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Theme

Study of Iodine-129 Transmutation in Nuclear Reactors

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Introduction

Nuclear power has become an important source of energy in the world. The increase in energy demand expected in the coming years and the depletion of fossil resources will be a real challenge. Nuclear energy has several advantages, given the considerable amount of energy it can produce. Moreover, it has made it possible to reduce dependence on hydrocarbons. Nevertheless, nuclear energy poses two major challenges to the scientist: nuclear waste and the use of its resources.

The separation of long-lived radionuclides is based on the industrial maturity of various process which are currently in use in reprocessing plants, we cite PUREX, DIAMEX, SANEX and SESAME. These processes are a satisfactory means can be developed for recovery of iodine-129 contained in spent fuel. Radionuclides can be trapped using : activated carbons and zeolites. Type of management can theoretically be envisaged : conditioning for deep geological disposal.

Currently, storage appears as a reference solution for the management of nuclear waste, but another way is being considered: separation followed by transmutation. The latter is based on the principle of transforming the long-lived radiotoxic element, by a nuclear reaction, into another element that is less radioactive or even stable. Among the most important long-live radio-elements, we distinguish iodine-129 with a half-life of $1,57\times10^7$ years. The other stable isotope available in spent fuel is iodine-127. Iodine is found in the form of metal iodides as irradiation targets. Transmutation of iodine-129 is applicable in nuclear reactors by neutron capture. Transmutation efficiency depends on neutron flux, neutron-cross section and irradiation time in reactors.

In order to show the potential interest of HFR (High Flux Reactor) reactors for the transmutation of iodine-129, we evaluate in the framework of this thesis, the transmutation performances in four research reactors namely: Petten (Netherlands), BR2 (Belgium), SM3 (Russia) and JOYO (Japan). This evaluation is based on the use of the transmutation calculation code ChainSolver 2.34. Other parameters calculated from this code are: transmutation rate R, transmutation amount C, transmutation support ratio TSR, average transmutation acceleration A to select the best reactor which provides a better rate.

The present manuscript is structured in four chapters. In the first chapter, some nuclear concepts are described and some basic notions of nuclear waste are presented as well as the three aspects of its long-term management.

In the second chapter, we describe the characteristics and methods of separation and transmutation of the long-lived fission product iodine-129.

The third chapter starts with the Bateman equation; the basis of the code, then the ORIP_XXI collection is explained in details. Its programs are: NKE, ChainFinder and ChainSolver. This collection is used for the creation and evaluation of the transmutation efficiency of iodine-129.

The fourth chapter is devoted to the numerical evaluation of the transmutation efficiency such as R, C, TSR and A.

Chapter I

From nuclear fission to nuclear waste

1. Atom, radioactivity and nuclear energy

From the 5th century BC, Leucippus, then Democritus, called "atom" (from the Greek "atomos" which cannot be cut, this small unit of matter, eternal, unlimited and constantly in rapid motion. In 1879, Joseph John Thomson discovered the electron. In 1911, an English scientist, Ernest Rutherford, saw that particles projected on matter pass through it as if it were a vacuum, except for a few, very rare ones which are reflected or even rebound. He thus discovered the atomic nucleus which concentrates almost the entire mass of the atom in a volume one hundred thousand times smaller than the latter. We then find that the positive electric charge of the nucleus is carried by the protons. In 1932, James Chadwick discovered the neutron, a neutral constituent of the nucleus. When alone, it transforms into proton and electron; when inside the atomic nucleus, it is stable. In 1969, physicists bombard nuclei with a beam of particles (electrons) accelerated enough to penetrate the core of nucleons, some electrons bouncing in seemingly bizarre ways. It can only be on even smaller corpuscles. This is how quarks are discovered. Each nucleon contains three [1]. Figure 1.1 illustrates some elementary particles [2].

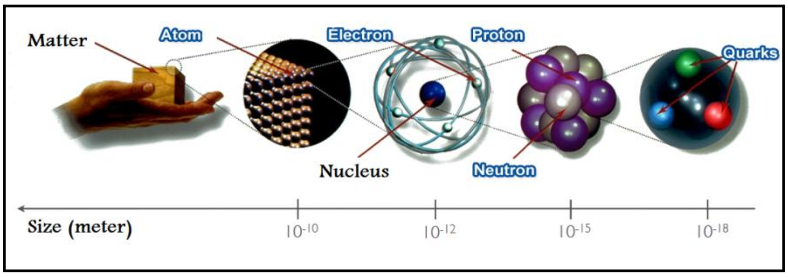


Fig. 1.1. Descent to the heart of matter : elementary particles.

In 1896, Henri Becquerel discovered the phenomenon of the radioactivity, also known as natural "radioactivity". Following the work of Henri Becquerel, Pierre and Marie Curie isolated polonium and radium in 1898. In nature, most atomic nuclei are stable, but not all. Some heavy nuclei formed outside the valley of stability. They have too many protons or too many neutrons, which makes them unstable. To return to the valley and therefore respect the laws of physics by finding the right balance between protons and neutrons, they are transformed into another lighter and more stable nucleus by ejecting very energetic particles. This transformation is called disintegration [3]. Radioactivity only concerns the nucleus, and to measure it, different units are used: the Becquerel (Bq), the Gray (Gy), the Sievert (Sv) and the Curie (Ci) [4].

Radioactivity is detected by various types of extremely sensitive detectors, including gas counter tubes (proportional counter, Geiger-Muller, ionization chamber), scintillators coupled to photomultipliers, semiconductors (silicon, germanium, etc.). Radioactivity decreases exponentially according to the law known "radioactive decay". The different types of decay are : alpha (α), beta (β) and gamma (γ). Figure 1.2 illustrates the penetrating power of different radiations [5].

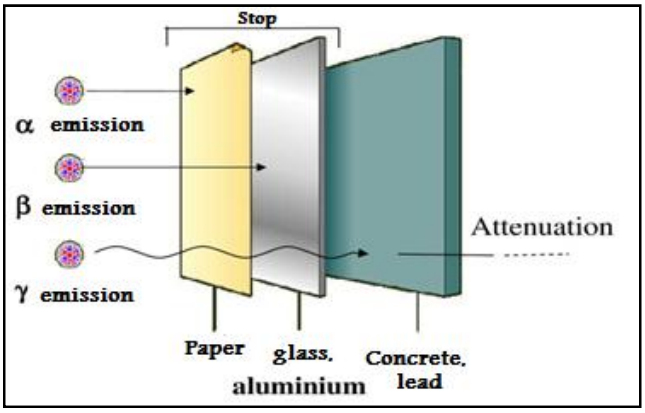


Fig. 1.2. Penetrative properties of radiation.

Energy is difficult to define simply other than through its effects and its variations. Its study is the basis of physics. The mastery of energy is therefore the engine of human activity. Man must store it, transport it, which some forms of energy allow better than others. For these reasons, it can be driven to transform energy. The discovery of electricity constituted a revolution: all known forms of energy can be transformed into electrical energy, the latter can then itself be easily transported and then transformed into movement or heat for the end user [6].

A nuclear system is formed by a nuclear reactor and the associated fuel cycle [7]. The use of nuclear energy consists in taking advantage of the high binding energy of the nuclei through exothermic reactions. The two main types of reactions are fission and fusion. Radioactivity itself releases great energy. For fission, we say that a nucleus if it fragments, spontaneously or induced, into two or more products plus the fission energy. Symmetrically, two nuclei fuse together if through nuclear reaction they produce a heavier nucleus plus a light fragment and fission energy. Heavy nuclei undergo fission, so light nuclei can undergo fusion [8]. Nuclear fission is the most concentrated source of energy that can be used today [6].

2. Nuclear history

A nucleus of certain large atoms has the property of breaking in two under the effect of a collision with the neutron, the latter without electrical charge has the ability to approach the nucleus sufficiently close without being repelled by electrical forces, the neutron can then penetrate inside this nucleus and break it into two pieces. The fragmentation of the nucleus is called fission reaction. An atom having the ability to break in two during a collision is said to be fissile, and the two pieces obtained after the fission are the fission products, and are most of the time radioactive. The best known of the fissile atoms are uranium-235 and plutonium-239 [9]. The sum of the masses of the fission products is lower than that of the initial atom of U-235. This decrease is due to the fact that this difference in mass has been transformed into energy. This energy is defined according to Einstein's mathematical formula $E = m.c^2$ where m, c are the mass and the speed of light respectively [10]. At the same time, two or three neutrons are also produced. These neutrons will also cause other fission reactions [6]. The energy produced is carried away by the fission products in kinetic form which will eventually be transformed into heat [9]. This heat can be used to generate electricity [6].

Following the Joliot-Curies in early 1934, other research teams attempted to create new radioactive nuclei by varying materials, targets and incident particles. protons suffered from a handicap for this: their positive electric charge made it necessary to provide them with kinetic energy beyond the reach of the accelerators of the time to make them cross the Coulomb barrier [11]. In 1934, the Italian physicist Enrico Fermi conducted experiments in Rome which showed that neutrons could split many types of atoms. The results surprised Fermi himself. When he bombards uranium with neutrons, he doesn't get the expected elements. The elements were much lighter than uranium [10]. The experiment, conducted in 1934, proved disappointing, visibly very many radioactive decays appeared in the reaction products without it being possible to identify the elements 93 or 94 that we were looking for [12].

It was not until the very end of 1938 that the Germans Otto Hahn and Fritz Strassmann, from the Kaiser Wilhelm Institute in Berlin, used a neutron source which contained radium and beryllium to bombard a uranium target, they were surprised to find lighter elements such as barium among the residues [10]. Mendeleev's table and not nearly anywhere near uranium are expected, this was evidence that the uranium nucleus had broken into two pieces, each approximately half the mass of the uranium. Fission was discovered. Lise Meitner and her nephew Otto Frisch announce it. A fission releases about two hundred million electron volts compared to the combustion of a carbon atom which releases four electron volts. This is an example of the order of magnitude [12].

In 1939, Bohr came to America. He shared with Einstein the findings of Hahn-Strassmann-Meitner. Bohr also met Fermi at a theoretical physics conference in Washington. They discussed the exciting possibility of self-perpetuating chain reaction. In such a process, atoms can be split to release large amounts of energy. Scientists around the world began to believe that a self-sustaining chain reaction might be possible. It would occur if enough uranium could be processed under specific conditions. The amount of uranium required to make a self-sustaining chain reaction is called a critical mass. Fermi and his associate, Leo Szilard, suggested a possible design for a uranium chain reactor in 1941. Their model consisted of uranium placed in a stack of graphite to make a cube-shaped framework of fissile material [10].

In 1942, in the United States, the first reactor was built by Fermi. It consists of a stack of 6 t of metallic uranium, 34 t of uranium oxide and 400 t of graphite [13]. This first reactor is also known as the Chicago pile [10]. In France, the first Zoé test reactor was built by the CEA, which operated for the first time in 1948 [13].

The first nuclear reactor was just the beginning. Most of the early atomic research focused on developing an effective weapon for use in World War II. This project was carried out under the code called the Manhattan Project. After the war, the United States government encouraged the development of nuclear energy for civilian purposes. Congress created the CEA Atomic Energy Commission in 1946. The commission authorized the construction of the first generation experimental reactor on the Idaho site. This reactor generated the first electricity of nuclear origin on December 20, 1951 [10]. In 1954, the Soviet Obninsk reactor paved the way for higher powers with 5 MW. The first French reactor producing electricity entered service at Marcoule in 1956. Its power was 40 MW [14].

3. Nuclear system

A nuclear system is formed by a nuclear reactor and the associated fuel cycle. It is globally optimized in its industrial implementation, from raw material to waste [7]. Figure 1.3 gives a general presentation of a pressurized water reactor and its main circuits [15].

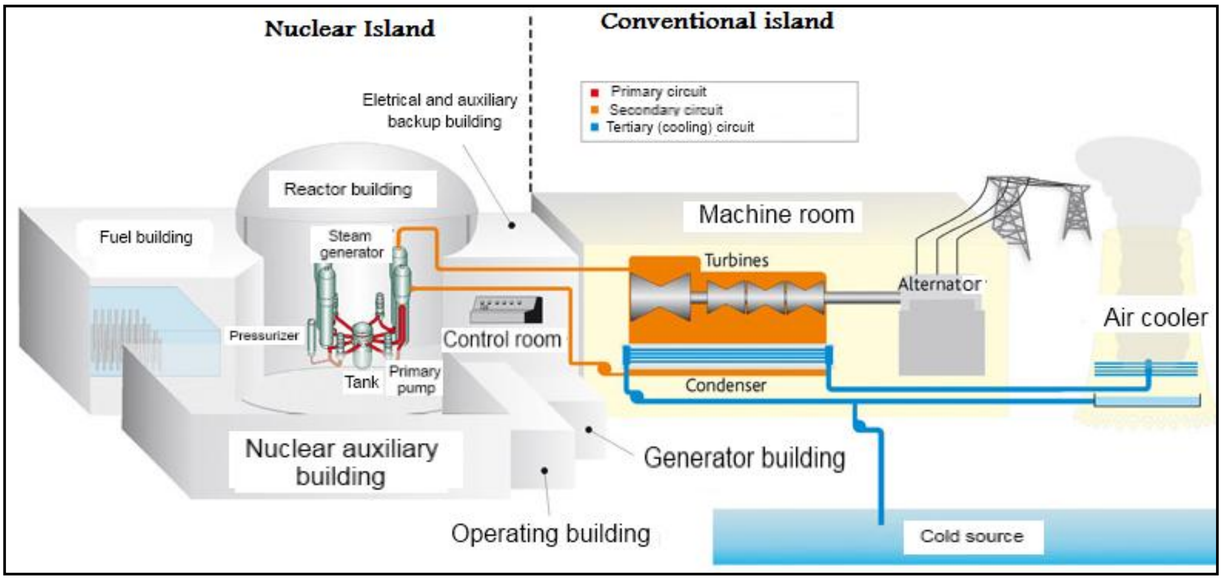


Fig. 1.3. General presentation of a pressurized water reactor and its main circuits.

3.1. Nuclear reactor

The purpose of a nuclear power plant is to generate electricity. Its structure is in principle very similar to that of a conventional thermal power plant. Fission produces heat which is used to heat water and make steam. The latter is used in a turbine to produce mechanical energy which is transformed into electricity with an alternator. The fission of a heavy nucleus gives two nuclei of average mass and a few neutrons: two or three neutrons (2.4 on average) depending on the case [14]. These neutrons released during fission will in turn be able to cause new fissions and the release of new neutrons and so on, this is the chain reaction. In a nuclear reactor, the chain reaction is controlled, that is to say a balance must be reached: a fission gives a fission, etc. and not one fission gives two fissions which gives four fissions, etc. [9]. This process is exothermic and, in the case of the fission of a uranium nucleus, releases about 200 MeV. Fission can be spontaneous or induced. The nuclear reactors used in the world to produce electricity are slow or the thermal neutron reactors [14].

Some nuclei, when they capture a slow neutron, fission. They are said to be fissile nuclei. This is the case of uranium-235, uranium-233, thorium-229, neptunium-238 or polonium-241. Other nuclei, called fertile, are those which, after capture of a neutron and radioactive decomposition, can lead to fissile nuclei. This is the case of uranium-238, uranium-234, polonium-238, polonium-240, americium-241 or thorium-232 [14].

Five main constituents are needed to design a reactor core [13]:

- A fuel in which fission occurs, only uranium-235 occurs naturally. It is therefore most often used in natural form or enriched uranium oxide, or even plutonium [11]. The nuclear fuel is placed in the reactor core [13].
- The coolant, which evacuates the thermal energy produced: water, liquid, or not, sodium [11]. It is a heat carrier outside the reactor [13].
- The moderator (except for fast neutron reactors) [13]. Which slows down neutrons by elastic scattering to promote their capture by uranium-235 nuclei: heavy water, graphite. For proper operation of the reactor, fuel and moderator must be alternated [11].

- Control rods, also called rods or control and stop rods, made of neutron - absorbing materials (boron, cadmium), ensure the chain reaction [7]. These bars are mobile, trapping neutrons [13].
- The steam generator, a heat exchanger [9].

The different types of nuclear reactors are distinguished by their physical or technological characteristics, as well as by their field of application. Electronuclear is currently the most industrially developed field. From a physical point of view two main families of reactors can be distinguished according to the neutron spectrum chosen: thermal or fast [16]. Among all the possible combinations between the different fuels, coolants or moderators, only some have been retained and have given rise to industrial achievements [13]. After the very first power reactors, such as UNGG (Uranium Natural Graphite-Gas), which constituted the first generation developed in the 50s/70s and which are now almost all shut down, large-scale reactors unit power, such as PWRs (Pressurized Water Reactor) constituted a second generation that entered into operation between the 70s and 90s, which constitutes the bulk of the current world fleet [17]. The Soviet sector RBMK (Reactor Bolchoe Molchnastie Kipiachie) mean high power boiling reactor is classified in the second generation, they were disqualified by the Chernobyl accident in 1986. There are also BWRs (Boiling Water Reactors). Finally, the CANDU-type PHWR (Pressurized Heavy Water Reactor) sector, of Canadian design. The third-generation sector corresponds to facilities that are beginning to be built with a view to commissioning from around 2010. It includes in particular the EPR (Evolutionary Pressurized Reactor) which was developed by the NPI (Nuclear Power International) subsidiary of Framatome and Siemens. The fourth generation under study, expected around 2040 on an industrial level. These are fast neutron reactors (FNR) [7]. The characteristics of these reactors is that they do not include a moderator, so the neutrons remain fast [9].

3.2. Nuclear fuel cycle

A fuel is a material that provides heat when it burns. The best known are wood, coal, natural gas or oil. By analogy, uranium, used in nuclear power plants, is called "nuclear fuel" because it also releases heat by fission and not by combustion [18]. The element uranium has three natural isotopes; U-238 (99.28 %), U-235 (0.718 %) and U-234 (0.0056 %). Uranium is a metal, with a density of 19, whose melting point is 1133°C. Uranium is relatively widespread in the earth's crust (2 to 3 g per ton of rock), more abundant in granites (10 to 30 g per ton) and very low content in water [19], the Figure 1.4 presents the nuclear fuel cycle [15].

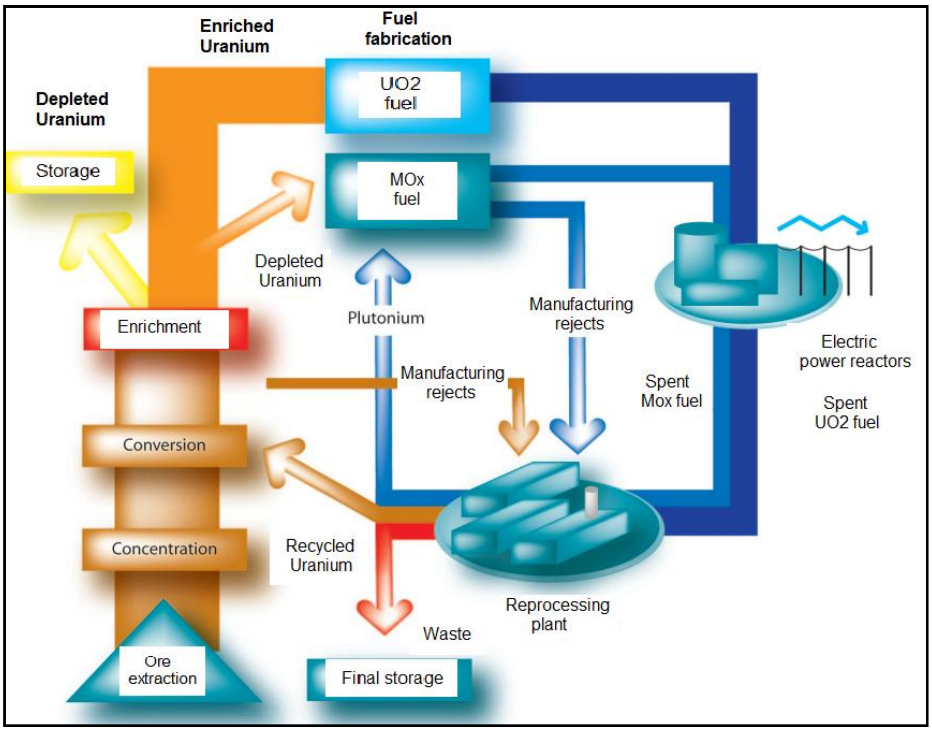


Fig. 1.4. The nuclear fuel cycle.

The nuclear fuel cycle (Fig. 1.4) is the set of operations necessary to supply nuclear reactors with fuel, then to store, reprocess and recycle this fuel after use: prospecting and extraction of ore, concentration, conversion and enrichment of uranium, manufacture of fuel elements, reprocessing and recycling of nuclear materials, waste storage [8]. A distinction is made between the front end of the nuclear fuel cycle and the back end [17].

