be used again to carry out all the analyses. Once the data is loaded on the program, the elements have the following characteristics:

Table 3.6. Composition by elements of the BP-1 before the irradiation

| | ATOMS | GRAM- ATOMS | GRAMS | BETA CURIES- MeV | kW | GAMMA CURIES- MeV | kW | ALPHA CURIES- MeV | kW |
|----|----------|----------------|----------|------------------------|----------|-------------------------|----------|-------------------------|----------|
| | | | | | | | | | |
| Be | 6.62E+25 | 1.10E+02 | 9.90E+02 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| 0 | 1.88E+23 | 3.13E-01 | 5.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Mg | 1.98E+22 | 3.29E-02 | 8.00E-01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Al | 2.01E+22 | 3.34E-02 | 9.00E-01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Si | 1.29E+22 | 2.14E-02 | 6.00E-01 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Fe | 1.08E+22 | 1.79E-02 | 1.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| Co | 1.02E+20 | 1.70E-04 | 1.00E-02 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 | 0.00E+00 |
| U | 1.27E+20 | 2.10E-04 | 5.00E-02 | 4.43E-10 | 2.62E-15 | 1.68E-10 | 9.96E-16 | 1.54E-07 | 9.15E-13 |
| | | | | | | | | | |

Initially the only radioactive element is uranium.

3.3.1. Formation of nuclides and percentage contribution of them to activity, dose rate and Heat during the irradiation phase

Certainly, unlike the previous cases where there was the analysis of the beryllium with only uranium as impurities, the presence of other materials involves variations on the activity, on the dose rate and on the heat. Since among the impurities there is also uranium, it is reasonable to assume to have nuclides with a very long half-life, such as the isotopes of plutonium and curium, as well as high values of the Dose Rate.

The first aspect evaluated is how the various isotopes affect the activity, following the irradiation 214 nuclides were formed and a total activity of 6.69758E + 14 Bq. In figure 29 is shown the contribution of the most important nuclide.



Figure 29: Percentage contribution of each nuclide to the activity.

Practically all the activity is produced by ⁶He, in addition to helium only tritium contributes appreciably to the activity of the system.



Figure 30: Percentage contribution of each nuclide to the heat.

Similar speech for the heat produced where again the nuclide that most influences the values is the ⁶He, unlike the activity the second nuclide that contributes appreciably is the ¹⁶N. The total heat produced is 1.6655E-01 kW.



Figure 31: Percentage contribution of each nuclide to the Dose Rate

In Figure 31 it's possible to see the contribution of the most important nuclides to the dose rate. This is the most interesting graph because there are several nuclides that affect the system, the total value of the dose rate is 6.6825E+03 Sv / hr. The largest contributor is the ¹⁶N then we find ²⁴Na, ⁶He, ⁵⁶Mn and ²⁷Mg that appreciably affect the Dose Rate.

3.3.2. Cooling phase

Also in this case the analysis was made in different periods following the end of the irradiation, the following were analysed: 1 Seconds, 3 Seconds, 5 Seconds, 25 Seconds, 40 Seconds, 1 Minutes, 7 Minutes, 12 Minutes, 30 Minutes, 45 Minutes, 45 Minutes, 1 hours, 5 hours, 9 hours, 17 hours, 1 days, 2 days, 5 days, 12 days, 20 days, 50 days, 120 days, 230 days, 1 years, 3 years, 8 years, 15 years, 50 years, 100 years, 200 years, 500 years, 1000 years.

In figure 32 is represented the activity, given the percentage composition of the previous paragraph it is assumed that in the first time the curve of activity is higher than that of tritium as there are isotopes such as ⁶He that have a great influence on the system.



Figure 32: Activity of the BP-1 and tritium activity during the cooling phase

The figure 32 confirms the assumption, in fact, initially the activity of the system is higher than that of tritium, this trend decreases quickly as the isotopes that affect decay very quickly, 16N has a half-life of 7.13(2) seconds. Subsequently, the tritium curve and the activity curve go together. As in the previous examples, the moment tritium stops contributing, the curve decreases rapidly.

In figure 33 there is the graph of the Dose Rate, given the presence of Uranium in impurities we expect the values to be higher than those of pure beryllium.



Figure 33: Dose Rate of the BP-1 and of the pure beryllium

In figure 33 the graph confirms the hypotheses made before, as seen in the red curve the Dose Rate in the case considered, therefore of BP-1, is much higher, this is due to impurities and the greater presence of Uranium in the system. The trend, on the other hand, is very similar.

The last graph analysed is the one related to heat shown in figure 34. Here instead is done the assumption, starting from the figure 31, to find results quite like the previous case.



Figure 34: Decay heat of the BP-1 and of the pure beryllium

In figure 34 the trend confirms our assumptions, in fact, the curves have very similar values, that of the BP-1 case has a less constant trend as impurities affect the values more.

4. FLiBe with Beryllium containing Uranium

4.1. Material composition and explanation of the analysis

In this chapter a FLiBe containing among the impurities a small percentage of uranium has been analysed. Indeed, it has been assumed beryllium that during the extraction had some parasitic uranium. The amount of U is 30 ppm which is the maximum limit allowed by ITER [15]. In the table 4.1. is reported the composition of the molten salt.

| | F | Li | Be | Fe | Cr | Ni | Na | Mg | Al | U |
|-------|-------|------|-------|--------|--------|--------|-------|-------|--------|-------|
| FLiBe | 76.79 | 14.1 | 8.997 | 0.0004 | 0.0003 | 0.0001 | 0.089 | 0.018 | 0.0033 | 0.003 |

Table 4.1. Mass percentage contribution of each element to the FLiBe

During the activation analysis, the behavior of the nuclides during the irradiation phase was better evaluated. The purpose of this analysis is to investigate some of the most important radionuclides:

- ³H;
- ⁶⁰Co;
- ²³⁹Pu;
- ²⁴¹Am;

These nuclides are important for several reasons: tritium is a key element in nuclear fusion as it can be used as a fuel along with deuterium according to the relation seen in chapter one.

The ⁶⁰Co, on the other hand, is a synthetic radioactive isotope of cobalt metal, it is used as a source of gamma rays with energies of about 1.3 MeV. ²³⁹Pu is a fundamental fissile product for most nuclear weapons. It has a very long half-life of twenty-four thousand and two hundred years and emits alpha radiation. Almost all of the plutonium is of synthetic origin, although very tenuous traces are found in nature in uranium ores. ²³⁹Pu can also be used as a fuel in next-generation nuclear power plants, which burn a uranium-plutonium mixed oxide fuel. The last isotope is the ²⁴¹Am, this is a trans-plutonic element that emits alpha radiation, with a half-life di over four hundred years.

The analysis was carried out mainly during irradiation, the time periods analysed are: 1 seconds, 5 seconds, 1 minutes, 10 minutes, 1 hours, 10 hours, 1 day, 10 days, 100 days, 200 days, 1 year, 5 years and 10 years.

4.2. Evaluation of the behaviour during irradiation of the target nuclides

As can be seen from figure 35, the most produced radionuclide is tritium among those analysed, this is due to the presence of Lithium and Beryllium inside the FLiBe. Indeed, these two elements can produce it through neutron capture. The second element in quantity is ²³⁹Pu, this is produced due to the presence of Uranium in our system, this element will be analysed more in details in the next paragraph.

The last two elements, which are ⁶⁰Co and ²⁴¹Am, are produced in much lower quantities and, above all, much further in time than the previous ones. The first one is characterized by a low half-life equal to 5.27 years, so when the irradiation ends it will disappear relatively quickly compared to the other isotopes analysed.



Figure 35: Behaviour of ³H, ⁶⁰Co, ²³⁹Pu and ²⁴¹Am isotopes during the irradiation time.

The long-lived isotopes are certainly Americium and Plutonium as they have very long half-lives, the first of 432.2 years and the second of 24.1E+03 years. The big problem related to the Americium is the large amount of radiation it emits. A few grams of ²⁴¹Am emit an intense gamma radiation that creates serious problems of exposure to those who have to handle the element.

4.2.1. Plutonium

Given the presence of Uranium in the system, it is useful to monitor the behaviour of Plutonium. There are several isotopes of Plutonium that are formed during the irradiation phase, at the end of the period under examination there are eleven of them. In figure 36 is possible to see the increase of the mass of the different isotopes.



Figure 36: Growth of the various isotopes of Plutonium during irradiation.

They all grow quite constantly during irradiation, obviously the times of birth are different since many are produced by the different transmutations of the same isotope. At the end of irradiation, the largest amount is related to the isotope ²⁴⁰Pu. Figure 38 instead describes the activity.



Figure 37: Trend of the Activity of the different isotopes of Plutonium during irradiation.

It is interesting to note that many of the isotopes examined have a slight decrease around five years of irradiation. However, the activity tends to grow with irradiation. In figure 38 is reported the heat emitted by the different decays: alpha, beta and gamma.



Figure 38: a) Beta decay heat of isotopes during irradiation b) Alpha decay heat of isotopes during irradiation c) Gamma decay heat of isotopes during irradiation

The heat generated by the alpha decay is the higher. The trend in all three graphs is quite similar, in fact is possible to see how the decay heat increases as the irradiation period increases.