The nuclear fuel cycle begins with the extraction of uranium ore in open-pit mines or in underground galleries [18]. The uranium content of the ore is generally quite low: 0.1 to 0.3 %. That is, only 2 to 3 kg of uranium per ton of ore extracted. The uranium is concentrated; the ore is first crushed and ground, then dissolved in sulfuric acid. The resulting solution is then chemically treated to obtain a concentrate called "yellow-cake" because of its color, and which contains about 70 % uranium. The treatment and concentration of 1000 tons of ore yields 1,5 tons of yellow cake. The yellow cake is not sufficiently pure, so it undergoes a purification step known as fluorination: various chemical treatments make it possible to obtain a very pure uranium oxide, which is then reacted with hydrofluoric acid to obtain uranium tetrafluoride (UF_4). From this UF_4, it is possible to obtain uranium hexafluoride (UF_6) intended for enrichment, which is in the gaseous state at a temperature of 56°C. Several processes for enriching uranium have been developed: gaseous diffusion and ultracentrifugation. All of them rely on the difference in mass that exists between the nuclei of the two isotopes of uranium in order to obtain a desired enrichment rate 3 %. After enrichment, the enriched UF_6 is transported in solid form to the fuel element manufacturing plant. It is first transformed into uranium oxide UO_2 in the form of a black powder. This powder is then sintered, that is, compacted with automatic presses that give it the shape of small cylindrical pellets 13 mm high and 8 mm in diameter, then brought to high temperature (1800 °C) in a furnace. After checking their characteristics, the UO_2 pellets are stacked in cylindrical zircolay tubes, called "rods"; 264 rods 4m long, arranged in a square section of 17×17 constitute an "assembly". The core of a 900 MW PWR reactor contains 157 of these assemblies, i.e. a total of 41.448 rods and more than 11 million pellets, and contains 72.5 tons of fuel [8]. An assembly remains in the reactor for three to four years. Out of 100 kg of fuel originally presents, for example, 3 kg of U-235 and 97 kg of U-238, there remains after three years of use.

- 1 kg of U-235 (2 kg were fissioned) ;
- 95 kg of U-238 (2 kg were transformed into Pu-239) ;
- 1 kg of Pu-239 (of the 2 kg obtained by the transformation of U-238, 1 kg was fissioned) ;
- 3 kg of fission products.

The spent fuel is stored in an on-site cooling pool. Then the assemblies are transferred to a deactivation pool and remain there for about a year [8].

Reprocessing consists, on the one hand, in recovering the material that is still usable, the plutonium and uranium, that is to say, the recycling of the energetic materials contained in the spent fuel, and on the other hand, in sorting out the radioactive waste that cannot be recovered by storage after its removal from the reactor. New fuels composed of a mixture of uranium oxide and plutonium oxide (called MOx; Mixed Oxides) are already used in certain PWR reactors. The recovered uranium can still be enriched to more than 3 % and follow a path similar to that of ordinary fuel [18].

The major drawback of nuclear energy is the generation of radioactive waste. Nuclear power will have to address the problems of waste management, plant safety, proliferation of radioactive materials and optimization of uranium resources [6].

4. Nuclear waste

4.1. Definition of nuclear waste

After leaving the reactor, nuclear fuel undergoes a series of operations before seeing the material is labeled "recoverable" or "waste" [19]. The production of nuclear waste is the price to pay for the production of energy from the phenomenon of nuclear fission [6]. According to the definition of the International Atomic Energy Agency (IAEA), radioactive waste is any material for which no use is foreseen and which contains radio-nuclides in a concentration higher than the values that the competent authorities consider acceptable in materials suitable for uncontrolled use [21]. The definition of radioactive refers to two problems that are not semantic problems [22]:

- That of ultimate radioactive waste. Ultimate waste is waste that cannot be treated under the economic and technical conditions of the moment, in order to extract the recoverable part or to reduce its polluting or dangerous nature.
- That of the exemption and release thresholds below which radioactive waste can or could be treated in a conventional disposal system.

Figure 1.5 shows the production of waste from the nuclear power industry; high-level and long-lived waste [23].

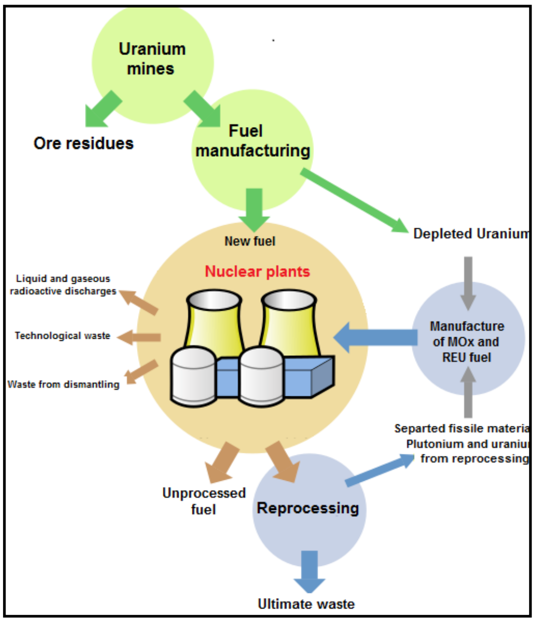


Fig. 1.5. Production of waste from nuclear power industry: HL and LL waste.

4.2. Nature and origin of nuclear waste

Radioactive waste is essentially produced in the context of nuclear activities [24]. Most nuclear waste comes from the back end of the nuclear fuel cycle and has its physical origin in fission reactions and neutron capture, sometimes associated with radioactive decay. In the first case, they are called fission products, and in the second case activation products, or "actinides" depending on the capturing body [25]. The main source of nuclear waste is spent nuclear fuel [26]. Radioactive waste is produced at each stage of the nuclear fuel cycle, which includes a series of activities: mining and milling of uranium ore, conversion and enrichment of uranium, fuel fabrication, operation of nuclear reactors, reprocessing of spent fuel (if any), dismantling of nuclear power plants and associated cycle facilities, and site rehabilitation [27].

The main wastes from nuclear energy have three origins [17]:

- Fission products, ashes of the chain reaction: these nuclei are almost all characterized by an excess of neutrons compared to stability and are therefore mostly β and γ emitters.

- Actinides (series of elements with atomic number between 89 and 104 in the periodic table) [6]. They are classified in two categories: the major actinides (uranium and plutonium) which have heavy nuclei and are fissile materials likely to be recycled in reactors and the minor actinides (neptunium, americium, curium) which are all α emitters and some also β emitters [17].
- Activation products, resulting from neutron capture by the structures of a reactor, for example cobalt 60. They are β and γ emitters [17].

Table 1.1 gives more details on the amount of energy associated with the fission of a U-235 nucleus, and Table 1.2 illustrates the different components of the inventory at the discharge of a pressurized water reactor in France per year [28].

Kinetic energy of F.P	165 MeV
Kinetic energy of the emitted neutrons	5 MeV
γ radiation	12 MeV
β radiation	8 MeV
Energy of antineutrinos	12 MeV
Total energy released	202 MeV

Tab. 1.1. Detail of the energy associated with the fission of a ^{235}U nucleus.

	Radionuclides	Period (y)	Discharge (kg/year)	Total (kg/year)
URANIUM	^{235}U	7.1×10^{8}	220	20500
	^{236}U	2.3×10^{7}	90	
	^{238}U	4.5×10^{9}	20200	
PLUTONIUM	^{238}Pu	88	3.3	210
	^{239}Pu	24120	123.1	
	^{240}Pu	6570	47.5	
	^{241}Pu	14.4	25.4	
	^{242}Pu	3.7×10^{5}	10.5	

MINOR ACTINIDES	^{237}Np	2.1×10^{6}	8.8	16
	^{241}Am	432.2	4.4	
	^{243}Am	7380	2.2	
	^{244}Cm	18.1	0.5	
MEDIUM-LIVED F.P	^{90}Sr	28	10.5	35
	^{137}Cs	30	24.3	
LONG-LIVED F.P	^{93}Zr	1.5×10^{6}	15.5	50
	^{99}Tc	2.1×10^{5}	17.7	
	^{107}Pd	6.5×10^{5}	4.4	
	^{129}I	1.6×10^{7}	3.9	
	^{151}Sm	93	7.7	

Tab. 1.2. Components of inventory at discharge of PWR per year.

It is important to understand the origin of these elements. Neutrons in the reactor do not only induce fissions, but a whole series of nuclear reactions, more or less probable [28]. It can be noted that the proportions in the spent fuel are (% by mass) [22]:

- U (Uranium) : 95.5 %, Pu (Plutonium) : 1.0 % ;
- F.P (fission products) : 3.5 % ;
- M.A (minor actinides) : 0.1 %.

After leaving the reactor, the spent fuel rests in a pool for four years before any intervention, to allow the fuel to cool. Uranium and plutonium must then be separated from the actinides and fission products [20]. Spent nuclear fuel is reprocessed in such a way as to [6]:

- Recovering volatile energy materials;
- Separate these materials from the other radioactive elements that constitute the waste;
- Conditioning waste in an inert and safe form by embedding it in glass, it is the vitrification process.

4.3. Characteristics of nuclear waste

Nuclear waste contains radioactive elements called radionuclides that emit radiation that can harm the body's cells, so it cannot be recycled or left in the environment. The physical and chemical nature, the level and type of radioactivity, are all characteristics that differ from one waste to another. Classification is therefore necessary. It defines the categories of classes of waste. Each type of waste requires the implementation or development of specific treatment and management procedures, from the producer to the outlet, appropriate to the risk it presents [29]. The radionuclides contained in radioactive waste may be of artificial origin. The radioactive characteristics of waste are [30]:

- The type of radionuclides contained and the radiation emitted (alpha, beta, gamma), the activity (number of atomic nuclei that spontaneously disintegrate per unit of time - expressed in Becquerel);
- The Half-life (time required for the activity of a radionuclide in a sample to decrease by half).

4.4. Classification of nuclear waste

4.4.1. According to the period

According to the radioactive half-life of radionuclides, we distinguish [31]:

- Waste whose radionuclides have a very short half-life (less than 100 d): VSHL waste.
- Waste whose main radionuclides have a short half-life (less than 31 y): SHL waste.
- Waste whose main radionuclides have a long half-life (over 31 y): LHL waste.

4.4.2. According to the mass activity

Radioactive waste is most often classified according to its mass activity, i.e., the quantity of radioactive elements contained per unit of mass [32] :

- Very Low Level Active (VLLA) elements with an activity between 1 Becquerel/gram (Bq/g) (the average level of natural radioactivity) and 100 Bq/g;
- Short-lived Low-Level waste (SLLL) and Long-Lived waste (LLLL), whose radioactivity varies from 0.1 to 10 million Bq/g;
- Short-Lived Medium-Level waste (SLML) and Long-Lived Medium-Level waste (LLML): they have a radioactivity of around 100 million Bq/g;
- High-Level and Long-Lived waste (HLLL) concentrates 92 % of the radioactivity and has activity levels of several billion Bq/g. Most of the high-level waste comes from the irradiation in nuclear reactors of fuel consisting of uranium pellets (UOX) and, in part, of mixed uranium-plutonium oxide (MOx).

4.4.3. According to waste manager

From the point of view of the "waste manager", three categories are, in fact, distinguished [17]:

- Category A: low or medium activity short-lived (LMA-SL) (by definition, maximum 30 y). In practice, these are β and γ emitters; the content of α emitters must not exceed a very low limit of 1.517 GBq per ton. They consist essentially of technological waste.
- Category B: low or intermediate level long-lived (LIL-LL), which may contain α, β and γ emitters. They are made of highly contaminated materials such as fuel cladding.
- Category C: high activity and long-lived HALL (including products with periods that are not very long but that must be stored for a very long time given the initial activity). This is essentially irradiated fuel, if not reprocessed, or fission products and minor actinides separated during reprocessing.

To these categories, must be added very low level waste (VLL).

Type	Activity	Period
A	Low Medium	Short
B	Low Medium	Long
C	Strong	Long

Tab. 1.3. Classification of nuclear waste.

4.5. Origin of different classes

For the origin of the different classes of waste [30]:

- Waste (VSHL) : a significant part of which results from medical applications of radioactivity (diagnostic or therapy) and whose radioactive elements have a half-life of less than 100 d;
- Waste (VLL) :14 % of the volume and 0.031 % of the radioactivity come from the nuclear industry, in particular from the dismantling operations of the facilities. Uranium ore processing residues can be attached to this category; they benefit from specific management;
- Waste (LMA-SL) :76.8 % of the volume and 0.055 % of the radioactivity come mainly from the nuclear industry, but also from a few research laboratories;
- Waste (LLLL) : 4.6 % of the volume and 0.013 % of the radioactivity are either radium-contaminated wastes (called radiferous) and mainly originating from the use of naturally radioactive raw materials in industrial processes;
- Waste (LLML) : 4.4 % of the volume and 8.2 % of the radioactivity, result mainly from the processing of spent fuel (spent fuel cladding, processing sludge) and from the maintenance of nuclear facilities;
- Waste (HALL) : 0.2 % of the volume and 91.68 % of the radioactivity are non-recyclable materials from the processing of spent fuel from nuclear power plants.

4.6. Nuclear waste management

The management of radioactive waste must be carried out with respect for the protection of nature, the environment and health, taking into consideration the rights of future generations [25]. The main source of waste is spent fuel. The vast majority of this waste (90 %) is low-level and short-lived and is stored in surface sites. In less than 300 y, its radioactivity will have returned to the level of natural radioactivity. The remaining 10 % have a long life, up to several tens of thousands of years. This waste can be safely stored temporarily [6].

There are two approaches to spent fuel [14]:

- The first is to leave the spent fuel as it is and to consider it as waste. This is called open cycle management.
- The second is to try to recover now or in the future what can be recovered from the spent fuel. The different constitutes are chemically separated by reprocessing the fuel. We say that we have a closed cycle management.

After three years of storage of a 1300 MW PWR, the uranium contains mostly ^{238}U; the plutonium produced represents only 1 %. Uranium and plutonium can be recovered or used in fast neutron reactors and cannot be considered as final waste. The vast majority of fission products and minor actinides are, on the other hand, ultimate waste that must be stored at great depths in a conditioning system appropriate for long-lived waste, or incinerated in fast neutron reactors. Plutonium can be used in the short term by making MOx fuel. MOx can be used, together with normal fuel, in dedicated nuclear reactors [14].

As with other wastes, the first aim for radioactive waste is to reduce its volume. Thus, it is compacted, incinerated or cut up and then packaged to form transportable package ready for storage or disposal. VLL waste is compacted and packaged in bags (bigbags) or in metal caissons. LMA-SL waste is conditioned and sometimes compacted, in solid form in several types of packages: metal drums, concrete shells, concrete caissons of filters used in nuclear power plants, or metal caissons. LLML waste is encased in bitumen or concrete and placed inside stainless steel or concrete container. HALL waste vitrified, i.e., packaged in special glass that is resisting to heat and radiation. These glasses are then poured into stainless steel tight containers. The containers are stored in ventilated shafts for cooling. Studies are underway on the conditioning and storage of HALL waste. It is currently stored on the surface, in complete safety, for several decades. Storage consists of placing the radioactive waste package in appropriate facilities without the intention of removing them [24].

The situation in France is governed by the 1991 Bataille law, which gives fifteen years to research to develop three solutions for managing nuclear waste [25]. This law concerns only the HALL type of waste, which is currently stored, since LLA and VLLA waste is considered to be permanently stored [17]. Work on the management of this HALL waste will be carried out in three research areas [33].

- Axis 1 : Of solution concerning the separation and transmutation of long-lived radioactive elements;
- Axis 2 : Possibilities of reversible or irreversible storage in deep geological formations, particularly through the construction of underground laboratories;
- Axis 3 : Conditioning processes and long-term storage.

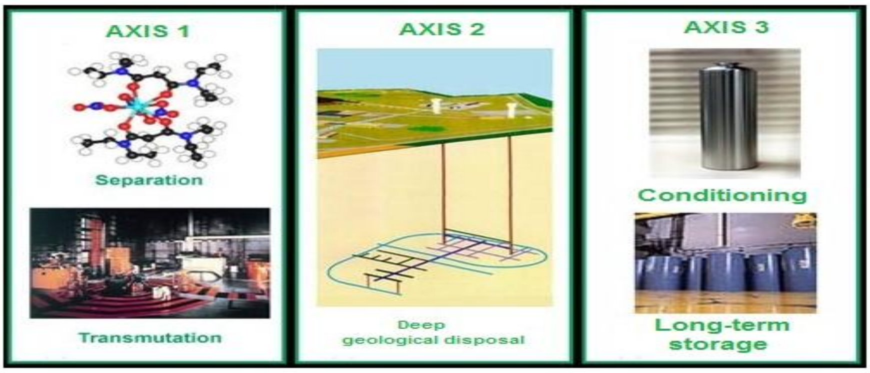


Fig. 1.6. Overview of the three research axes.

4.6.1. Separation and transmutation

Table 1.4 below gives the orders of magnitude of the amount of HALL in the fuel irradiated in PWR reactors, respectively minor actinides (mainly α emitters) and long-lived fission products (β and γ emitters). The activation products are, like the fission products, β and γ emitters [17].

	Nuclide	Period (y)	Abundance (g/t)
Minor Actinides MA	Neptunium-237	2 100 000	430
	Americium-241	430	220
	Americium-243	7 400	100
	Curium-243	28	0.3
	Curium-244	18	21,4
	Curium-245	8 500	1.2
Fission Products FP	Selenium-79	65 000	4.7
	Zirconium-93	1 500 000	713
	Technetium-99	210 000	810
	Palladium-107	6 500 000	200
	Tin-126	100 000	20
	Iodine-129	16 000 000	170
	Cesium-135	2 300 000	360

Tab.1.4. Main minor actinides and fission products present in PWR reactors (UOX fuel irradiated at 33 000 MWd/t; 3 years after unloading.

The objective of advanced separation and transmutation is to sort and transform certain long-lived wastes into other, less radiotoxic, short-lived wastes. This approach would significantly reduce the quantity and long-term harmfulness of the waste. Concerning more precisely advanced separation, the objective of this process is to extract from spent fuel, in addition to the uranium and plutonium already extracted, other radioactive and long-lived elements contained in the waste. In concrete terms, within the framework of research in this area, researchers are working on the development of advanced separation techniques, as well as on the definition of concepts for nuclear reactors and systems capable of transmuting long-lived radioactive elements. This research is led by the CEA, which has several research laboratories in this field (Phénix reactor for studies on transmutation) [24]. Transmutation will certainly be useful in reducing the quantities of HALL waste, but it will not eliminate all of it [17]. Figure 1.7 defines the transmutation process [34].

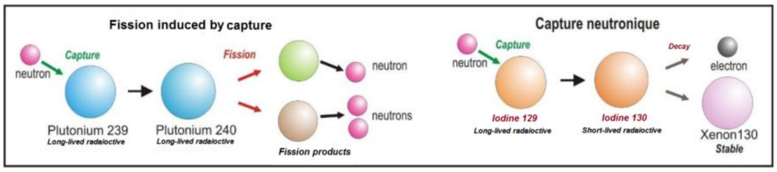


Fig. 1.7. Transmutation by causing fission (left) / simple capture of a neutron (right).

4.6.2. Deep storage

Research in this area is aimed at developing underground storage arrangements and technologies, several hundred meters below the surface. Unlike storage, which is temporary in nature, deep geological disposal makes it possible to place long live radioactive waste permanently in a site that guarantees its containment, while reserving the possibility of recovering it if necessary or appropriate. Three types of waste are being studied for deep geological disposal: SLML, HALL, and spent fuel that would be conditioned without treatment [24]. For a man-made repository, not only is the geological terrain optimally chosen - the main criteria being the least possible circulation of water (clay, granite, salt mine), geological stability, and the absence of exploitable mining resources - but at least three artificial barriers would be added: matrix (e.g., a glass), containers, and structures. Under these conditions, models prove the safety of storage over periods longer than the periods of the products to be stored [17]. Deep storage is illustrated in Figure 1.8 representing the CLAB installation in Sweden [34].

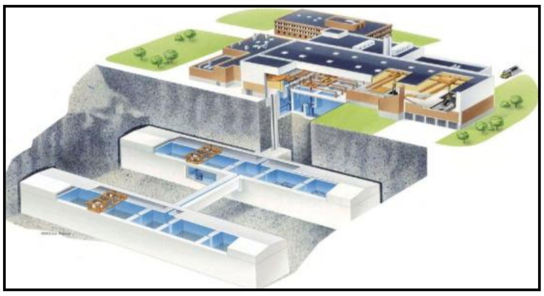


Fig. 1.8. The CLAB facility in Sweden for underground storage (about 30m deep of spent fuel under water).

4.6.3. Conditioning and long-term storage

The objective of the researchers on conditioning and long-term storage is to develop conditioning for all types of waste and to study the feasibility of storage facilities designed from the outset to operate for a period of 100 to 300 y while guaranteeing the protection of man and the environment. The facility concepts were examined for two different locations, one above ground and one below ground. Storage is an interim solution, designed to retrieve waste packages for final disposal or recycling if new techniques are found. For specific, researchers are studying materials known to be highly resistant to alteration. The research focuses in particular on the study of materials belonging to the family of ceramics [24]. Vitrification remains the best qualified process for all high-level waste extracted by the separation process [21]. The concept of surface and subsurface storage is illustrated in Figure 1.9 [34].

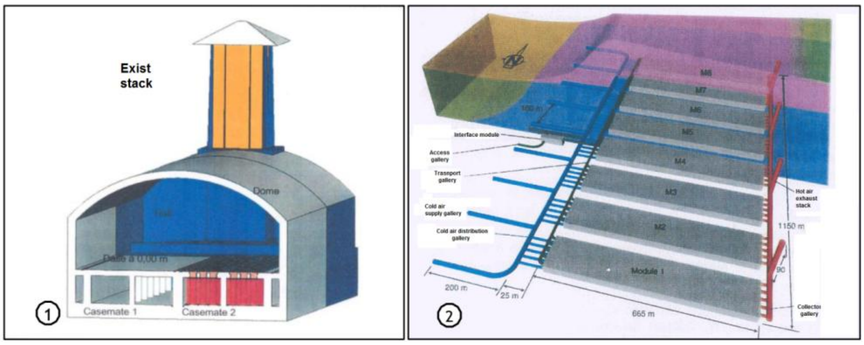


Fig. 1.9. CEA's concept of storage for HA and CU (1) above ground, (2) below ground.