The last graph is the one relating to the Dose Rate in figure 39. The purpose is to identify which are the isotopes of Plutonium that most affect this parameter.



Figure 39: Trend of the Dose Rate during the irradiation phase of the different isotopes.

The first thing that is possible to notice is the similarity with Graph 33c in fact the dose rate is mainly influenced by gamma radiation. The quantity at the end of the irradiation can be divided into two macro-groups, there is a part that reaches higher quantities, on the order of 10⁻⁹ Sv/h, while a second group has two orders of magnitude less, therefore about 10⁻¹¹ Sv/h. The Dose Rate of all isotopes however tends to rise as irradiation increases, the only isotope that has an anomalous behaviour is ²⁴³Pu which, after a growing phase, consistent with the other isotopes, decays and disappears from the system no longer contributing to the Dose Rate. This behavior is due to the fact that ²⁴³Pu if it is irradiated with a sufficiently high neutron flux decade in beta turning into ²⁴³Am.

4.2.2. Track fission and actinide products

In this section, actinides and fission products in the irradiation and cooling phases were evaluated. As in the previous case, the FLiBe has been irradiated with a flow of 10^{15} $\frac{neutrons}{cm^2 s}$ for ten years. The composition of FLiBe is that with impurities, including Uranium, for the percentages see in the previous case.

- Actinium, Ac;
- Thorium, Th;
- Protactinium, Pa;
- Uranium, U;
- Neptunium, Np;
- Plutonium, Pu
- Americium, Am;
- Curium, Cm;

The first case analysed is the changing of the mass of the actinides during the irradiation time. The periods analysed are: 1 seconds, 5 seconds, 1 minutes, 10 minutes, 1 hours, 10 hours, 1 day, 10 days, 100 days, 200 days, 1 year, 5 years and 10 years. Below is the graph with the results.

Figure 40 shows the evolution of the actinides inside the system.



Figure 40: Trend of the mass of actinides during the irradiation phase.

The trend of actinides in the system during the irradiation phase is very particular, we see how in the early stages of irradiation they all have moderate growth, excluding uranium which remains constant. After two hundred days actinium, americium and curium appear in the system. All the elements continue to grow until the end of irradiation. At the end the quantity of Uranium decreases a little because part of that turn into Plutonium.

4.3. Activation analysis of the FLiBe

An activation analysis of the FLiBe is also carried out to see the trend of activity, dose rate and decay heat. Given the presence of uranium in the system, the time scale analyzed is longer. In this way it is possible to see well the behavior of some elements that have a long half-life such as plutonium. The following are the periods analyzed: 1 sec, 5 seconds, 1 minutes, 10 minutes, 1 hours, 10 hours, 1 days 10 days, 50 days, 100 days, 200 days, 1 years, 5 years, 10 years, 20 years, 50 years, 100 years, 250 years, 500 years, 750 years, 1000 years, 1500 years, 2000 years, 5000 years, 10000 years.

The first graph analyzed is that of both total and tritium activity alone. The tritium having a half-life of about twelve years at some point will no longer affect the system. Figure 41 shows the activity during the cooling time.



Figure 41: Total activity and tritium activity of the FLiBe during the cooling phase

From Figure 41 at the beginning the total activity is influenced more by unstable radionuclides such as ¹⁶N which however have a very low half-life. Subsequently, the trend follows that of tritium which becomes the largest contributor. After two hundred and fifty years, tritium disappears no longer contributing to the activity, therefore, only isotopes with a sufficiently long half-life remain to affect the system, such as ²⁴³Pu.

The following graph shown in Figure 42 represents the trend of the Dose rate.



Figure 42: Dose rate of the FLiBe during the cooling phase

The dose rate graph has a decreasing trend. Initially it is more influenced by very unstable isotopes such as ¹⁶N or ¹⁸F which have very small half-life. Once decayed, the largest contributor becomes ²²N up to fifty years after the beginning of the cooling phase. With the disappearance of ²²N from the system there is a peak in the trend of the dose rate up to a value around 10⁻⁶ Sv / h. Subsequently, the value stabilizes having as the largest contributor the ²⁶Al which has a half-life of 7.2×10^5 years.

In figure 43 it is possible to see instead the graph of the decay heat.



Figure 43: Decay heat of the FLiBe during the cooling phase

5. Discussion

In this paragraph, the different types of FLiBe studied in the previous chapters have been compared. In this way it was possible to identify the impact of impurities on the system. In Table 5.1. the different compositions of the FLiBe are reported.