The general diagram that summarizes the waste management cycle is given in Figure 1.10 [29].

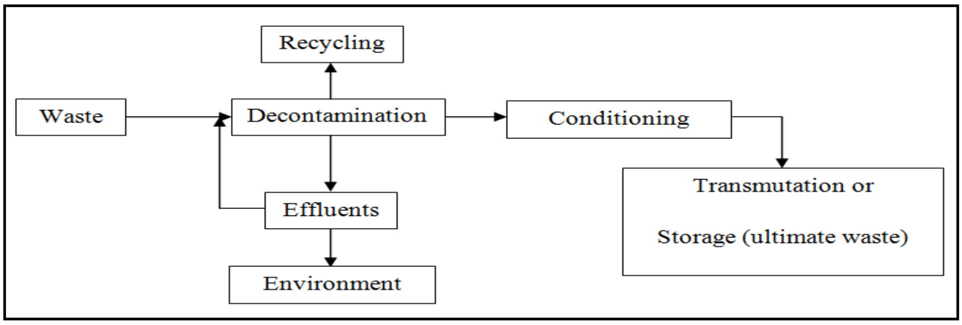


Fig. 1.10. General waste management cycle.

Chapter II

Separation and Transmutation of Iodine-129

I. Separation of Iodine-129

1. Spent fuel

1.1. Medium - and long-lived fission products

The production of waste is the price to pay for the production of energy from the phenomenon of nuclear fission. There are two categories of nuclear fission products; medium-lived and long-lived. The medium-lived fission products are Strontium-90 (28.8 y) and Cesium-137 (30.07 y). The long-lived fission products are: Selenium-79 (327×10^{3} y), Zirconium-93 (1.53×10^{6} y), Technetium-99 (710×10^{6} y), Palladium-107 (6.5×10^{6} y), Tin-126 (0.23×10^{6} y), Iodine-129 (15.7×10^{6} y), Cesium-135 (2.3×10^{6} y) and Samarium-151 (93 y), in total eight fission products [26].

1.2. Inventory of long-lived fission products from a PWR reactor

The basic fuel for light water nuclear reactor is uranium in oxide form, slightly enriched in U-235 (< 5 %), which is its only fissile isotope. As it produces energy, the nuclear fuel is depleted of fissile material, until it becomes necessary to unload the fuel assemblies to replace them with new ones [36]. Under the effect of the nuclear reactions that occur within the nuclear fuel, it undergoes a certain number of important transformations, leading to an evolution of its composition throughout its use in the reactor. At the end of its cycle in the reactor, the fuel is called spent fuel [26].

The average composition of long-lived fission products LLFPs at the end of the fuel cycle, initially enriched in ^{235}U to 3.5 %, of a PWR reactor with a burn-up of 33 GWd/t and after 3 years of operation, is given in the following Table 2.1:

Radionuclides	Period (y)	Discharge (kg/y)
Selenium-79	70000	0.11
Zirconium-93	1.5×10^6	15.5
Technetium-99	2.1×10^5	17.7
Palladium-107	6.5×10^6	4.4
Tin-126	10^5	0.44
Iodine-129	1.57×10^7	3.9
Cesium-135	2×10^6	7.7
Samarium-151	93	0.33
Total long lived fission products		50.1

Tab. 2.1. Inventory of LLFPs at PWR reactor discharge.

1.3. Problems posed by long-lived fission products

The destruction of selected long-lived fission products (Technetium-99, Iodine-129 and Cesium -135) in reactors poses many problems:

- ✓ They are pure consumers of neutrons;
- ✓ Their capture cross-sections (and thus transmutation rates) are low in the fast spectrum (< 0,5 b) and in the thermal spectrum (a few barns at most);
- ✓ They are in the minority (except for ^{99}Tc) compared to the stable or medium-lived isotopes of the element produced in the reactor, as shown in the previous table;
- ✓ The chemical form that can be introduced into the reactor is not necessarily the pure element (especially for easily fusible or volatile bodies, such as iodine and cesium).

The problems mentioned are susceptible to various solutions.

For the first point, hybrid reactors are more adapted to the transmutation of LLFPs, thanks to their external source of neutrons, but this gain is limited by the maximum acceptable intensity of the beam. In a critical reactor, the destruction of LLFPs can only use the surplus of neutrons released by the fission of actinides.

2. Iodine-129 in the fuel cycle

2.1. Characteristics of Iodine

Iodine is an element of the halogen family with atomic number 53. It crystallizes in an orthorhombic structure with a melting point at 113.7 °C. Its valence number can be -1, 0, 1, 5 or 7 and its atomic radius is 130 pm. Various iodines are described in the literature: IO, I_2O_4, I_2O_5 and I_4O_9. For example, IO is a purple gas that reacts with ozone. The other oxides are very soluble in water and react to form iodic acid (HIO_3) and di-iodine I_2. Their melting temperature is low, between 75 ºC (I_4O_9) to 300 ºC (I_2O_5) and they are rarely obtained pure. Iodine has five major isotopes: 123, 125, 127, 129 and 131. Iodine-123 and iodine-125 decay to tellurium-123 and tellurium-125 by electron capture and emission of gamma radiation of energy 127 keV and 35 keV, respectively. The isotope 127 is the stable isotope of iodine. The isotope 129 and 131 undergo β^- decays (energy 156 keV and 236 keV respectively) and emit γ-radiation with energy 40 keV and 364 keV respectively, to form stable xenon forms; xenon-129 and xenon-131 [37].

Thirty-seven isotopes of iodine are currently known, with atomic masses ranging from 108 to 144; twenty-eight of them have half-lives shorter than one day. In the case of irradiated fuel, the production of iodine consists mostly of iodine-129 (82 %) whose half-life is equal to $1.57.10^7$ y, but also of iodine-127 (stable isotope) and iodine-131 with a period of 8.05 d [38].

The radioactive decay of iodine-129 occurs according to the following scheme [38]:

$$ {}^{129}_{53}I \xrightarrow{\beta^-(156\,keV);\ 1.57.10^7\,years} {}^{129m}_{54}Xe \xrightarrow{39.6\ keV\ ;\ 1.0\ ns} {}^{129}_{54}Xe(stable) $$

The heat released is about 34 MW/t of fuel. Chemically very active, iodine is rarely found in elemental form. It combines easily and forms organic compounds (HOI, ICN, alkyl iodide) but also inorganic compounds (I_2, HI). The literature reports the possibility of very complex mixtures including high molecular weight compounds

[39]. Iodine is one of the first fission products to escape from the fuel in case of cladding rupture. It reacts with the water in the primary circuit of pressurized water reactors (PWR). In practice, iodine is only released from the fuel when it is dissolved during reprocessing. At this stage, three years after the reactor's exit, practically only the isotope 129 remains, the iodine-131 having disappeared by decay [38].

Because of its very long half-life, iodine-129 has a low specific activity that implies a relatively low radiotoxicity compared to the other nuclides in radioactive waste. However, it is active over a very long period compared to the human scale, migrates easily in the environment, and readily concentrates in a critical organ (the thyroid) [40]. The activity of iodine-129 for a mass of 8.09 kg is 52,91 GBq, and its radiotoxicity is 2.7×10^{-1} Sv/g ; values for 1GWe×y [41].

2.2. Thermal power

The thermogenic or non-thermogenic character of the isotopes to be conditioned must be taken into account in the choice of the most appropriate management method. This depends on the residual thermal power at time 't' of each isotope 'i', named P_{th} (i, t). It is expressed according to the equation :

$$P_{th}(i,t) = A(i,t) \times k \times [E_\alpha(i) + E_\beta(i) + E_\gamma(i)] \quad \text{... (Eq. 2.1)}$$

With : P_{th}(i, t) : specific residual thermal power of the isotope 'i' (in Watt).

A(i, t) : Activity of the isotope ‘i’ at time ‘t’ (in Bq).

k : $1.602176487\times10^{-16}$ J/keV is Planck's constant.

E_α, E_β, E_γ : Energies of the α, β, γ decays respectively, in keV.

In the case of iodine, two radioactive isotopes are predominantly present in spent fuel: ^{129}I and ^{131}I. For a UOx fuel, after storage in a pool for 4 years, the activity of iodine-129 is about 1.1 GBq, that of iodine-131 is neglected because of a half-life of 8 d for this isotope, which is to be compared to the cooling time in the pool. Knowing that the β^- decay of ^{129}I produces an electron of 156 keV and an electromagnetic radiation γ of 40 keV, and considering a probability of 1 for each of these two emissions and 0 for the other decay modes, the residual thermal power of iodine-129 brought back per gram of isotope is about 2.1×10^{-7} W. For comparison, the thermal power per gram of cesium-137, an isotope classified as thermogenic, is 0.6 W. Iodine is therefore not a thermal emitter [42].

2.3. Radiotoxicity of Iodine : health effects

The radiotoxicity depends on the dose factor and the mass activity of the element according to the following equation [37]:

$$Radiotoxicity\ \left(\frac{Sv}{g}\right) = Dose\ Factor\ \left(\frac{Sv}{Bq}\right) \times MassActivity\ (Bq/g) \text{.............(Eq. 2.2)}$$

The dose factor depends on the mode of absorption of the element by the body (inhalation or ingestion). Table 2.2 shows the dose factors (for the thyroid, the organ of preferential iodine fixation in mammals), the mass activity and the radiotoxicity (for ingestion or inhalation) for two major radioactive isotopes of iodine [43]. A is the activity of the nucleus in Bq. The activity of a radionuclide is the number of disintegrations of its nuclei per second. It evolves over time as a function of the number of nuclei present N(t) according to the relation [44]:

$$A(t) = \lambda . N(t) \text{.. (Eq. 2.3)}$$

Where λ is the decay constant of the nucleus; it is the probability of decay of a single nucleus in one second. Table 2.2 shows that with a radiotoxicity of the order of a tenth of sievert per gram, the 129 isotope is weakly radiotoxic compared to the 131 one, which is considered very radiotoxic [37].

Isotope	Dose Factor (Sv/Bq)		Mass Activity (Bq/g)	Radiotoxicity (Sv/g)	
	Ingestion	Inhalation		Ingestion	Inhalation
Iodine-129	7.4×10^{-8}	4.7×10^{-8}	6.74×10^{6}	5.0×10^{-1}	3.2×10^{-1}
Iodine-131	1.8×10^{-7}	7.2×10^{-8}	4.60×10^{15}	8.3×10^{8}	3.3×10^{8}

Tab. 2.2. Dose factor, mass activity and radiotoxicity of iodine 129 and 131.

The dangerousness of the element also depends on the effective period of radioactive emission in the body. It corresponds to the time after which the activity in the body will have been divided by two, under the combined effect of radioactive decay and the biological half-life (time after which half of a quantity ingested or inhaled is eliminated from the body by purely biological processes). The effective half-life is determined by the following equation [42]:

$$\frac{1}{T_e} = \frac{1}{T_b} + \frac{1}{T_r} \ldots\ldots\ldots\ldots \text{(Eq. 2.4)}$$

T_e, T_b and T_r are the effective, biological and radioactive periods respectively.

Table 2.3 shows that the isotope (129 or 131), the effective half-life of iodine does not exceed one month [37].

Isotope	Biological period	Radioactive period	Effective period
Iodine-129	30 d	15.7×10^6 y	30 d
Iodine-131		8 d	6,3 d

Tab. 2.3. Biological, radioactive and effective period of iodine 129 and 131.

In the case of iodine-129, the predominant mechanism governing the decrease in its effect on the body is biological elimination, whereas for iodine-131, the mechanism of radioactive decay is predominant. The first clinical signs of radioactive contamination appear from 1 Sv and the annual dose received is about 3.7 mSv (of which 2.4 mSv is of natural origin). The body contains about 15 to 20 mg of iodine, which would represent about 6,4 mSv if all of this iodine were iodine 129 (i.e., 6 times more than the dose obtained artificially per years) [37]. In radiation protection, we are interested in the energy deposited by the radiation in the unit of mass of the material, which is called the absorbed dose (in J/kg, or Gray - abbreviation Gy) [28].

2.4. The risks of Iodine

They can be radiological, chemical or physical (heat). When we talk about nuclear waste and the associated risk, we are interested in the radiological risk. A distinction is made between the potential risk, to which any person would be subjected without any radiation protection measures, and the residual risk, which takes into account possible protection. The effect on living matter will differ according to the nature of the radiation and its energy. The equivalent dose (and radiation weighting factor W_r), measured in Sievert Sv, is defined by the relation [28]:

$$Equivalent\_Dose = W_r \times Absorbed\_Dose\ (Gy) \text{.. (Eq. 2.5)}$$

While the quantity W_r is characteristic of the radiation alone, it does not have the same effect depending on whether it reaches a particular tissue or organ. Hence the introduction of a tissue weighting factor W_t and an effective dose:

$$Effective\_Dose\ (Sv) = W_r \times W_t \times Absorbed\_Dose\ (Gy) \text{...........................(Eq. 2.6)}$$

A distinction is made between external irradiation and contamination; the latter, if the elements are fixed on the organs, may be followed by internal irradiation. As the definitions below are hardly operational, the International Commission on Radiological Protection (ICRP) evaluates the dose, following an ingestion or an inhalation of a given radioelement, by the following formula:

$$Effective\_Dose\ (Sv) = Activity\ (Bq) \times Dose\_Factor\ (Sv/Bq) \text{............... (Eq. 2.7)}$$

The committed effective dose factor is evaluated using a model of the pathway of the radioelement in the body [28].

2.5. Generated flow and current management

In the nuclear power industry, iodine comes from the fission of uranium-235 and plutonium-239. Several isotopes of iodine are present in spent fuel. Table 2.4 summarizes their characteristics [45]:

Iodine isotope	Emission	Period	Mass Activity (Bq/g)
127	Stable	Stable	/
129	β^-, γ	15.7×10^{6} y	6.74×10^{6}
131	β^-, γ	8 d	4.6×10^{15}
133	β^-, γ	20.8 h	4.2×10^{16}
135	β^-, γ	6.6 h	1.3×10^{17}

Tab.2.4. Characteristics of iodine isotopes in spent fuel.

Of the different isotopes present in the spent fuel, only iodine-127 and iodine-129 remain to be considered after storage for 4 to 5 years for cooling. Since the half-life of iodine-129 is nearly 16 million years, and its activity nearly 7 MBq/g (or 7 GBq/kg), iodine-129 is considered to be a long-lived intermediate-level waste (LLIL) [42].

Table 2.5 illustrates the amount of iodine contained in a fuel rod [37].

Fuel generation	UOx1	UOx2	MOx
Cooling time	4 y	5 y	5 y
^{235}U enrichment	3.5 %		
tihm/y Base	800		
Burn-up	33 GWd/t	45 GWd/t	45 GWd/t
^{127}I	37 g ≡ 29.6 kg/y	52 g ≡ 41.6 kg/y	37 g ≡ 50.4 kg/y
^{129}I	169 g (1.1 GBq) 135.2 kg/y	234 g (1.5 GBq) 187.2 kg/y	282 g (1.8 GBq) 225.6 kg/y
^{131}I	Negligible quantity after a few years of storage due to the radioactive period of 8 days.		

tihm: ton irradiated heavy metal

Tab. 2.5. Quantity and activity of Iodine produced per ton of irradiated heavy metal fission after storage in pool for cooling.

The higher the burnup in the reactor, the greater the amount of iodine (two thirds more for MOx fuel than for UOx).

We can also note that in a fuel, before cooling in a pool, the mass quantity of iodine per isotope is distributed as follows: 15 % iodine-127, 82 % iodine-129 and 3 % iodine-131 [37].

At the spent fuel reprocessing plant, iodine is present in three types of waste streams, liquid, solid and gaseous, with a heterogeneous distribution:

- 96 % by mass of the iodine inventory is in liquid form, which undergoes isotopic dilution by discharge into the sea;
- 3 to 4 % by mass of the iodine inventory is trapped on filters in solid form (silver iodide). These filters are currently stored awaiting a storage solution;
- Approximately 1 % by mass, corresponding to the residual fraction, is divided between atmospheric releases and immobilization in high activity glasses.

The source reactions for the production of iodine-129 involve either activation or fission. In the field of nuclear waste, iodine-129 is produced from the reprocessing of spent fuel. Iodine-129 is not produced directly by the fission of uranium-235 or plutonium-239, but by the intermediary of fission products: Antimony-129 and two isomers of Tellurium-129.

The total ^{129}I/I ratio in the reactor is 80 %. The amounts of iodine present in the fuel at the time of reprocessing depend essentially on the neutron flux and the duration of irradiation in the reactor and the cooling time after leaving the reactor, Table 2.6 [46].

Fuel	PWR	PWR	MOx	FBR
Burnup	33000	45000	45700	85000
Fissile content (cooled after 1y)	3,5 % ^{235}U	3,5 % ^{235}U	6,95 % Pu + Am	28 % Pu + U
^{129}I/I (g/t fuel)	208 (1.11 GBq/t)	290	390	756 (3.071 GBq/t)

PWR: Pressurized Water Reactor FBR: Fast Breeder Reactor MOx: Mixed Oxides of uranium and plutonium

Tab. 2.6. Quantities of iodine produced in different reactors.

For a reprocessing plant producing of 1500 t/y of waste, the amount of iodine leaving is only 0,3 t/y [47].

3. Reprocessing of Iodine-129

The behavior of iodine during reprocessing has been the subject of numerous studies in several countries. Two specific features of the chemical element iodine should be noted: the variety of its oxidation states (I^-, I^0, I^{+1}, I^{+5}) and the ability to form organic compounds easily; in addition to this, the volatility of certain compounds (elemental iodine, organic iodine compounds) [48]. Iodine in fuel is present in the form of soluble iodides; and in particular cesium iodide. The principle of separation of the Iodine contained in the dissolution of irradiated fuel is based on two points :

- ✓ Oxidize non-volatile iodides into volatile iodine, this is favored by the presence of HNO_3;
- ✓ Have a sufficient gas/liquid ratio to de-sorb the Iodine from the solution, this is promoted by boiling.

The trapping of gaseous iodine is carried out according to two processes:

- ✓ By absorption in liquid phase;
- ✓ By adsorption on a solid support.

Studies carried out on the trapping of gaseous iodine in the liquid phase have naturally focused on the precipitation of very poorly soluble compounds such as silver iodide, lead iodide or copper iodide or barium iodate, but also on adsorption on sorbents such as silver clinoptilolite or natural cinnabars and ion exchange on an inorganic ion exchanger. The fate of iodine 129 in the reprocessing of irradiated fuel is presented in Figure 2.1 [49].

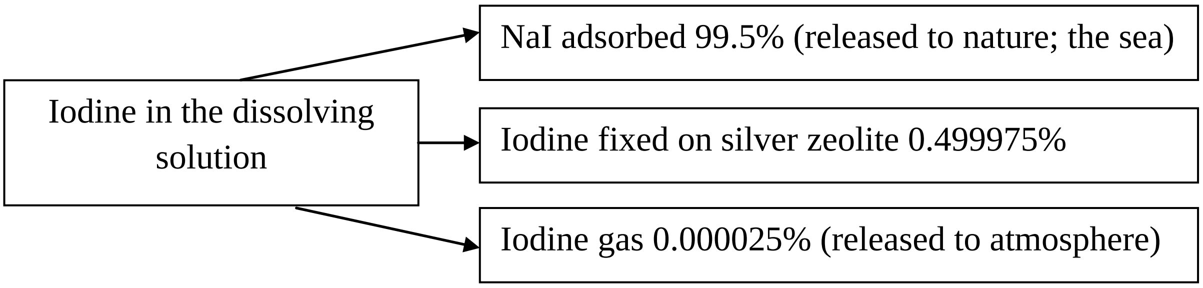


Fig. 2.1. The fate of iodine-129 in the reprocessing of irradiated fuel.

4. Alternative management of Iodine-129

4.1. Advanced separation

Plutonium is the principal element contributing to the radiotoxic inventory, followed by minor actinides and fission products. This observation must be qualified according to the mobility of these elements in the geosphere. Indeed, some fission products, as iodine or cesium, are more mobile than actinides and are likely to have a greater impact on the environment [50]. The distribution of the various radionuclides in the spent fuel is as follows: for 500 kg of uranium initially contained, the fuel consists, after use, of 475 kg of residual uranium, 5 kg of plutonium, 20 kg of fission products, and less than 500 g of minor actinides [21].

The reference separation scheme for these radionuclides is shown in Figure 2.2 below [43].

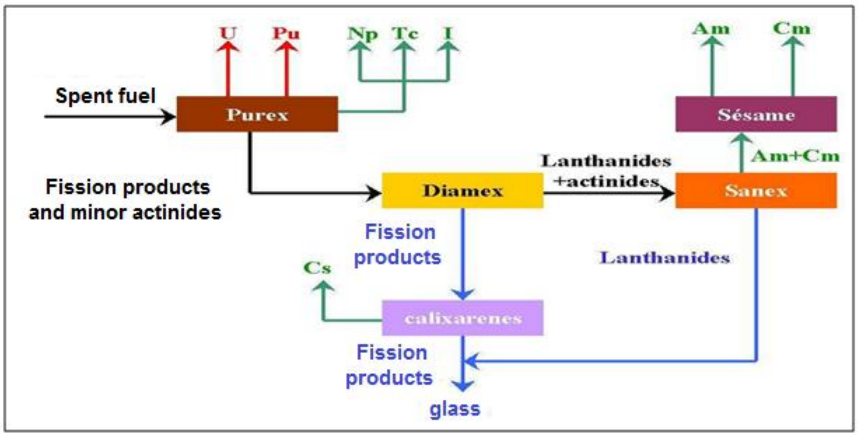


Fig. 2.2. Reference scheme for the separation of long-lived radionuclides.

The separation of long-lived radionuclides is based on the industrial maturity of the PUREX (Plutonium Uranium Refining by EXtraction) process, which already allows the separation of uranium and plutonium [51]. Filters can then be used to isolate the gaseous iodine by precipitation of an insoluble compound (yield of about 99 %) [52]. It is also possible to take advantage of the extracting capacities of tributyl-phosphate to recover neptunium and technetium [53]. The addition of downstream steps then allows the separation of the other main radiotoxic elements.

The actinide/lanthanide separation, because of very similar chemical properties, requires the succession of three steps: their non-selective isolation from the fission products by molecules of the diamide family in the DIAMEX (DIAMide EXtraction) process (yield greater than 99.9 %) [54], then that of americium and curium by various polydentate nitrogenous molecules in the SANEX (Selective Actinides EXtraction) process [55] and finally the separation of these elements by the SESAME (Selective Extraction and Separation of Americium by Means of Electrolysis) process [56]. The extraction yield is 99.1 % for Americium and 97.5 % for Curium.

According to this scenario, at the end of the advanced separation stage, the various radiotoxic elements have been isolated. Their harmfulness by transmuting them into shorter-lived or non-radioactive elements or by confining them in specific matrices.

4.2. Iodine extraction from fission product solutions

In spent fuel, it is often assumed that iodine combines with electropositive elements, and in particular with cesium, which is present in greater quantities (about 2.6 kg per ton of irradiated fuel for UOx fuel). Iodine is therefore considered at this stage in the form of cesium iodide (CsI). The spent fuel is then dissolved while hot in a nitric acid solution. The iodine oxidizes and passes from the oxidation state -1 to 0 (molecular form I_2) which gives it a volatile character (limited solubility in aqueous phase). The di-iodine is carried away by water vapor and they trapped in a soda washing column where it is reduced to form sodium iodide (NaI). It should be noted that a small proportion of the iodine combines with organic residues to form alkyl iodides whose majority representative is CH_3I and which are carried along with the molecular iodine I_2. In contact with water, di-iodine undergoes a dismutation reaction according to the chemical reaction [37]:

$$I_2 + 3H_2O \leftrightarrow 6H^+ + IO_3^- + 5I^-$$

All the iodine in liquid form thus represents 96 % by weights of the iodine inventory and is then released into the sea where it undergoes isotopic dilution (seawater containing Iodine-127 at a rate of 50 μg/L, Figure 2.3 [37]. In practice, the conditions of dissolution of the spent fuel thus ensure a separation in principle of the iodine from the other fission products. In the gaseous outlets of the dissolution reactor, iodine is associated only with krypton 85 and carbon-14. Only the latter is likely to interfere with iodine during subsequent trapping and therefore to be associated with the iodine that exists and allows the isolation of this elements from carbon-14.

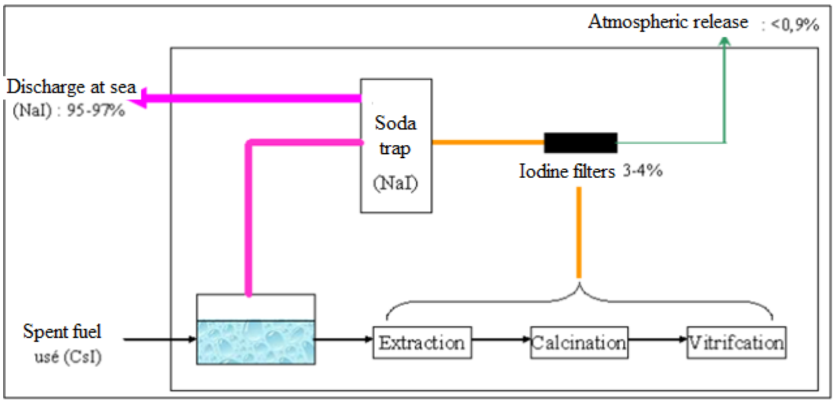


Fig. 2.3. Schematic diagram of iodine management in the reprocessing plant.

They can either be based on extraction techniques from liquids (which requires a selective trapping step upstream of the soda trap, which is then a secondary trap mainly intended to immobilize the carbon-14), or on filtration techniques using series of zeolites whose cage dimensions are adapted to the nature of the elements to be fixed [45]. It should also be noted that the principle of fixing the entire iodine flow from the dissolution reactor on a solid support has already been validated on an industrial scale, since this solution has already been implemented at the Rokashomura spent fuel reprocessing plant in Japan, where the iodine flow is trapped in the form of silver iodide in alumina or silica-based supports [45].

4.3. Iodine trapping

4.3.1. Activated carbons

If iodine management by isotopic dilution were to be completely abandoned, it would be necessary to capture the entire iodine inventory in the gaseous emissions from the reprocessing plant. This trapping would always be done on a solid support, such as porous alumina-based supports impregnated with silver nitrate. However, these supports are not the only ones that can be used. Indeed, there is a great diversity of materials studied for the trapping of iodine. Among them are activated carbons, most often impregnated, which have excellent trapping capacities, but which present risks of explosiveness [37].

4.3.2. Zeolites

Zeolites are also widely studied as iodine filters. They are porous materials with a controlled geometry, and their iodine trapping efficiency depends on their stability under acidic conditions (the iodine to be captured can be found within acidic flows), this being higher when the Si/Al ration increases. Mordenite ($Al_7Si_{17}O_{48}$), impregnated with silver, has a high Si/Al ratio of 2.4 and is therefore particularly used for iodine capture. MOF (Metal Organic Framework) materials can also be found, such as ZIF-8 (Zeolit Icimidozalate Framework-8) which has higher adsorption capacities than mordenite. Finally, more original materials have also been developed in recent years, such as microporous polymers or porous argaeoles based on cellulose impregnated with silver or based on chalcogenides [42].

The trapping of iodine on a solid support (as well as its eventual advanced separation) opens the way to the possibility of alternative management of this element. Two types of management can theoretically be envisaged : management by transmutation (as for minor actinides) or conditioning for deep geological disposal.

5. Conditioning (packaging) of iodine-129

The objective of a conditioning matrix is to guarantee the durable containment of radionuclides. It must therefore meet a certain number of specificities determined by the properties of the isotopes to be immobilized. In particular, it must have good resistance to irradiation damage. These corresponds to the breakage of chemical bonds and atomic displacements created by the production of daughter nuclei, helium nuclei and radiation during radioactive decay. Because of the thermogenic power of certain isotopes, a conditioning matrix must also be able to withstand significant heating during the first years of storage. In addition, in the event of contact with a vector for the dissemination of radionuclides into the biosphere such as water, it must have good resistance to leaching, i.e., a high degree of chemical durability [43].

The matrix considered for the conditioning of iodine-129 belongs to the mineral family of apatites; it has the formula $Pb_{10}(VO_4)_{1.2}I_2$ [38]. This phase is obtained by reaction in a closed environment between a lead vanado-phosphate with lead iodide, which is the envisaged form of the waste, according to the balance equation [57]:

$$3Pb_3(VO_4)_{1.6}(PO_4)_{0.4} + PbI_2 \rightarrow Pb_{10}(VO_4)_{4.8}(PO_4)_{1.2}I_2$$

The conditioning material is obtained by sintering under load at 700°C under 25 MPa from a PbI_2 core embedded in a $Pb_3(VO_4)_{1.6}(PO_4)_{0.4}$ gangue. The initial rate of alteration of iodo-apatite at 90 °C, based on the released of iodine, at pH 5.7 (25 °C), in a strongly renewed medium is equivalent to $2.5.10^{-3}$ g/(m^2.d). This rate decreases by a factor of 20 after one year ($4.4.10^{-4}$ g/(m^2.d)). Iodine release is then limited by diffusion through a residual layer of lead phospho-vanadate of formula $Pb_2V_{1.16}P_{0.27}O_{7.66}$ [57].

II. Transmutation of Iodine-129

1. Basic concepts

Transmutation consists in modifying the nuclei of certain isotopes to transform them into stable or shorter-lived bodies. Three solutions have been considered to achieve these results : photonic transmutation, proton transmutation and neutron transmutation.

Since they have no electrical charge, neutrons, through capture and fission phenomena, appear to be best candidates for transmutation of radionuclides [58]. Produced in the core of nuclear reactors, they have the advantage of being able to use existing or future reactors. Efficiency of transmutation depends on the neutron cross-sections of capture and fission, expressed in barns (b), which are complex and vary from one element to another, but also, for the same element, from one isotope to another [58].

2. Benefits and risks

The benefit/risk ratio of transmutation appears to be quite different for long-lived fission products than for actinides, as the following arguments show [59]:

- A reduction in the inventory of radiotoxic actinides in stored waste reduces the risk of storage facilities in human intrusion scenarios. Also in this case, the number of storage facilities or the volume of waste stored at each facility could be reduced.
- Exploitation of the full energy content of actinides could reduce fuels costs and waste streams from mining and spent fuel. However, transmutation could also induce new risks due to a more complex fuel cycle or to a change in safety parameters. In this respect, the fraction of delayed neutrons and the reactivity coefficients depending on temperature and vacuum are relevant.
- After decay of the shorter-lived waste in the storage facilities, a weapon material could be extracted from the spent fuel and, therefore, future proliferation and criticality risks could be eliminated by separation/transmutation. On the other hand, the separation/transmutation process itself has increased these risks to the present population.
- The dose risks from leakage of mobile elements such as Rn-222 and the metalloid fission products technetium and iodine should be reduced. The price to be paid is related to the efforts needed to avoid the ecological risks of the separation/transmutation process itself, since radiotoxicity should not be spread by procedures such as machining or solvent spillage. It seems sensible to distribute the efforts in such a way that the total dose risk is minimized for a given amount of spent fuel.

3. Transmutation process

The isotopic composition of iodine in the spent fuel is 16 ~ 23 % for iodine-127 and 77 ~ 84 % for iodine-129. By neutron capture, ^{129}I produces two states of ^{130}I, an isomeric (metastable) state noted ^{130m}I produced at ~ 66 % with a half-life of 9 min, and a fundamental (ground) state noted ^{130g}I produced ~ 33 %, with a period of 12.36 h. ^{130m}I makes an isomeric transition at 84 % characterized by the emission of a photon of 39.95 keV, and de-excites by β^- emission, at 16 % towards the excited states of ^{130}Xe. These levels transition to the stable ground state by emission of γ photons [41].

The ^{130g}I, formed directly by neutron capture and indirectly by isomeric transition decays in 12.36 h entirely by β^- radioactivity toward the excited levels of ^{130}Xe; as before a γ-cascade signs the relaxation towards the fundamental. The γ rays at 536.1 keV and 586.1 keV are present in each of the decays of the ^{130}I states, which will have to be taken into account in the analysis of the spectra. The rest of the de-excitations consists of electronic captures forming ^{128}Te, with half-life 7.7×10^{24} y. Figures 2.4 and 2.5 schematize these decays, with the direct and total level population probabilities in parentheses respectively [60]. Table 2.7 shows the strongest γ lines of ^{130}I and ^{128}I [60].

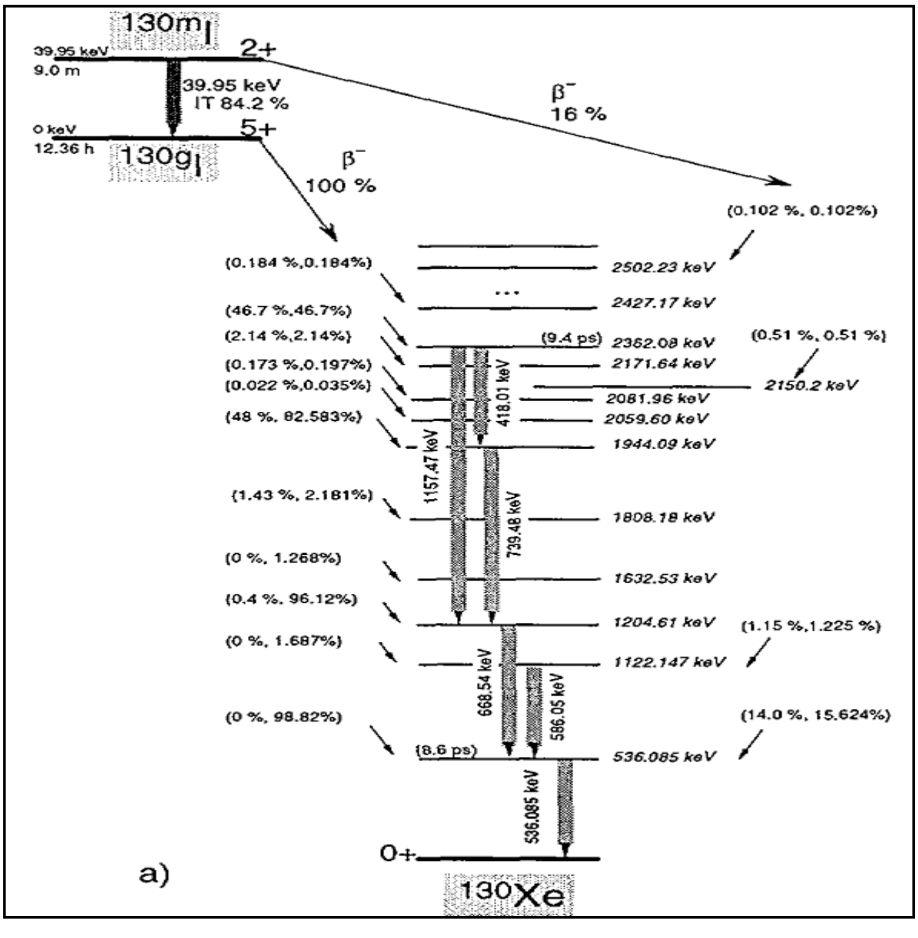


Fig. 2.4. Simplified diagram of the de-excitation of ^{130m}I and ^{130g}I.

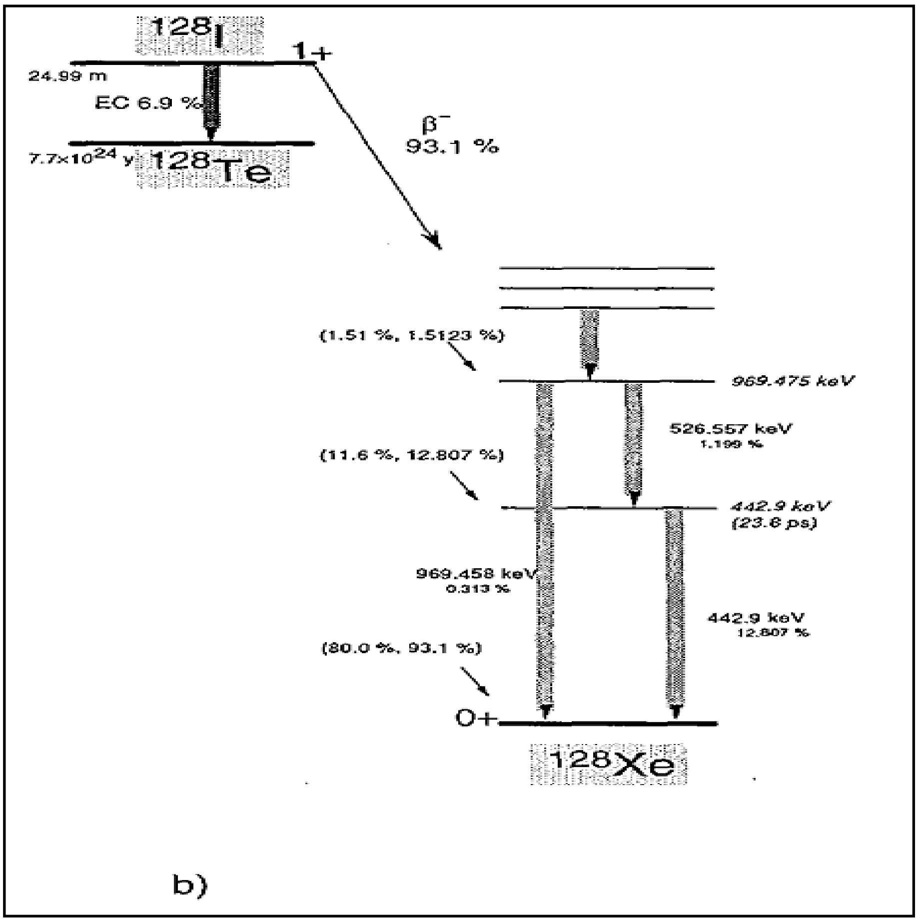


Fig. 2.5. Simplified diagram of the de-excitation of ^{128}I.

Energy (keV)	(Intensity ± err) %		Energy (keV)	(Intensity ± err) %
^{130m}I (9.0 min)			^{130g}I (12.36 h)	
536.1	15.643 ± 0.1		418	34.57 ± 0.1
586.1	1.06 ± 0.01		536.1	98.823 ± 0.1
^{128}I (24.99 min)				586.1
442.9	12.807 ± 0.1		668.6	96.199 ± 0.1
526.6	1.1993 ± 0.01		739.5	82.583 ± 0.1
			1157.5	11.41 ± 0.1

Tab. 2.7. Most intense γ rays of ^{130}I and ^{128}I.

4. Ways of transmutation

4.1. Use of photons

If a target is bombarded with a beam of electrons provided by an accelerator, it is possible to obtain photons (Bremsstrahlung effect) which can give rise to reactions (γ, xn), i.e. x neutrons are rejected from the nucleus under the effect of incident gamma radiation. By applying it to bodies too rich in neutrons such as some fission products that are unstable. These reactions lead to stable bodies. This method has not been adopted, given the very low yield and the high level of electron current required, which is still unattainable [61].

4.2. Use of protons

This way was proposed in the early 90's, using accelerator-assisted subcritical reactors, which are called hybrid systems or ADS (Accelerator Driven System). ADS must be able to deliver high currents (tens of mA) at energies in the GeV, with a high level of reliability. The protons reach a target in which a reaction called spallation occurs. Each proton emits on average about thirty neutrons which are added to the fission neutrons. Very big difficulties are posed for the spallation target [28].

4.3. Use of neutrons

Produced in the core of nuclear reactors, the efficiency of transmutation depends on the neutron capture and fission cross-sections which are a function of the neutron spectrum (flux, energy) [43].

Fission products are produced by capture reactions and the mass inventory at the exit of a PWR (Pressurized Water Reactor) is dominated by plutonium (after uranium). The use of higher burnup or MOx fuels increases the relative proportion of heavy transuranics. The total quantity of fission products (FP) is directly proportional to the thermal energy released by the core. The formation of LLFPs depends only slightly on the fissioning nucleus, and the only way to create less LLFPs, is to increase the proportion of energy actually used. Making cogeneration or high temperature reactors, produces less FP.

The average yield of iodine-129 is 7.7 kg/y in a PWR of 900 MWe (UOx 3.5 % at 33 GWj.t^{-1}) of discharge [25].

Reaction cross-sections are the physical quantities that drive the evolution of nuclei in the reactor. The cross-section σ reflects the probability for the reaction to take place. For a number N of target particles in a flux Φ of incident particles, the reaction rate R is by definition of σ:

$R = \sigma . \Phi . N$ .... (Eq. 2.8)

The cross-section is a function of the nature of the nucleus and the energy of the incident neutron. In the case of iodine-129, cross-section is 0.35 b for the fast neutron spectrum, and 3.12 b for the thermal neutron spectrum. These values were calculated by the ORIGEN code for a thermal flux of the order of 1×10^{14} n.cm^{-2}.s^{-1} and a fast flux of the order of 1×10^{15} n.cm^{-2}.s^{-1} [62].

5. Transmutation mode

The only realistic principle of transmutation envisaged is neutron irradiation, inducing fissions on the heavy elements, and captures on the fission products FP to result by β^- transition in short-lived isotopes. The fission reactor is considered the only device where a high enough flux of neutrons can be maintained for years.

The addition of fission products (FPs) in the core of a reactor modifies its neutronic behavior and may therefore require new assembly configurations. Two modes of transmutation are distinguished, homogenous and heterogeneous. The choice between these two modes depends on the behavior of each element in the reactor, and its influence on the fuel cycle [63].

5.1. Homogenous mode

In this case, the fission product is homogeneously mixed with the fuel, figure 2.6 [64].