In the second part of the discussion the purpose is to analyse and compare two FLiBe salts with a different composition.

| | F | Li | Be | Fe | Cr | Ni | Na | Mg | Al | U |
|--------------------|-------|-------|-------|--------|--------|--------|-------|-------|--------|-------|
| FLiBe ¹ | 76.79 | 14.12 | 9.09 | | | | | | | |
| FLiBe ² | 76.79 | 14.1 | 9.0 | 0.0004 | 0.0003 | 0.0001 | 0.089 | 0.018 | 0.0033 | |
| FLiBe ³ | 76.79 | 14.1 | 8.997 | 0.0004 | 0.0003 | 0.0001 | 0.089 | 0.018 | 0.0033 | 0.003 |

Table 5.1. Mass percentage contribution of each element to the FLiBe

FLiBe¹ is the pure salt, so the composition is made from fluorine, lithium and beryllium alone. While in FLiBe² there are impurities, these derive mainly from lithium and beryllium, in fact the mass percentage of fluorine remains constant. In FLiBe³ there is also parasitic uranium caused by the extraction of beryllium. The amount of uranium is 30 wppm i.e. the limit allowed by law and is the worst case scenario. Indeed the quantity of uranium is less than 20 wppm.

The substances were irradiated with a flux of $10^{15} \frac{neutrons}{cm^2 s}$ for ten years. The analysis was made both for the irradiation phase and for the cooling phase.

Initially the values of 1 kg of FLiBe will be shown, then the typical amount of salt inside a fusion reactor will be studied, i.e. $7*10^5$ kg. In this way the purpose is to evaluate the absolute values that the FLiBe produces after an irradiation period of ten years.

5.1. Activation analysis

During the irradiation phase there are no appreciable differences between the different salts. Figure 44, for example, shows the behavior of the three FLiBe. It is immediately noticeable that this is in no way affected by the presence of impurities.



Figure 44: a) Total activity and b) tritium activity of the three molten salts during the irradiation phase; c) Decay heat and d) Dose Rate during the cooling phase

Even in the cooling phase there are no differences between the different salts. This is because the activity is highly affected by tritium. The production of tritium is not affected by the presence of impurities as can be seen in figure 44b.

The most interesting graphs are those related to the decay heat and the dose rate during cooling time. Figure 44c shows a zoom on the last period of the cooling time of the decay heat. The presence of uranium slightly changes the values of the decay heat. This happens because of the irradiation some isotopes such as ²³⁹Pu have formed, which contribute considerably to the decay heat. These isotopes have a very long half-life as seen in chapter 4 and have a considerable impact on the system. To demonstrate this, after ten thousand years ²³⁹Pu contributes 31.31% of the total decay heat produced, while ²⁴⁰Pu

contributes 1.85%. Figure 47 shows the dose rate during the cooling time. The graph related to the dose rate is the one that brings the most interesting results. The presence of impurities does not allow this value to zero. In fact, in the pure case after ten days from the end of irradiation the dose rate is zero. This happens because initially the largest contributor is ¹⁸F, an isotope with a half-life of 109.771 (20) minutes. Subsequently, the largest contributor becomes the ²²Na, with a half-life of 2.602 years. In the final phase of our analysis, the effects of uranium are again felt, in fact the isotopes of plutonium slightly raise the dose rate values. For example, at the end of irradiation ²³⁹Pu contributes 4.41% of the total dose. A final aspect that could be of interest is the possibility to recycle the FLiBe. There are two different ways to recycling the material, and both are connected to specific limit of the dose rate of contact. The first one is inside the system, the reference value is the HOH (Hands-On Handling) of 1E-05 Sv/h, this limit is the one to which the exposure dose of structural materials must be reduced in order to be handled by workers exposed to radiation [18]. The second way is outside the system, in this case the limit is the one for public exposure equal to 1E-07 Sv/h. In figure 47 is possible to see that only FLiBe¹ and FliBe² can be recycling. Indeed, the pure FLiBe goes to zero after ten days, after that the salt could be reused both inside and outside the system. The FLiBe² reaches the dose rate level useful to be recycling inside the system after fifty years and never outside. The last FLiBe analyzed shoes the most interesting results, indeed the presence of uranium inside the system doesn't allow to recycle the salt. The minimum value of the dose rate of contact is 1.37E-05 Sv/h after a thousand of years.