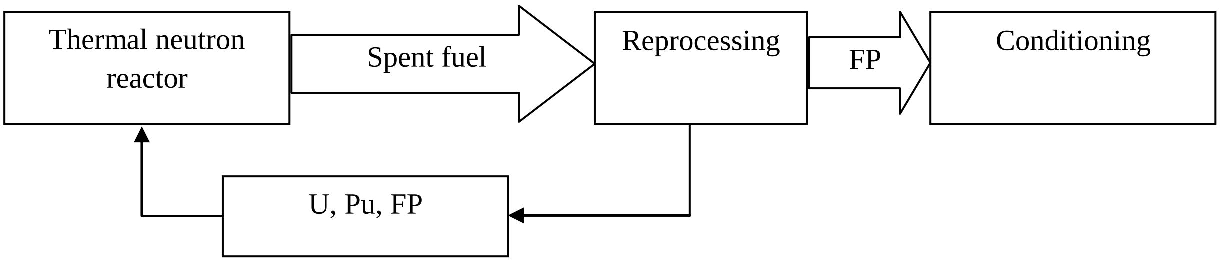


Fig. 2.6. Transmutation in homogenous mode.

5.2. Heterogeneous mode

The fission products are located to be transmuted into special rods without any fuel. These rods, will form groups of 24 units intended to be inserted into the guide tubes of the 17×17 assembly exactly as control groups. The fissile content of the assemblies receiving these groups is kept identical to those without groups. Figure 2.7 describes the heterogeneous mode [64].

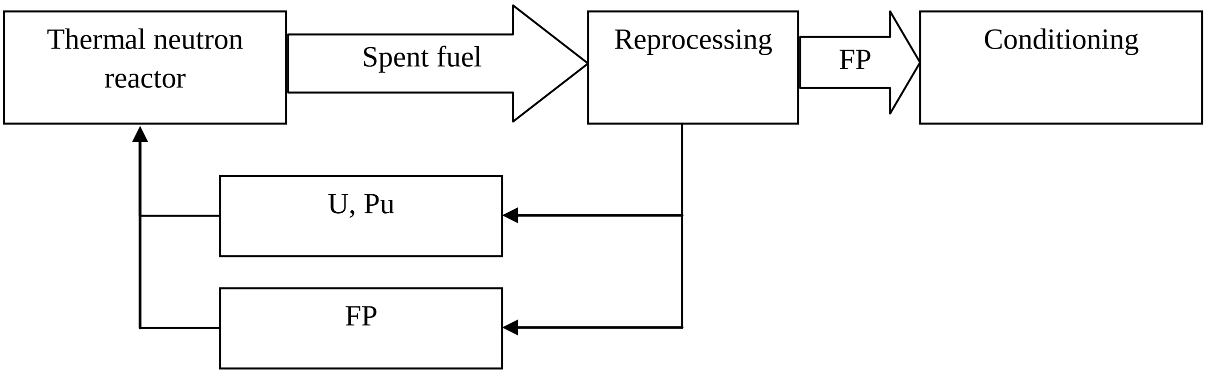


Fig. 2.7. Transmutation in heterogeneous mode.

6. Transmutation of long-lived fission products : Iodine-129

6.1. Introduction

The long-term risk of a geological repository for nuclear waste is generally dominated by the LLFP citing iodine-129; ^{129}I. The latter is generally more mobile than the actinides. Iodine also has a relatively high radiological toxicity. In most repository designs, the early dose contribution from primarily LLFP and their order of importance depend on the characteristics of the repository. To reduce the long-term dose, it has been suggested that LLFP be transmuted into stable or short-lived isotopes, or that they be specially packaged for disposal.

Activities on LLFP transmutation are mainly focused on the transmutation of iodine-129 and technetium-99, since they can be transmuted quite efficiently in nuclear reactors due to their standard neutron absorption cross-section (30 b for iodine-129).

6.2. Neutronic consideration of the transmutation of Iodine-129

The elemental iodine in spent fuel consist of iodine-127 and iodine-129, with a typical composition of 23 % ^{127}I and 77 % ^{129}I in the case of PWR. Because the half-lives of the unstable nuclides ^{128}I, ^{130}I, ^{131}I and ^{132}I are very short, ^{127}I and ^{129}I are mainly converted by neutron capture to ^{128}Xe and ^{130}Xe, respectively. The conversion of ^{127}I to ^{129}I is very small, as ^{128}I decays rapidly to ^{128}Xe. As a result, ^{127}I and ^{129}I are transmuted to stable xenon isotopes by neutron capture reactions. In the thermal energy range, the capture cross-sections of ^{127}I and ^{129}I and ^{130}Xe are inversely proportional to the neutron velocity, and ^{129}I has a much larger capture cross-section than the other two isotopes. ^{129}I has a resonance series, and the resonance integral is 36 b. In an iodine target, the self-shielding effects are generally small [65]. Figures 2.8 and 2.9 illustrate the transmutation process of iodine-129 and the cross-sections of ^{127}I and ^{129}I from ENDF data [65].

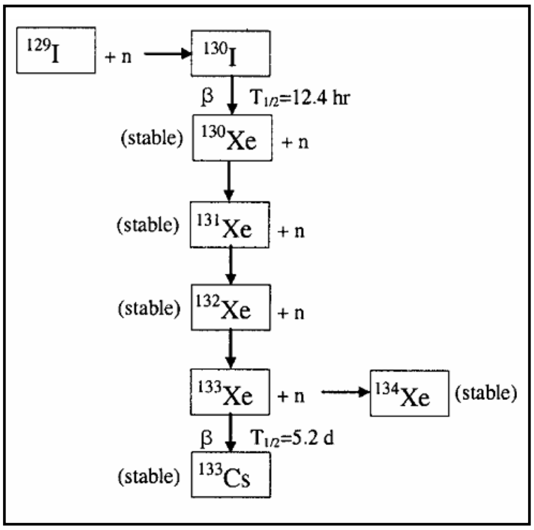


Fig. 2.8. Transmutation process of the iodine-129.

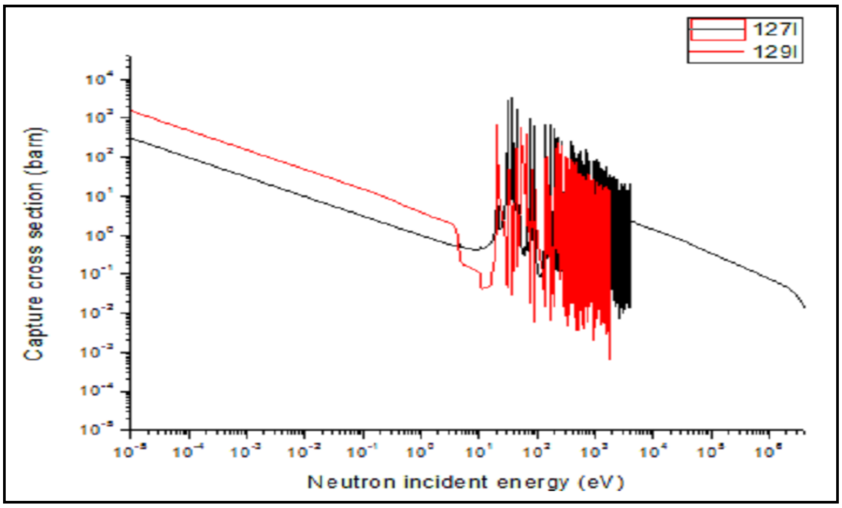


Fig. 2.9. ^{127}I and ^{129}I cross-sections from ENDF data.

6.3. Selection of iodine-129 targets

The target consists of a support matrix, which is transparent to neutrons and allows the adjustment of the power density of the fuel, and a chemical compound containing the element to be transmuted. Irradiation experiments were performed to select the iodine chemical compound, the compatible matrix material and the appropriate moderator material [63].

Elemental iodine, I_2, is not a suitable chemical form of the target for transmutation of iodine-129 because its low melting point of 386 K and boiling point of 457 K and it is very corrosive. Some attempts have been made to select suitable chemical forms for the target through literature study and irradiation tests. They focused on these criteria: (1) high melting point of the metal component, (2) low thermal neutron capture cross-section of the metal component, (3) no production of long-lived nuclide, (4) sufficient iodine content, (5) compatibility with cladding materials, (6) chemical stability in air for safe handling and storage. Unfortunately, no compound met the criteria [66].

The properties of a selection of the most promising metal iodides are summarized in Tables 2.8 and 2.9 [65, 66, 67] and figure 2.10 [66, 67].

Compound	σ_{metal} (b)	N_{atoms} $10^{22}/cm^3$	Melting point (K)
CeI_3	0.73	1.97	1033
MgI_2	0.064	1.94	1906
YI_3	1.30	1.76	1238
NaI	0.543	1.47	934
PbI_2	0.18	1.59	683
ZnI_2	1.10	1.80	719

Tab. 2.8. Properties of some metal iodides (at E_0=0,0253 eV).

Compound	Melting point (K)	N_{atoms} (Iodine) $10^{22}/cm^3$	Production of long-lived nuclide: half-life	Chemical stability
NaI	933	1.47	No	Deliquescent
MgI_2	907	1.84	No	Deliquescent
CaI_2	1052	1.62	^{41}Ca: 1.03×10^5 y	Deliquescent (light sensitive)
CuI	868	1.78	^{65}Zn: 244.26 d; ^{63}Ni: 100.1 y	Stable in the air
$Ca(IO_3)_2$	823	1.40	^{41}Ca:1.03×10^5 y	Stable in the air

Tab. 2.9. Properties of iodine compounds.

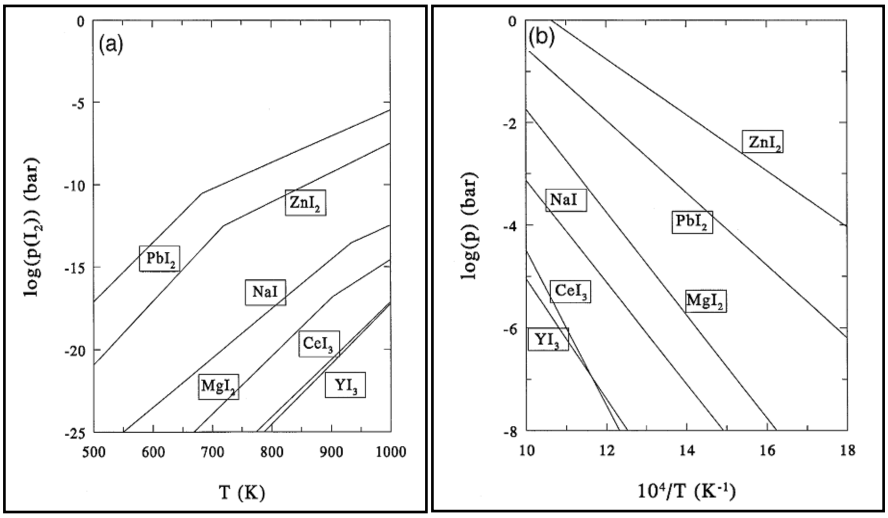


Fig. 2.10. The dissociation pressure $\log(p(I_2))$ as a function of temperature (a) and the vapor pressure as a function of reciprocal temperature (b) of some metal iodides.

Cerium tri-iodide CeI_3 and yttrium iodide YI_3 clearly seem to be the most promising materials, having a relatively low cross-section for low thermal neutron capture, a reasonably high melting point, low vapor pressure and high thermodynamic stability. However, they have the disadvantage of being unstable to moisture and oxygen and are so far difficult to prepare in a pure form. Lead iodide PbI_2 and zinc iodide ZnI_3 are the lowest stable in terms of dissociation from each other, have the highest vapor pressure among the compounds considered, but are relatively easy to handle [65, 66, 67]. These compounds have a relatively low melting point, so they will be in the molten state under the conditions of most power reactors. Magnesium iodide MgI_2 and sodium iodide NaI have intermediate characteristics, and are in most respects less promising than CeI_3 and YI_3. Since NaI is less sensitive to moisture than MgI_2, it seems to be a better candidate [65, 66, 67].

A secondary criterion for target material selection is the compatibility of the target preparation with the reprocessing technology. Liquid scrubbing as well as solid adsorbents have been suggested for the removal of iodine from gas streams in reprocessing facilities. In the case of liquid removal processes, iodine is bound in metal compounds such as NaI in (caustic scrubber), HgI_2 in (Mercuryx) or $Ba(IO_3)_2$ in (Iodox). In the case of a solid adsorbent, only silver Ag (is quite expensive) seems to be an attractive binding agent. PbI_2 and NaI have the advantage of being possible chemical forms in which the fission product iodine can be isolated in reprocessing plants by the PUREX process [65, 67].

6.4. Preparation and characterization of Iodine-129 target

In all transmutation experiments, the iodine targets should contain the natural isotope ^{127}I because it was not possible to prepare samples containing ^{129}I in the required amount (1-2 g). Since the transmutation process of ^{127}I is almost identical. Materials that contain iodine were purified before use, after which they were stored in a glove box filled with argon. PbI_2 and NaI were purified by heating in an argon stream at 400 K, CeI_3 was purified by sublimation at high temperature [68].

The materials will be encapsulated in 15-15 Ti stainless steel tubes, which had an outer diameter of 6.55 mm, an inner diameter of 5.65 mm and a length of 100 mm as shown in the following figure 2.11 [67].

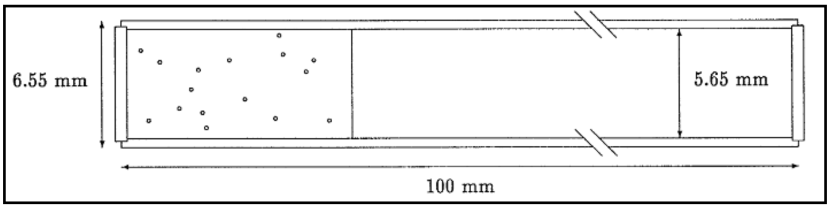


Fig.2.11. The metal iodide irradiation capsule; The left part represents the sample material.

The bottom and top are made of 15-15 Ti rod. The bottom of the capsules were air welded, but due to the highly hygroscopic nature of the materials, the top had to be welded in an inert atmosphere. Before the start of irradiation, X-ray photographs of the targets were taken to confirm that the sample materials do not occupy only the bottom section of the capsule [67].

6.5. Irradiation conditions and transmutation rate of iodine-129

The transmutation of fission products such as iodine has been demonstrated in the JOYO experimental fast reactor (140 MWt thermal power) at the Oarai Engineering Center of JNC (Japan Nuclear Cycle Development Institute) in Japan. Iodine was considered as the target long-lived fission product to be transmuted. In the core fuel region, the total neutron flux is 5×10^{15} n.cm^{-2}.s^{-1}. Many materials have been considered as neutron moderators in fast reactors, including hydride materials (zirconium hydride ZrH), B_4C, graphite and beryllium (Be). The maximum transmutation rate of iodine-129 was 7.7 % $ZrH_{1.65}$ (60 %) as the moderator material [69].

A small fast reactor such as the Fast Flux Test Facility (FFTF) can be an effective transmutation device to destroy long-lived fission products such as iodine. The FFTF is a 400 MWt sodium-cooled fast reactor SFR with a well-characterized irradiation environment and the demonstrated ability to accommodate metal hydride-moderated assemblies. The target form of iodine chosen was cerium iodide CeI_3 because of its quantity of iodine in a given volume. The calculated transmutation rate of iodine-129 was 14 % per year, using yttrium hydride ($YH_{1.7}$) as the moderator [70].

The BR2 material testing reactor at MOL, Belgium with, in current operation, an effective thermal neutron flux ranging, in the core, from 2 to 4×10^{14} $n.cm^{-2}.s^{-1}$, in the reflector position, up to 4×10^{14} $n.cm^{-2}.s^{-1}$, and an important epithermal contribution (about 0.4×10^{14} $n.cm^{-2}.s^{-1}$ in the core position) [71]. The BR2 is a heterogeneous high-flux thermal reactor designed in 1957 by the Nuclear Development Corporation of America (NDC). The reactor is cooled and moderated by pressurized light water (12 bars) in a compact core made of highly enriched uranium; the reflector is a beryllium matrix. The thermal power has been increased to 100 MW. Because of its higher volume concentration of iodine and its higher melting point, calcium iodide CaI_2 was chosen as the reference compound. The percentage of transmutation obtained after 200 days of irradiation (≈ 1 year) is 9.5 %; 7.5 g transmuted on 78.94 g of charge [71]. In another experiment, using the iodine $Ba(IO_3)_2$, for an irradiation time of 200 d, the rate will be 10 % for a neutron flux of 2×10^{14} $n.cm^{-2}.s^{-1}$. This rate will be 23 % for an irradiation duration of 500 d for the same neutron flux [72].

Irradiation tests for the transmutation of technetium and iodine fission products were carried out in the high flux reactor of Petten (Netherlands). Samples of three different metal iodides (CeI_3, NaI and PbI_2) were irradiated in the central C5 position in the core for eight reactor cycles (192.95 days at full power). The calculated total neutron fluence rate was approximately 9×10^{18} $n.m^{-2}$ with a thermal component of 1.5×10^{18} $n.m^{-2}$ for the 8-cycle irradiation period, the total neutron fluence was 1.5×10^{26} $n.m^{-2}$ and the thermal neutron fluence was 2.5×10^{26} $n.m^{-2}$. The transmutation rate was calculated to be about 3 % for the iodine samples [67].

The transmutation of fission products loaded into irradiation tube of the 30 MWth Korean Multipurpose Research Reactor (KMRR) was estimated using the code TRIFON; developed at the Russian Institute of Theoretical and Experimental Physics (ITEP). The transmutation rate of iodine-129 was estimated to be 9,5 % over a burnup period of six cycles (203 days) in the KMRR core center square, a higher flux of 5.3×10^{14} $n.cm^{-2}.s^{-1}$ [73].

7. Transmutation reactor systems

For the implementation of transmutation, studies have focused on the use of these reactor systems [36]:

- Existing reactor systems: i.e. the reactors of the current fleet of pressurized water reactor (PWR) and those to come (sodium-cooled fast reactors SFR), whose technology has been mastered.
- Innovation technology reactors are the fast neutron nuclear reactors developed within the framework of the international "Generation IV" initiative for energy production by 2023, citing gas-cooled fast neutron reactors (GFRs) and molten salt reactors (MSRs).
- Hybrid systems are sub-critical coupled to accelerators, which are still under study.

8. Scientific feasibility of iodine-129 transmutation

The scientific feasibility of transmutation of long-lived waste has been studied for many years by the scientific community. Scientific studies are conducted to thoroughly evaluate the feasibility of recycling these elements in a nuclear reactor and to identify of thermal neutron energy ranges for transmutation of certain fission products, technetium and iodine. The transmutation efficiency depends initially on two key parameters: *cross-sections* of target and *neutron flux level*. Studies have focused on the use of existing reactor systems (PWR, SFR) and innovative technology reactors such as gas-cooled fast reactors (SFR). Hybrid systems have been studied. The evaluation of a waste management solution can only be done with a global view of the cycle (fuel fabrication, reactors operation, cycle plants and reprocessing facilities) with a complete analysis of the whole nuclear cycle through scenario studies [74].

Chapter III

Simulation codes and « The ORIP_XXI collection »

1. Simulation of the parameters of nuclear transmutation

The transmutation rate R is one of the most important parameters for nuclear transmutation. This parameter expresses the amount of radioelement transmuted during an irradiation period. The transmutation rate parameter is determined either experimentally or by evaluation with a suitable calculation code. From the transmutation parameter, several other parameters can be calculated: transmutation quantity C, transmutation support ratio TSR, average transmutation acceleration A.

The transmutation processes of radiotoxic elements directly concern their evolution under irradiation in a nuclear reactor. The Bateman equations are the mathematical models that describe the abundances and activities in a transmutation chain as a function of time. The solution of these equations is performed by various methods: *the search for an analytical solution, the matrix exponential method and discrete numerical methods* [75].

The codes of calculation of nuclear transmutation are numerous: MCNP, Trifon, Citation, D-Chain, Scale, Appolo, Origen2, Darwin, OpenMC [69, 70, 71, 72, 73].

In this work, the evaluation code adopted is the calculation code "ChainSolver 2.34".

2. Theoretical basis of the code

The ChainSolver calculation code is based on the solution of the ordinary differential equation, or Bateman equation. This equation describes the evolution of the states of an isotope according to a spontaneous decay or by transmutation induced by neutrons according to the equation [6]:

$$\frac{dN_i(t)}{dt} = -N_i\mu_i + \sum K_{ji}N_j \quad \text{.........(Eq. 3.1)}$$

N_i: the nuclear density of the state of the i-th isotope.

K_{ji}: the transmutation constant from the j^{th} isotope to the i^{th} isotope.

μ_i: the constant of the i-th state, it is given as follows:

$$\mu_i = \lambda_i + \sum \sigma_g \phi_g k_g \text{ .... (Eq. 3.2)}$$

λ_i: the decay constant of the i-th state.

σ_g: the incineration cross-section.

ϕ_g: the neutron flux.

k_g: the self-shielding factor.

g: the neutron energy group.

2.1. Ordinary differential equation DOE

The decay curve of any radionuclide can be solved by equation Eq. 3.3 [76]:

$$N(t) = N_0 . e^{-\lambda t} \text{ .... (Eq. 3.3)}$$

Where t is the time in years. λ is the decay constant.

N(t) is the size of population of radioactive atoms at a time t.

N_0 is the size of population of radioactive atoms at t=0.

The first step is to determine the decay curves. For any nuclide, the growth-decay equation can be solved by integrating Eq. 3.4 with respect to t.

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \text{ .... (Eq. 3.4)}$$

The equations include both growth and decay. Looking at the basic equation, $\lambda_1 N_1$ represents the growth of the daughter and $\lambda_2 N_2$ represents the decay of the daughter product. For the first daughter product, where there is no growth of the parent; the integration resolves to Eq. 3.5.

$$N_2 = N_{01} \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right) \text{ .... (Eq. 3.5)}$$

For the second daughter product, the basic equation is rearranged, as in Eq. 3.6.

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3 \qquad \text{(Eq. 3.6)}$$

Including the growth of the daughter product, N_2 as a function of time and integration in time, we obtain the equation Eq. 3.7.

$$N_3 = \lambda_1 \lambda_2 N_{01} \left(\frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_2)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right) \qquad \text{(Eq. 3.7)}$$

This method of solving is complicated for several reasons. Accounting for chain branching and joining is computationally intense and is handled by using partial decay constants for growth in the above equations. However, as chains become longer, these equations become unwieldy and progressively more difficult to solve [6, 76] .

2.2. Solving the ordinary differential equation

The evolution of a system subjected to a neutron flux Φ can be calculated by such a calculation code; using the Bateman equation Eq. 3.8. This relation balances the disappearances and appearances of N_i nuclei i over a time interval 'dt' [77].

The first term corresponds to the disappearance of the nucleus i by nuclear reaction (capture, fission). The second term represents its formation by interactions undergone by a nucleus j. The third term is the loss of the considered isotope by natural decay. The last term corresponds to its creation by the set of nuclei j whose decay channels lead to the isotope i.

The first term and the second term correspond to the reaction. The third term and the last one correspond to the decay.

$$\frac{dN_i(t)}{dt} = -\sigma_i^{abs} \phi N_i + \sum_{i \neq j} \sigma_{j \to i} \phi N_j - \lambda_i N_i + \sum_j \lambda_{j \to i} N_j \qquad \text{(Eq. 3.8)}$$

N_i : the number of nuclei i.

$\sigma_i^{abs} \phi$: the absorption reaction rate of the nucleus i.

$\sigma_{j\to i}\phi$: the reaction rate of the nucleus j producing i.

λ_i : the total decay constant of the nucleus i.

$\lambda_{j\to i}$: the decay constant from nucleus j into nucleus i.

The reaction rates and $\sigma_i^{abs}\phi$ and $\sigma_{j\to i}\phi$ are defined by equation Eq. 3.9 where $\sigma(E)$ and $\phi(E)$ are the cross-section and the neutron flux as a function of the incident neutron energy.

$$\sigma\phi = \frac{\int \sigma(E)\phi(E)dE}{\int dE} \text{ ....(Eq. 3.9)}$$

Harry Bateman found a general explicit formula for the quantities by taking the Laplace transform of these variables. The concentration of the nth nuclide after a time t was given by Bateman as follows [77]:

$$N_n(t) = \frac{N_1(0)}{\lambda_n}\sum_{i=1}^{n}\lambda_i.\alpha_i.e^{-\lambda_i t} \text{ ....(Eq. 3.10)}$$

Where α_i is given by the following equation:

$$\alpha_i = \prod_{j=1;\, j\neq i}^{n} \frac{\lambda_j}{\lambda_j - \lambda_i} \text{ ....(Eq. 3.11)}$$

2.3. Incineration in a nuclear reactor

The temporal evolution of a quantity N of an isotope under neutron irradiation Eq. 3. 12 [80] :

$$\frac{dN(t)}{dt} = -\left(\lambda + \sigma\phi\right)N \text{ ....(Eq. 3.12)}$$

Where ϕ is the flux, σ is the cross-section, and λ is the decay constant. If decay is the predominant reaction, then the time evolution of the quantity N is defined by Eq. 3.13.

$$\frac{dN(t)}{dt} = -\lambda N \text{ ....(Eq. 3.13)}$$

This equation leads by integration to Eq. 3.3.

The time needed to halve the initial quantity Eq. 3.14 is then [81]:

$$t_{1/2} = \frac{\ln 2}{\lambda} = \tau.\ln 2 \qquad \text{(Eq. 3.14)}$$

Where τ is the time constant of the isotope.

By extension, if neutron absorption is the predominant reaction, then the time evolution of the quantity N is defined by Eq. 3.15 [81]:

$$\frac{dN(t)}{dt} = -\sigma.\phi.N \qquad \text{(Eq. 3.15)}$$

The solution of this equation is Eq. 3.16 [80].

$$N(t) = N_0.e^{-\sigma\phi t} \qquad \text{(Eq. 3.16)}$$

This equation leads to the concept of effective half-life when transmutation is considered. The effective half-life is defined by the relation Eq. 3.17 [81].

$$t_{1/2} = \frac{\ln 2}{\sigma\phi} \qquad \text{(Eq. 3.17)}$$

3. Nuclear databases

The databases contain a large amount of nuclear data, citing the cross-sections of interaction with a particle stream, decay properties. The data presented in the nuclear libraries have been evaluated from experimental measurements. The main nuclear databases evaluated are [78]:

- ENDF/B: Evaluated Nuclear Data File, United States.
- JEFF: Joint Evaluated Fission and Fusion file, Western Europe.
- JENDL: Japanese Evaluated Nuclear Data Library, Japan.
- CENDL: Chinese Evaluated Nuclear Data Library, China.
- BROND: Biblioteka Rekomendovannykh Otsenjonnykh Nejtronnykh Dannykh, Russia.

These libraries may differ in certain energy ranges, for certain isotopes. Moreover, we can find a disagreement between identical measurements made with different techniques.

4. Simulation Codes "ORIP_XXI"

The transmutation efficiency of a radioelement is estimated by calculating transmutation parameters. The transmutation rate TR, transmutation quantity C, transmutation support ratio TSR, average transmutation acceleration A are quoted.

The determination of these parameters is done either experimentally or by numerical evaluation. In the present work, the calculation code used for the evaluation is ChainSolver 2.34. The cross-section curves presented are obtained from the JANIS code.

ORIP_XXI, a software suite is developed for the study of transmutation chains of radioactive and stable isotopes, i.e. networks of nuclides with possible feedbacks. This suite is developed by E.G. Romanov for the numerical simulation of nuclear transmutation. It is possible to estimate various quantitative characteristics of a nuclide transmutation chain, both under irradiation in neutron fluxes and in case of spontaneous radioactive decays. The main programs of the ORIP_XXI software are :

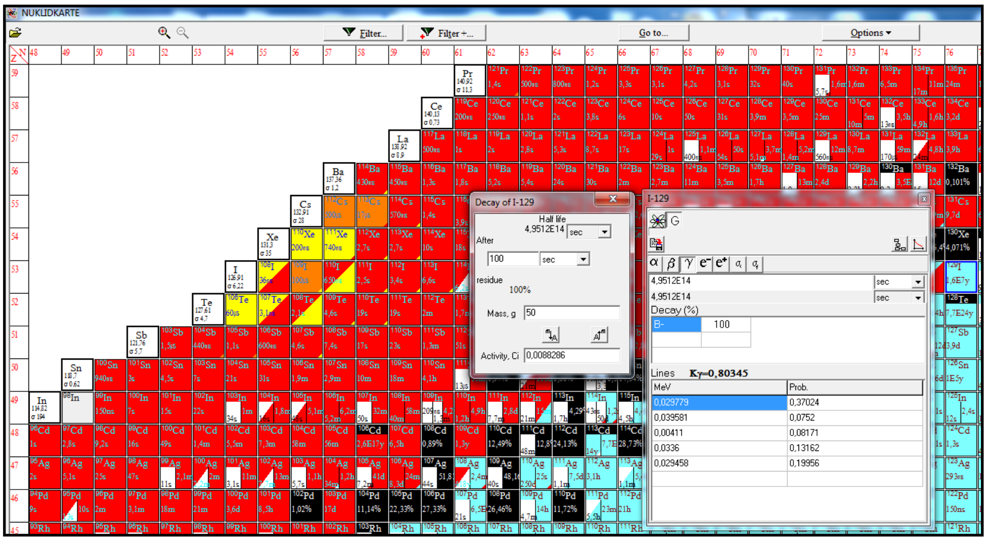
- The NKE electronic card,
- The ChainFinder nuclear transmutation chain search program,
- The complete simulation program of nuclide production in a nuclear reactor ChainSolver.

All programs use a common data file, which contains nuclear constants and decay data for over 2800 nuclides with atomic weights from 1 to 293 (nuclear charge from 1 to 118) and chemical element characteristics. The file also includes fission product yield data for thermal and fast neutron induced fission of 22 heavy isotopes. ChainSolver allows users to take into account neutron flux depression and self-shielding factors. Once the calculations are complete, various features are calculated based on the estimated isotopic densities: isotopic masses, element masses, isotopic activities, isotopic parts, mass yields, depression coefficients, self-shielding factors, and estimated fission energy deposits at all time steps [79].

4.1. The electronic card of nuclides "NKE"

The NKE program is developed to visually represent basic nuclear physics data (half-life, neutron reaction cross-section, radioactive decay data, fission yield, etc.) and to search for nuclides meeting certain criteria, Figure 3.2 [79].

The program can perform complex searches, which allows analysis of all available data in the ORIP_XXI software suite data file. The user interface allows to specify exactly what information should be displayed on the nuclide card. It is possible to quickly switch to any nuclide. When double-clicking in a nuclide cell, general information about the nuclide is displayed. The user can also request detailed information for any isomeric state.



Decay of I-129
Half life 4,9512E14 sec
After 100 sec
residue 100%
Mass, g 50
Activity, Ci 0,0088286

I-129
4,9512E14 sec
4,9512E14 sec
Decay (%)

B-	100

Lines **Kγ=0,80345**

MeV	Prob.
0,029779	0,37024
0,039581	0,0752
0,00411	0,08171
0,0336	0,13162
0,029458	0,19956

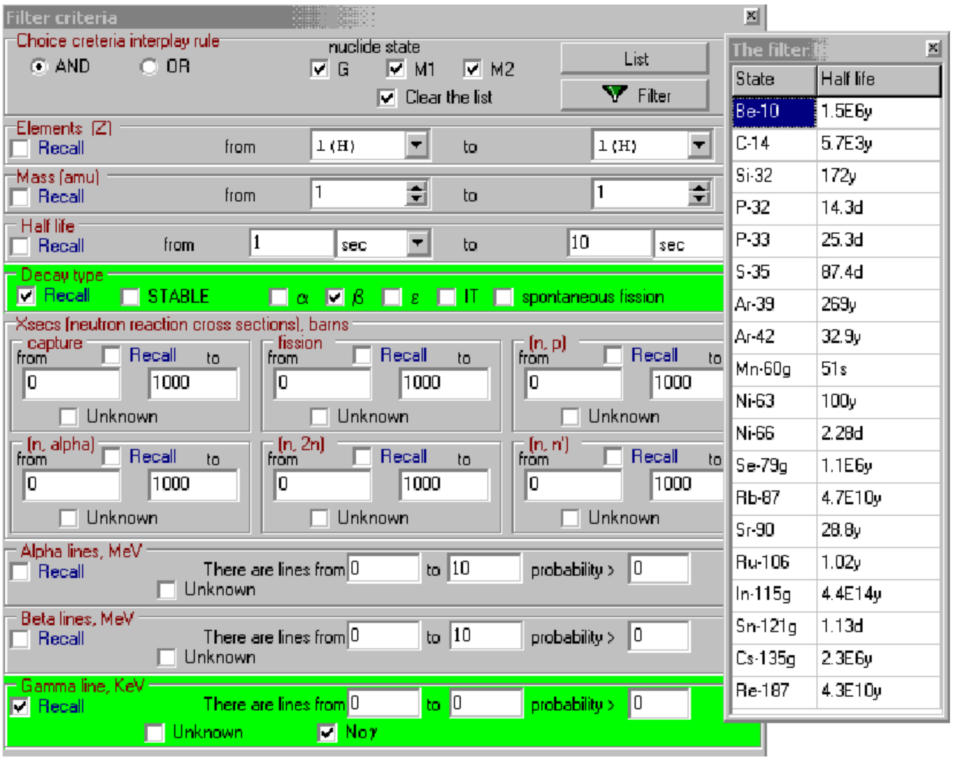
Fig. 3.1. The main windows of NKE program.

The data displayed includes information on:

- ✓ *Neutron reactions for chemical elements*: properties of stable/fundamental/meta-stable states of nuclides (half-life, percentages of decay channels).

- ✓ *Information on particles and their alpha-beta-gamma decays*, emitted electrons and positrons, estimation of the ionizing range constant K_{γ}.
- ✓ *Fission product yields* for various fissile nuclei in fast and thermal neutron spectra; thermal and resonance (capture and fission) cross-sections; cross-sections of (n, p), (n, α), (n, 2n), (n, n') threshold reactions averaged over a fission neutron spectrum.

It is possible to combine several filtering conditions by the operators 'AND' or 'OR', figure 3.2 [80]. Such nuclides seem to be promising for medical applications [79].



Choice creteria interplay rule: ● AND ○ OR

nuclide state: ☑ G ☑ M1 ☑ M2 ☑ Clear the list

Elements (Z): ☐ Recall from 1 (H) to 1 (H)

Mass (amu): ☐ Recall from 1 to 1

Half life: ☐ Recall from 1 sec to 10 sec

Decay type: ☑ Recall ☐ STABLE ☐ α ☑ β ☐ ε ☐ IT ☐ spontaneous fission

Xsecs (neutron reaction cross sections), barns

capture: ☐ Recall from 0 to 1000 ☐ Unknown

fission: ☐ Recall from 0 to 1000 ☐ Unknown

(n, p): ☐ Recall from 0 to 1000 ☐ Unknown

(n, alpha): ☐ Recall from 0 to 1000 ☐ Unknown

(n, 2n): ☐ Recall from 0 to 1000 ☐ Unknown

(n, n'): ☐ Recall from 0 to 1000 ☐ Unknown

Alpha lines, MeV: ☐ Recall There are lines from 0 to 10 probability > 0 ☐ Unknown

Beta lines, MeV: ☐ Recall There are lines from 0 to 10 probability > 0 ☐ Unknown

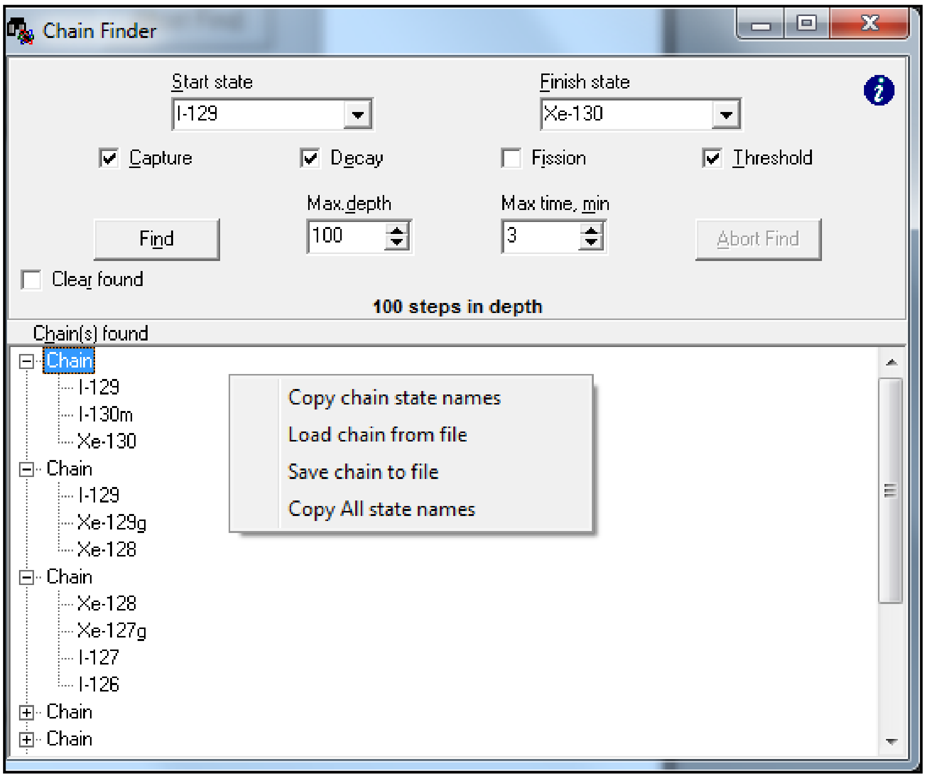
Gamma line, KeV: ☑ Recall There are lines from 0 to 0 probability > 0 ☐ Unknown ☑ No γ

State	Half life
Be-10	1.5E6y
C-14	5.7E3y
Si-32	172y
P-32	14.3d
P-33	25.3d
S-35	87.4d
Ar-39	269y
Ar-42	32.9y
Mn-60g	51s
Ni-63	100y
Ni-66	2.28d
Se-79g	1.1E6y
Rb-87	4.7E10y
Sr-90	28.8y
Ru-106	1.02y
In-115g	4.4E14y
Sn-121g	1.13d
Cs-135g	2.3E6y
Re-187	4.3E10y

Fig. 3.2. Selection of nuclide states of interest to the user.

4.2. Chain search program "ChainFinder"

The main research function of the ChainFinder program is to elaborate the nuclide chains that can be realized in nuclear reactors, figure 3.3. The program also allows to discover the reason for the appearance of unexpected isotopes in the irradiated material. ChainFinder automatically constructs possible transmutation chains, given start and end states. The chain search process takes into account the decay "sub-branch" data available in the data file. Thus, the data used includes all the data in the data file, not just the data displayed by NKE. ChainFinder allows the user to choose whether or not to take into account decay reactions, thermal neutron captures, neutron-induced fission, and fast neutron threshold reactions [79].



Start state: I-129
Finish state: Xe-130
☑ Capture
☑ Decay
☐ Fission
☑ Threshold
Max.depth: 100
Max time, min: 3
☐ Clear found

100 steps in depth

```
⊟ Chain
   I-129
   I-130m
   Xe-130
⊟ Chain
   I-129
   Xe-129g
   Xe-128
⊟ Chain
   Xe-128
   Xe-127g
   I-127
   I-126
⊞ Chain
⊞ Chain
```

Fig. 3.3. Main window of ChainFinder program.

The context menu allows to transfer the list of chain state names to the windows clipboard, or to save a chain to a file. A saved chain can be loaded later into the ChainSolver program to perform nuke production calculations.

4.3. Transmutation simulation program "ChainSolver"

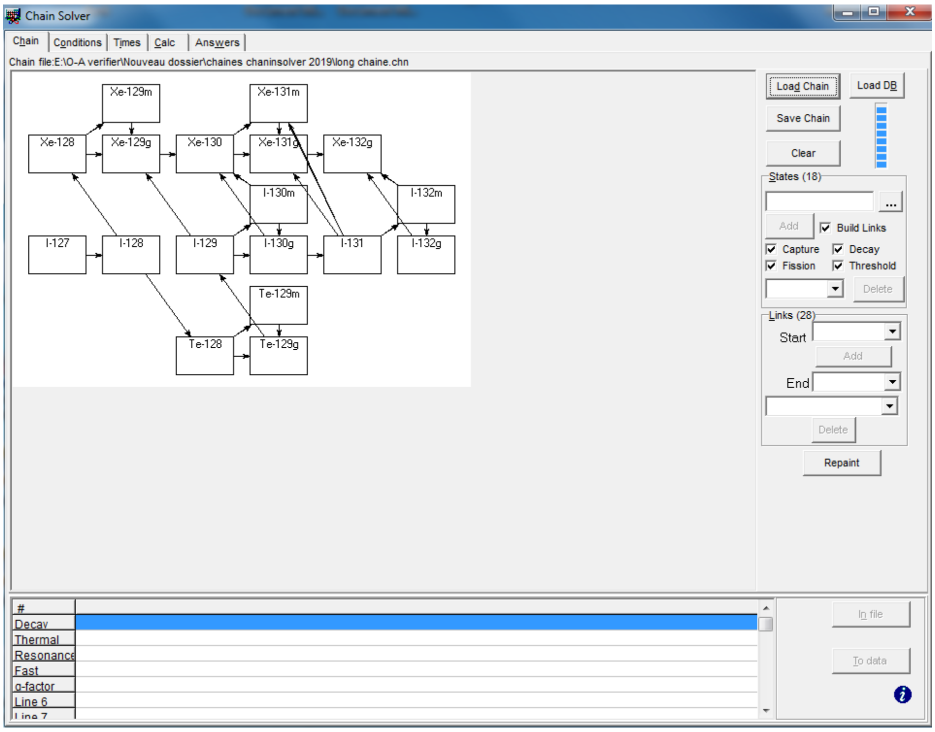
The ChainSolver program is developed for the numerical simulation of nuclear transmutation in the final stage before irradiation or before isotope shipment. Self-shielding by neutron capture resonance, thermal neutron flux depression and irradiation schedule can be taken into account [6, 26, 36, 79].

Making these calculation is extremely tedious because at each irradiation time step, the specific neutron fluxes, the reaction cross-sections and the initial isotopic composition must all be taken into account. The calculations allow to choose more precisely the irradiation mode and to estimate the yield of the products as well as the inevitable impurities. The main approximation used in transmutation simulations is the following: changes in the user-specified neutron spectra due to change in the composition of the irradiation material are assumed to be negligible [6, 26, 36, 80].

The program interface uses a Delphi visual component, which allows to display several pages on a main form. Each page contains a group of parameters necessary for the numerical evaluation of nuclear transmutation and derived quantities.

4.3.1."Chain" tab

The "Chain" tab in the main ChainSolver window allows the user to create a transmutation chain and edit it, figure 3.4. Editing means adding states and/or links changing their numerical characteristics. A link is an arrow corresponding to the transmutation between two isotopic states, possibly two states of the same nuclide. Some associated numerical values can be created in several ways. The chain can be loaded from a file created by ChainSolver. The chain can also be created by starting with an empty chain and adding nuclide states [6, 26, 36, 79].