In order to be reused, FLiBe must be purified from tritium, in doing so part of beryllium and lithium is lost. As it's possible to see in the previous chapters these two elements produce tritium through neutron capture. For this reason, the decrease involves a series of complications since they are necessary to maintain the value of TBR greater than 1. So, a quantity of fresh FLiBe must be added periodically to the system

5.1.1. Real case

Obtaining a totally pure FLiBe is complicated and very expensive, for this reason the most likely substance that it's more probable to find on the market is the one with impurities. Among the major problems related to the impurities certainly there is the corrosion of the surrounding materials, in fact HF, O and the metal impurities influence the corrosion rate of FLiBe. In addition, the beryllium and the fluorine with their impurities have threshold limit values (TLVs) for airborne exposure [17].

Subsequently there is an analysis of a plausible amount of FLiBe inside the ARC reactor blanket. The aim is to see the evolution of activity, dose rate and decay heat. An evaluation of the production of the plutonium and of the actinides within the system during the irradiation phase. Figure 48 shows the activity, the dose rate, the decay heat and the tritium activity during the irradiation phase.



Figure 45: Evaluation during the irradiation phase of the a) Total activity b) Dose Rate during c) Decay Heat d) Tritium activity

As can be seen from the graphs, a greater amount of substance leads to higher values for all aspects evaluated except for the dose rate. The growth of total activity is linked with that of tritium, this being one of the largest contributors. The total amount of tritium produced at the end of the irradiation phase is 705.4 kg. The total activity is 7.37E+20 Bq, the final contact dose rate is equal to 5.82E+05 Sv/h and the final heat output is 3.19E+05 kW.

Very interesting is the development of the plutonium and other actinides within the system following the presence of 21 kg of uranium in the system shown in figure 49.



Figure 46: Evaluation during the irradiation phase of the a) Actinides b) Isotopes of the plutonium

The presence of uranium inside the system allowed the development of actinides. The largest quantity element after uranium is plutonium. The isotopes that compose plutonium are shown in Figure 49b. The most present isotope is 239 Pu with 1.76E+03 grams. Also relevant are the 238 Pu and 240 Pu. The first has a half-life of 87.74 years and there are 3.03E+02 grams, the second has a half-life of 6.5E+03 years and there are 1.40E+02 grams. It is very important to view the development of plutonium as its compounds are highly toxic and radioactive.

6. CONCLUSION

The present work aimed to analyse the FLiBe molten salt used as a blanket in the ARC fusion reactor. To do that the program FISPACT-II was used. With this program it was possible to do an activation analysis of the salt and evaluate the activity, the decay heat and the contact dose rate after different period of irradiation. Several compositions of the FLiBe are analysed to verify how the impurities acted on the system. Focus has been made on uranium. Its presence within the system can bring several complications.

In the first part the pure substance is studied with the purpose to investigate the behaviour of the FLiBe and the tritium after different periods of irradiation. FLiBe, in fact, can produce tritium through neutron capture thanks the presence of lithium. The results shows that the irradiation period is linked to the production of tritium in the system. The level of dose rate and decay heat, in the first period after irradiation, have as major contributors some very unstable isotopes like ¹⁶N, ¹⁸F and ¹⁹O.

The analysis of the FLiBe containing impurities immediately show important differences. The first one is the greater number of nuclides at the end of the irradiation. The increased of the number also led to a variation in the impact that the isotopes have on the system regarding the activity, the dose rate and the decay heat. The value of the total activity compared to the one produced by tritium is much higher. Very interesting is the impact of the impurities on the contact dose rate, indeed they don't allow to this value to go to zero.

Subsequently an activation analysis on the beryllium is done. This material has been analyzed because during the extraction can have parasitic uranium. The value that has suffered most from the presence of uranium is certainly the contact dose rate, higher percentages of uranium correspond to higher values of the dose rate. Subsequently, an experimental beryllium composition called BP-1, designed for nuclear applications, is analyzed. At the level of activity there are no major differences with a pure beryllium. Instead for the dose rate the values are very different, the BP-1 following a ten-year irradiation, shows higher values. At the end an analysis of the FLiBe with a little percentage of uranium is done. Very interesting is the evolution of the actinides inside the system. It is clear from the analysis that the presence of the impurities has a big impact on the system. Especially the presence of the uranium led to some difficulties in fact it does not allow to recycle the FLiBe because the contact dose rate has value too much higher. In addition, there is the production of elements very harmful to the system like plutonium, americium and curium. Indeed, in the real case in which 7E+05 kg of FLiBe is considered, the presence of 21 kg of uranium produces more than 2 kg of plutonium.

Surely the FLiBe presents itself as an excellent molten salt as blanket. Future works will compare FLiBe with some other substance to evaluate an alternative with lower production costs and without possible uranium impurities. Eliminating the presence of uranium would in fact have great advantages, including the possibility of recycling the material and not having to dispose of some fission products such as plutonium.

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