Chain file:E:\O-A verifier\Nouveau dossier\chaines chaninsolver 2019\long chaine.chn

☑ Build Links
☑ Capture ☑ Decay
☑ Fission ☑ Threshold

Fig. 3.4. "Chain" tab of ChainSolver program.

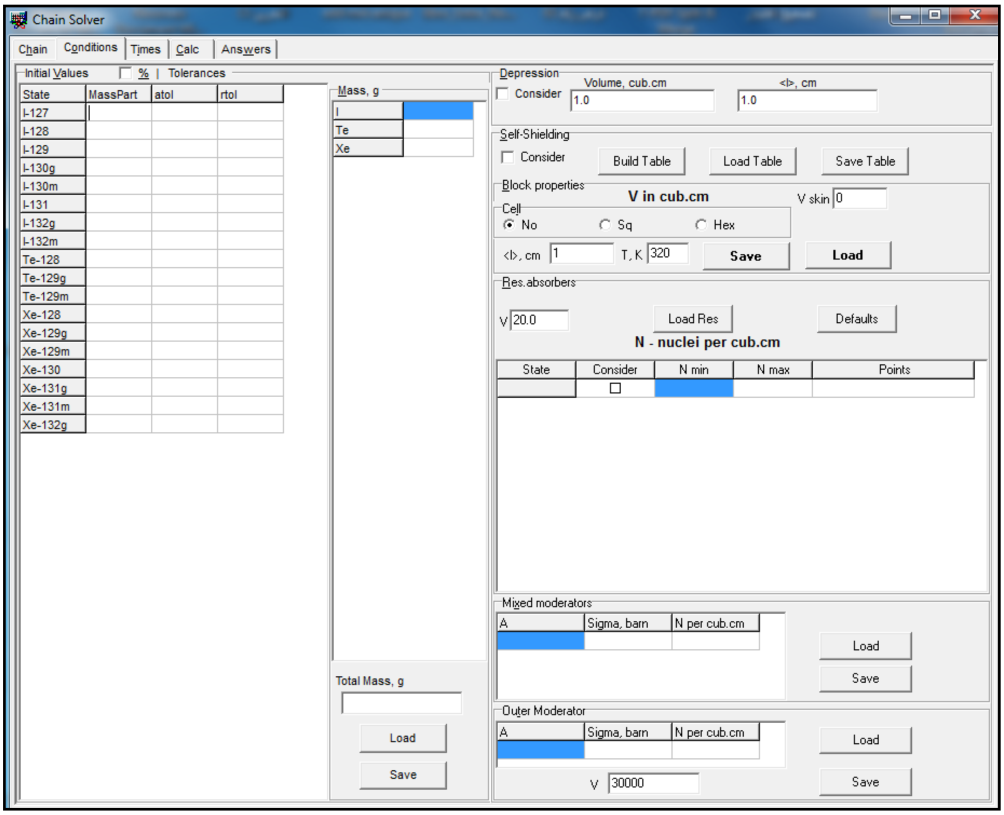
4.3.2. "Conditions" tab

The "Conditions" tab is where the irradiation conditions are defined: the initial composition, the required precision, and the heat flux depression and resonance self-shielding data, figure 3.5. The initial mass parts of the nuclide states, the element weights, a sample piece, and the absolute and relative tolerances (*atol* and *rtol* respectively) [6, 26, 36, 79].

The local tolerance for the ith nuclide concentration y(i) at each time step is determined by the formula:

$$TOL(i) = rtol(i).abs\left(y(i)\right) + atol(i)$$

Calculations taking into account resonance self-shielding are extremely tedious and require many additional parameters.



State	MassPart	atol	rtol
I-127			
I-128			
I-129			
I-130g			
I-130m			
I-131			
I-132g			
I-132m			
Te-128			
Te-129g			
Te-129m			
Xe-128			
Xe-129g			
Xe-129m			
Xe-130			
Xe-131g			
Xe-131m			
Xe-132g			

Mass, g	
I	
Te	
Xe	

Depression: ☐ Consider; Volume, cub.cm 1.0; <l>, cm 1.0

Self-Shielding: ☐ Consider

V skin 0

Cell: ● No ○ Sq ○ Hex

<l>, cm 1 T, K 320

V 20.0

State	Consider	N min	N max	Points
	☐			

A	Sigma, barn	N per cub.cm

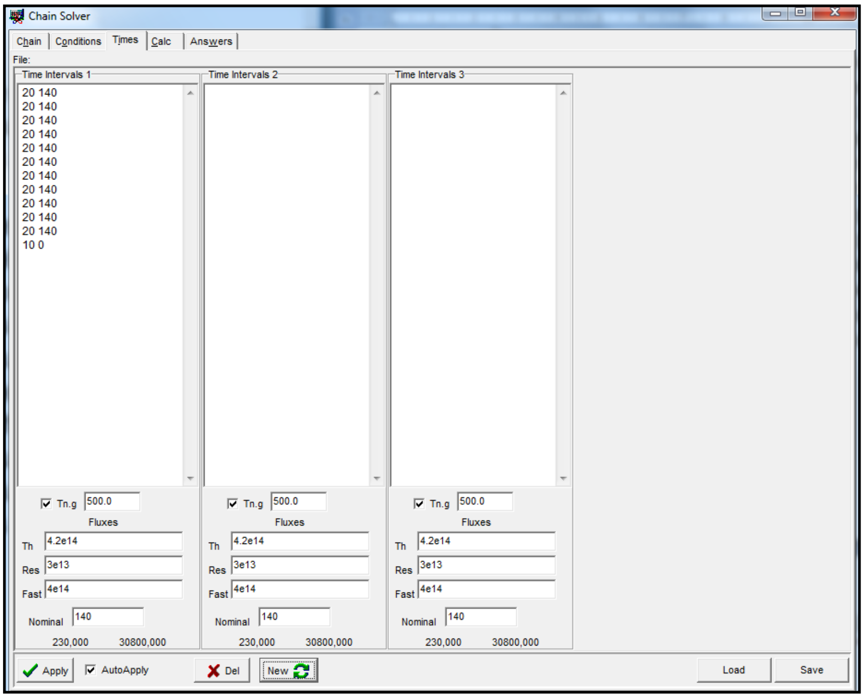
A	Sigma, barn	N per cub.cm

V 30000

Fig. 3.5. "Conditions" tab of ChainSolver program.

4.3.3. "Times" tab

The "Times" page is where the user defines the irradiation program. In the multi-line editor, paired values are defined: time interval (in days) and corresponding power level (in MW), figure 3.6. On this page, the following fields are to be filled in: neutron gas temperature (in degree Kelvin), thermal, resonance and fast neutron fluxes respectively for the nominal power of the reactor. During the calculations, the neutron flux for the time interval is defined as the product of the flux value for the nominal power and the ratio between the current power level and the nominal level. The program allows to simulate multistage irradiation processes when the irradiated material is placed in different positions in the reactor at different stages, or even in different reactors and/or endurance pools with different neutron spectra [6, 26, 36, 79].



Time Intervals 1

```
20 140
20 140
20 140
20 140
20 140
20 140
20 140
20 140
20 140
20 140
20 140
10 0
```

☑ Tn.g 500.0
Fluxes
Th 4.2e14
Res 3e13
Fast 4e14
Nominal 140
230,000 30800,000

Time Intervals 2

☑ Tn.g 500.0
Fluxes
Th 4.2e14
Res 3e13
Fast 4e14
Nominal 140
230,000 30800,000

Time Intervals 3

☑ Tn.g 500.0
Fluxes
Th 4.2e14
Res 3e13
Fast 4e14
Nominal 140
230,000 30800,000

☑ AutoApply

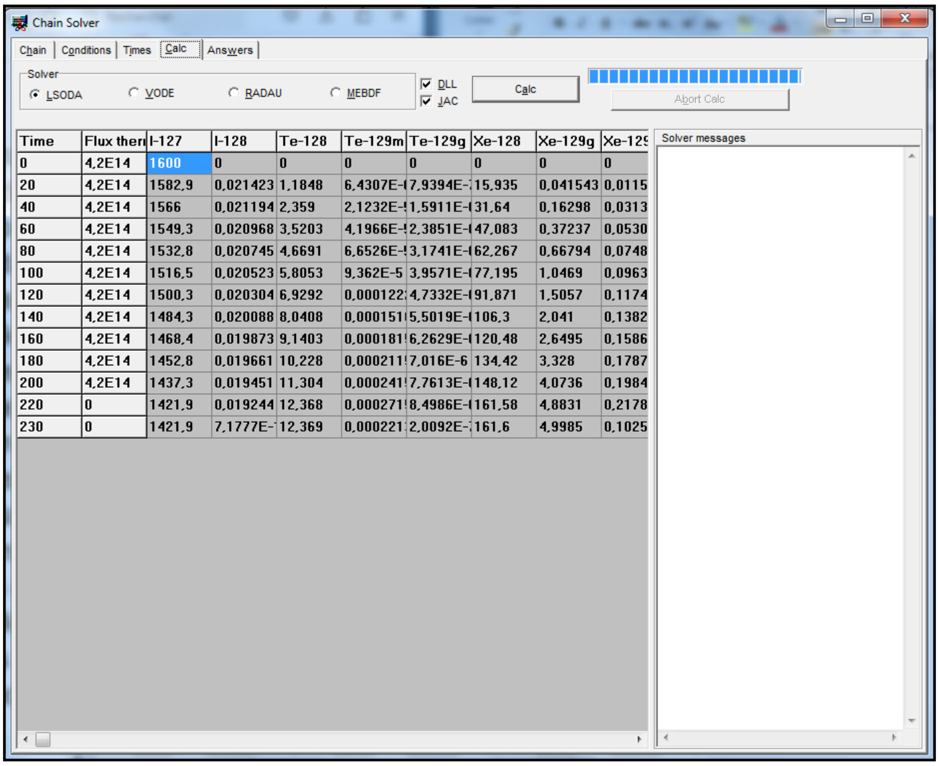
Fig. 3.6. "Times" tab of ChainSolver program.

4.3.4. "Calc" tab

The calculation page "Calc", presented in the figure 3.7, gives the interface to the various methods used to solve the system of ordinary differential equations of Bateman, which describes the chain of transmutation of the nuclide studied. Four solver sub-programs are available in this page: VODE, LSODA, RADAU and

MEBDF, which call upon dynamic libraries loaded "DLL". The solutioners use methods of calculations of the Jacobian, by selecting the box "JAC" [6, 26, 36, 79].

After the selection of the solver, the calculation starts by pressing the "Calc" button. In the table, the calculated weights of all isotopes present in the constructed chain for each time interval are displayed, indicating the corresponding heat fluxes.



Solver: ● LSODA ○ VODE ○ RADAU ○ MEBDF

☑ DLL
☑ JAC

Time	Flux ther	I-127	I-128	Te-128	Te-129m	Te-129g	Xe-128	Xe-129g	Xe-12
0	4,2E14	1600	0	0	0	0	0	0	0
20	4,2E14	1582,9	0,021423	1,1848	6,4307E-	7,9394E-	15,935	0,041543	0,0115
40	4,2E14	1566	0,021194	2,359	2,1232E-	1,5911E-	31,64	0,16298	0,0313
60	4,2E14	1549,3	0,020968	3,5203	4,1966E-	2,3851E-	47,083	0,37237	0,0530
80	4,2E14	1532,8	0,020745	4,6691	6,6526E-	3,1741E-	62,267	0,66794	0,0748
100	4,2E14	1516,5	0,020523	5,8053	9,362E-5	3,9571E-	77,195	1,0469	0,0963
120	4,2E14	1500,3	0,020304	6,9292	0,000122	4,7332E-	91,871	1,5057	0,1174
140	4,2E14	1484,3	0,020088	8,0408	0,000151	5,5019E-	106,3	2,041	0,1382
160	4,2E14	1468,4	0,019873	9,1403	0,000181	6,2629E-	120,48	2,6495	0,1586
180	4,2E14	1452,8	0,019661	10,228	0,000211	7,016E-6	134,42	3,328	0,1787
200	4,2E14	1437,3	0,019451	11,304	0,000241	7,7613E-	148,12	4,0736	0,1984
220	0	1421,9	0,019244	12,368	0,000271	8,4986E-	161,58	4,8831	0,2178
230	0	1421,9	7,1777E-	12,369	0,000221	2,0092E-	161,6	4,9985	0,1025

Fig. 3.7. "Calc" tab of ChainSolver program.

4.3.5. "Answers" tab

The "Answers" page, a page of answers, presents the results of the calculations performed as shown in figure 3.8. The check box "A_Z" allows the alphabetical classification of the nuclide names. The unit of the activities obtained can be chosen in the box "Activity Units": (Ci, Bq). Type of measurements is determined from the choice "What" [6, 26, 36, 79].

"Nuclei": number of nuclei of the chain isotopes for each time interval.

"Mass": weights (in grams).

"Activity A": alpha activity.

"Activity B": beta activity.

"Activity EC": disintegration activity by electronic capture.

"Activity IT": decay activity by isomeric transition.

"Activity Total": total activity.

"Activity per element g": total activity for one gram of the element mass.

"Isotope parts": isotopic partition of the elements of the chain.

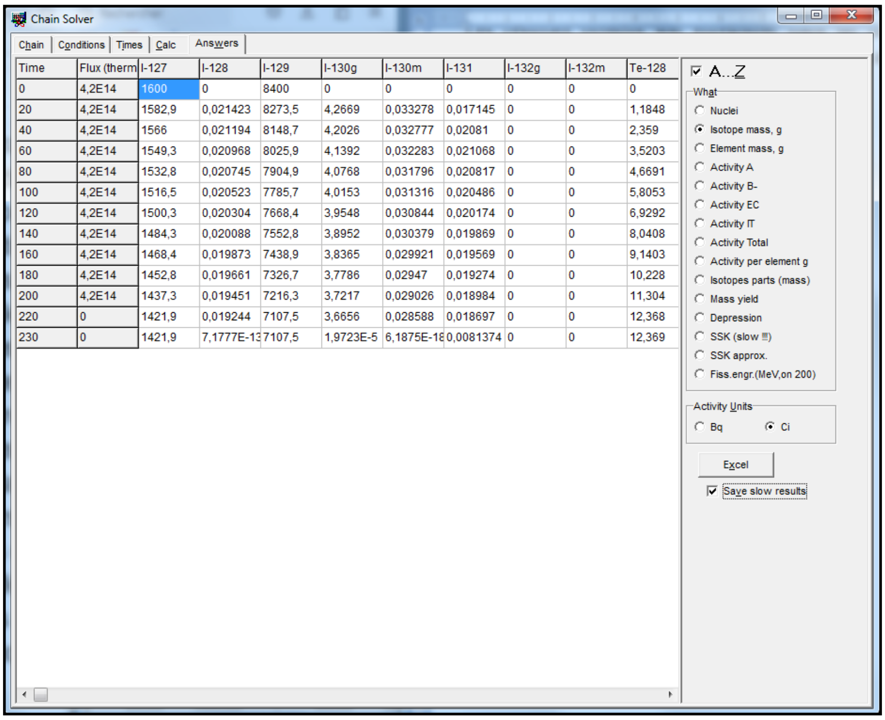
"Masse yield": yield of mass per gram of initial elements.

"Depression": factors of depression.

"SSK": self-shielding factors.

"Fiss. engr": value of the fission energy (in MeV).

The results obtained are saved in an Excel sheet type file, by selecting the "Save slow results" menu, then pressing the "Excel" button.



Time	Flux (therm	I-127	I-128	I-129	I-130g	I-130m	I-131	I-132g	I-132m	Te-128
0	4,2E14	1600	0	8400	0	0	0	0	0	0
20	4,2E14	1582,9	0,021423	8273,5	4,2669	0,033278	0,017145	0	0	1,1848
40	4,2E14	1566	0,021194	8148,7	4,2026	0,032777	0,02081	0	0	2,359
60	4,2E14	1549,3	0,020968	8025,9	4,1392	0,032283	0,021068	0	0	3,5203
80	4,2E14	1532,8	0,020745	7904,9	4,0768	0,031796	0,020817	0	0	4,6691
100	4,2E14	1516,5	0,020523	7785,7	4,0153	0,031316	0,020486	0	0	5,8053
120	4,2E14	1500,3	0,020304	7668,4	3,9548	0,030844	0,020174	0	0	6,9292
140	4,2E14	1484,3	0,020088	7552,8	3,8952	0,030379	0,019869	0	0	8,0408
160	4,2E14	1468,4	0,019873	7438,9	3,8365	0,029921	0,019569	0	0	9,1403
180	4,2E14	1452,8	0,019661	7326,7	3,7786	0,02947	0,019274	0	0	10,228
200	4,2E14	1437,3	0,019451	7216,3	3,7217	0,029026	0,018984	0	0	11,304
220	0	1421,9	0,019244	7107,5	3,6656	0,028588	0,018697	0	0	12,368
230	0	1421,9	7,1777E-13	7107,5	1,9723E-5	6,1875E-18	0,0081374	0	0	12,369

☑ A...Z

What
○ Nuclei
● Isotope mass, g
○ Element mass, g
○ Activity A
○ Activity B-
○ Activity EC
○ Activity IT
○ Activity Total
○ Activity per element g
○ Isotopes parts (mass)
○ Mass yield
○ Depression
○ SSK (slow !!!)
○ SSK approx.
○ Fiss.engr.(MeV,on 200)

Activity Units
○ Bq ● Ci

☑ Save slow results

Fig. 3.8. "Answers" tab of ChainSolver program.

Chapter IV

Transmutation Efficiency of Iodine-129

1. Definition of High Flux Reactors "HFRs"

The present study is based on different types of "High Flux Reactors" HFRs, namely Petten, BR2, SM3 and JOYO.

1.1. Petten high flux reactor

The Petten HFR (Netherlands) is a tank pool type reactor. It operates at a constant power of 45 MW using a low-enriched uranium plate type fuel, cooled and moderated with light water. The core is composed of 33 fuel rods, 6 control rods, 25 reflector rods and 17 experimental stations [36]. The neutron fluence can reach 10^{26} m^{-2} with a thermal component up to 10^{25} $n.m^{-2}$, a fast component up to 10^{25} $n.m^{-2}$ [67]. The core configuration is shown in the following Figure 4.1 [82]:

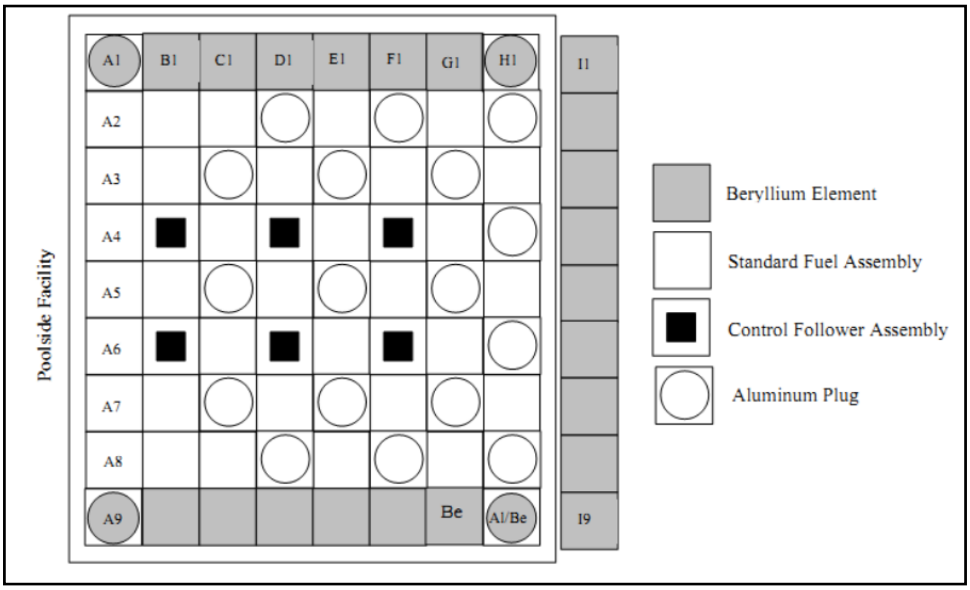


Fig. 4.1. The Petten HFR Reactor core configuration.

1.2. BR2 high flux reactor

The BR2 (Belgian Reactor 2) is a 100 MWth HFR dedicated to materials testing. Commissioned in 1991, BR2 operates with highly enriched uranium (39.6 % ^{235}U)

with water under pressure as coolant and moderator [83]. The availability of high thermal neutron fluxes up 10^{15} n.cm^{-2}.s^{-1} offers an important opportunity for routine production of radioisotopes for medical and industrial applications [84]. The following Figure 4.2 shows a cross section of BR2 HFR [83].

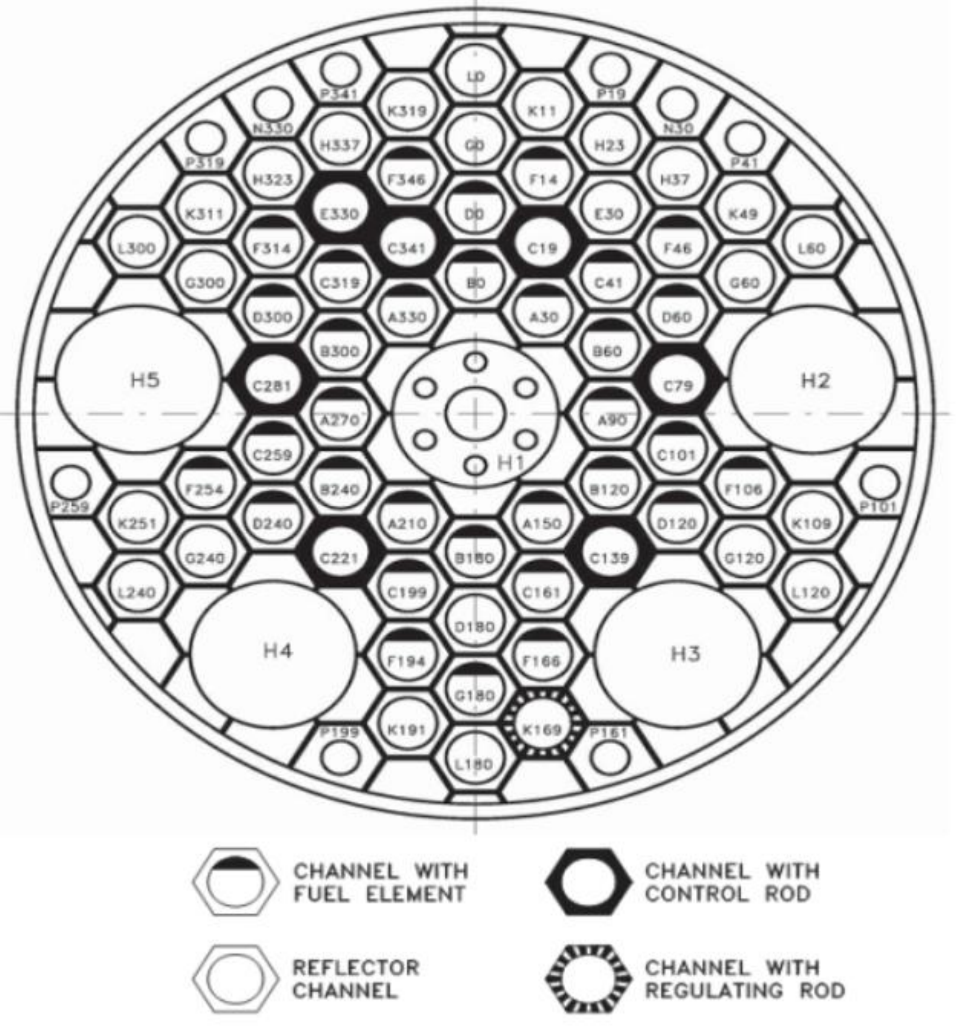


Fig. 4.2. BR2 HFR cross-section.

1.3. SM3 high flux reactor

The SM3 (Dimitrovgrad, Russian Federation) HFR is a water-cooled vessel with a trap. It uses UO_2 fuel with high enrichment. The thermal power is up to 100 MW [85]. The neutron flux density is higher than 10^{15} n.cm^{-2}.s^{-1}. A horizontal cross-section of the core geometry of the SM3 reactor is shown in the following Figure 4.3 [86]:

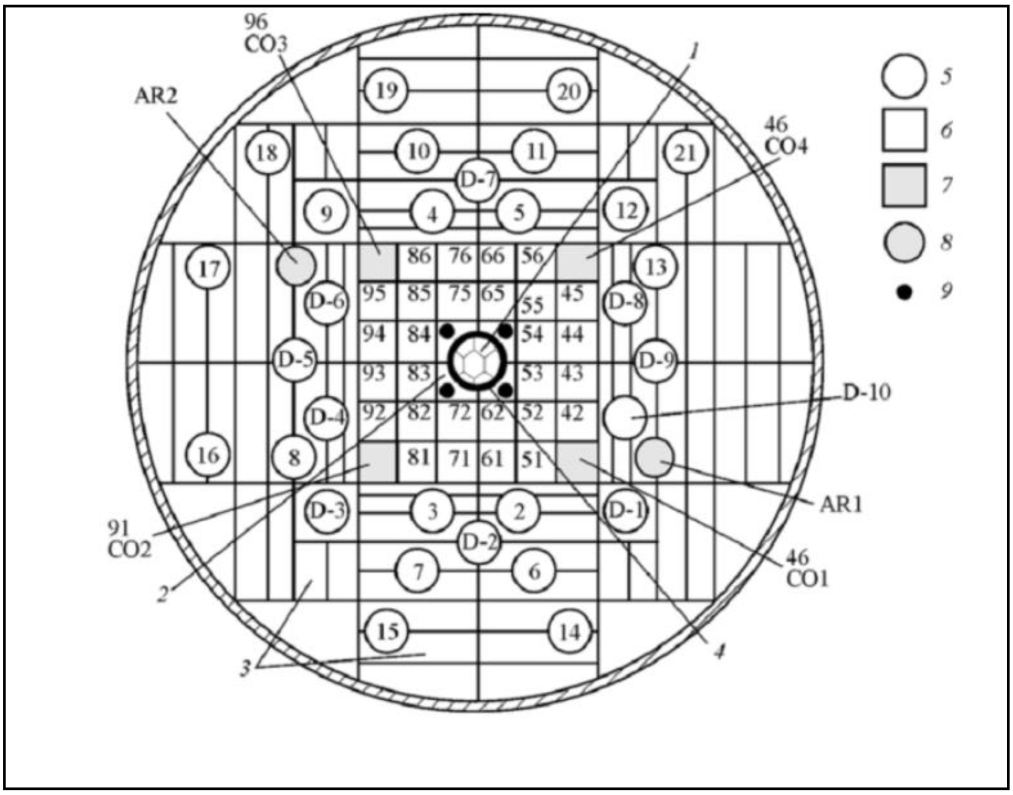


Fig. 4.3. Map of the SM3 reactor: (1) central block of transmutation targets, (2) beryllium bushes, (3) beryllium block of the reflector, (4) central compensating controller, (5) channels, (6) core sub-channels, (7) compensating controllers, (8) automatic controllers, and (9) safety rods.

1.4. JOYO fast reactor

The JOYO experimental fast reactor was built as the first step in the development of a sodium-cooled fast reactor with mixed oxide MOx fuel in Japan. JOYO reactor reached initial criticality in 1977 with the MK-I breeder core. From 1983 to 2000, JOYO operated at 100 MW for thirty-five operational cycles with the MK-II irradiation test core. In 2003, the upgrade of the JOYO reactor to the 140 MW MK-III core was completed to increase the irradiation test capability of JOYO. The maximum fast neutron flux can be increased from 3.2×10^{15} $n.cm^{-2}.s^{-1}$ to 4.0×10^{15} $n.cm^{-2}.s^{-1}$ and therefore the reactor power is increased from 100 to 140 MW. The core design is shown in Figure 4.4 [87].

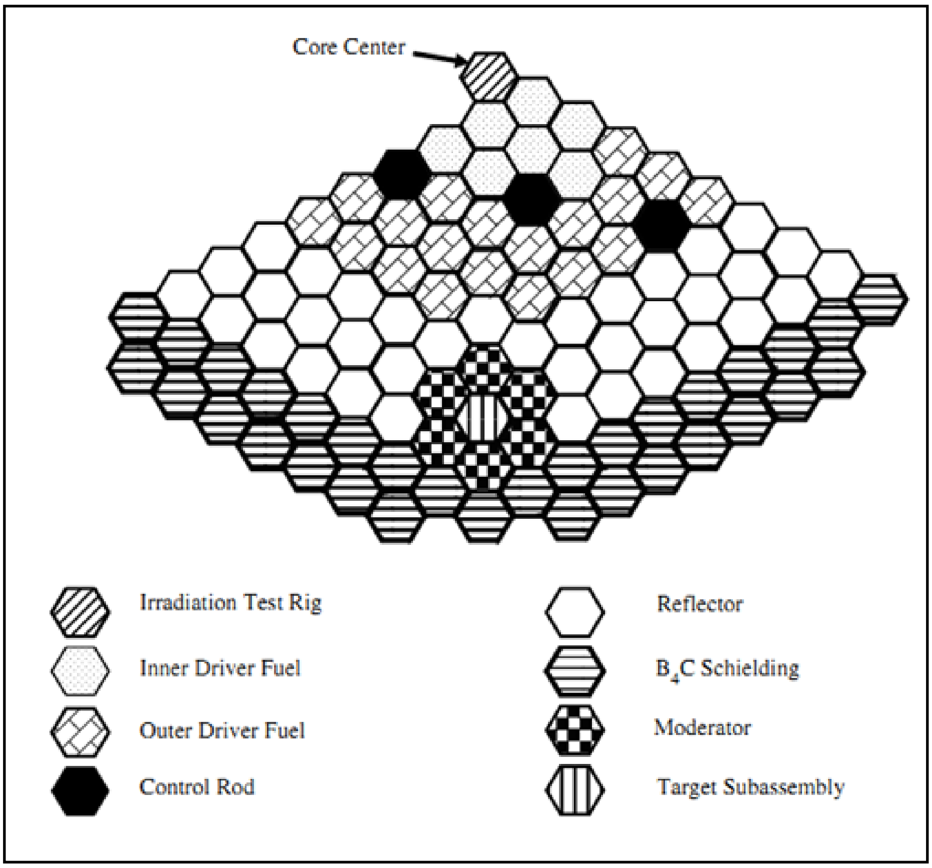


Fig. 4.4. Iodine target location in JOYO core.

2. General information on Iodine-129 and its transmutation

The accumulation of long-term radioactive waste in the spent fuel of nuclear power plants around the world represents the greatest challenge to the continuity of electricity generation from nuclear power plants. The traditional solution to dispose of this radioactive waste is geological disposal and this requires a very high cost due to engineering constraints in the construction and maintenance of such repositories [88].

Spent nuclear fuel consists of actinides and fission products. Fission products cannot fission because they are tightly bound, but they can transmute into other low radioactive nuclides through other nuclear interactions [89]. The seven long-lived fission products are considered the most hazardous waste. Nuclear incineration of LLFPs and minor actinides has been studied as an alternative means of reactor waste disposal [90]. Transmutation of LLFPs to shorter-lived or stable nuclides is an effective method of disposal [91].

2.1. Characteristics of Iodine-129

Iodine-129 is one of the most important LLFPs found in spent nuclear fuel. It contributes significantly to long-term radiological effects [67]. The most important characteristics of any radioactive element are half-life ($T_{1/2}$) and mobility in the environment rather than quantity. Because of its relatively long half-life of 1.57×10^{7} years combined with high mobility in the environment and uptake by human bodies, iodine-129 is highly suitable considered for the fission product transmutation option. Iodine-129 is produced by nuclear reactors with a cumulative yield of 0.75 % and 1.40 %, for the thermal neutron fission of uranium-235 and plutonium-239 respectively [66]. The production of iodine-129 is 1.6 to 4.6 kg/GWe.y, for a 33 GWt.d burnup spent fuel after 3 years of cooling [92]. It's radiotoxicity is about 0.2696 Sv/g in a PWR. For the half-life of iodine-129 is about 63 y in fast neutron and 70 y in thermal neutron. These calculations have been calculated under thermal flux of 1×10^{14} $n.cm^{-2}.s^{-1}$ and fast flux of 1×10^{15} $n.cm^{-2}.s^{-1}$ [62]. Thermal power of iodine-129 is 1.3×10^{-14} W/Bq, its ingestion dose is 7.4×10^{-8} Sv/Bq and its fraction in an irradiated fuel is 170 g/t [93].

2.2. Principle and scheme of the experimental transmutation of iodine-129

Nuclear transmutation can be induced by any particle capable of penetrating nuclei and interacting with nucleons. Transmutation by neutron occurs in nuclear reactors with a flux range 10^{14}-10^{15} $n.cm^{-2}.s^{-1}$. For iodine-129, the neutron capture cross section is 0.35 b for fast neutrons and 3.12 b for thermal neutrons, making it transmutable in nuclear reactors [81]. Transmutation of iodine-129 can be accomplished by a single neutron capture, followed by a fast β^- decay to stable isotope xenon-130. Additional neutron capture are possible and can yield other stable isotopes Xe-131 and Xe-132 [66]. The techniques are based on either the (n, γ) or the (γ, n) capture reaction. The nuclides produced are all short-lived. However, the efficiency of transmutation methods further depends on the level of (1) particle flux and (2) the effective cross sections involved [62].

$$^{129}I + n \rightarrow {}^{130}I \rightarrow {}^{130}Xe + \beta^{-}$$

For the transmutation scheme of iodine-129 and iodine-127 and the formation of xenon-130 and xenon-128 under neutron irradiation are presented respectively in Figure 4.5 and Figure 4.6.

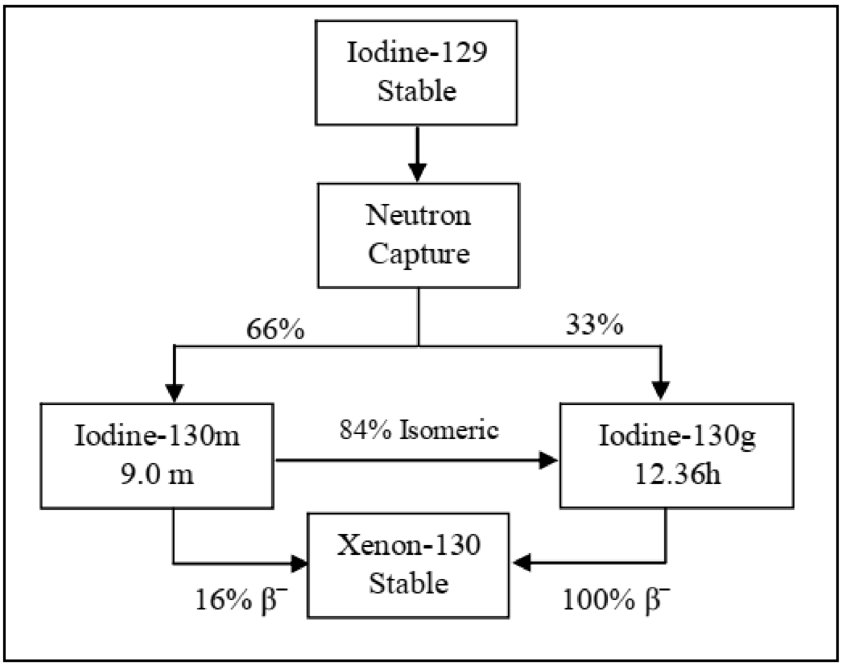


Fig. 4.5. Transmutation scheme of I-129.

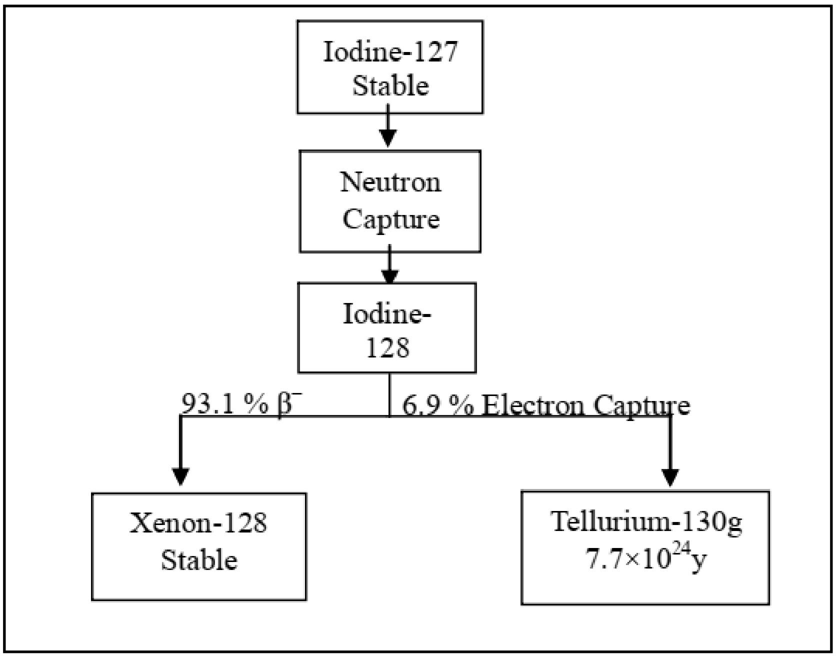


Fig. 4.6. Transmutation scheme of I-127.

For the first scheme (Fig 4.5), the ground state of iodine I-130g (half-life of 12.36 h) decays to the stable isotope Xe-130. For the second scheme (Fig 4.6), 94 % of the I-128 nuclides decay to the xenon xe-128. The transmutation schemes ^{127}I and ^{129}I are quite similar, but some differences can be identified, the resonance integral of ^{127}I is five times larger than that of ^{129}I, in contrast to the thermal cross-section, which is five times smaller. The effective cross-sections of ^{127}I and ^{129}I as a function of neutron energy are shown in Figure 4.7 [94]. It can be seen that ^{129}I has a weak thermal cross-section and higher resonance absorption peaks in the epithermal and fast energy range.

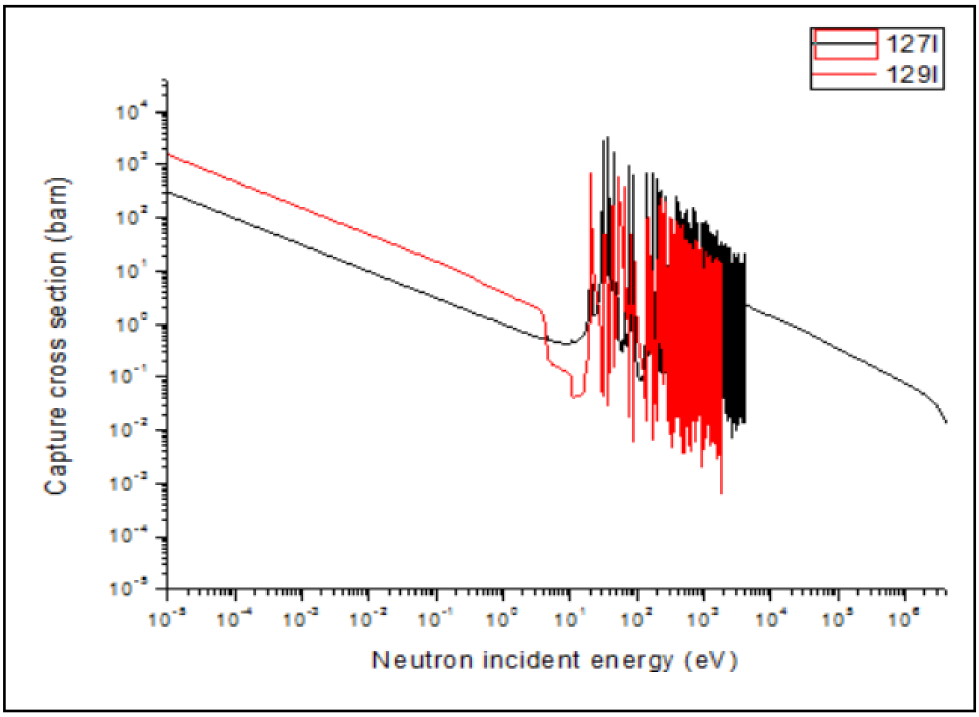


Fig. 4.7. Cross-sections (n, γ) for I-127 and I-129 from ENDF data.

2.3. Isotopic composition of iodine in spent fuel

Within the framework of the OMEGA (Options, Making Extra Gains from Actinides) program in Japan [95], the Japan Atomic Energy Research Institute (JAERI) has initiated a literature review and experimental studies concerning the transmutation of iodine-129 [70]. The isotopic composition of iodine in the LWR spent fuel is 16 % for ^{127}I and 84 % for ^{129}I [94]. For the PWR spectrum, the isotopic composition of ^{127}I and ^{129}I are 22.98 % and 77.02 % respectively [62]. Transmutation of ^{129}I to ^{130}Xe presents some challenges; it is theoretically possible, but at a low rate, in addition to irradiation targets with sufficient masses (1-2 g) [67]. In the case of iodine transmutation experiments, metal iodide compounds were irradiated in the Petten high flux reactor [67].

2.4. Studies in the literature on the transmutation of iodine-129

Some of the performed ^{129}I transmutation experiments have been described in the literature. In the Fast Flux Test Facility (FFTF), a milligram-sized physical sample of ^{129}I was irradiated [70]. One could also mention the experimental feasibility of the transmutation target (EFTTRA-T1) performed by Konings [67], where metal iodides were irradiated in the Petten HFR as target materials [94]. Code calculations on iodine transmutation in the BR2 HFR and the JOYO experimental fast reactor [69] were performed to optimize the best conditions (neutron flux, moderating materials and reflecting materials) in order to achieve a high transmutation rate. The properties of iodine compounds were compared in order to select the best target materials for ^{129}I transmutation. According to the reactor heating experiments, the candidate target materials composed with suitable chemical forms were NaI, MgI_2, CaI_2, CuI et $Ca(IO_3)_2$ [66].

In the BR2 high-flux reactor, Na, Ca and Ba iodides were considered and CaI_2 was chosen as the reference compound [71]. About 400 g of iodine can be transmuted per year [96]. For the iodine separated from the dissolver off-gas in the reference fuel cycle, it can be transformed into a stable $Ba(IO_3)_2$ matrix that can be used as an irradiation matrix to destroy the iodine [97]. Code-based Iodine transmutation studies have been performed with the MCNP/MCNPX [98], ORIGEN [99], CITATION [100], MVP [101] and TRIFON [102].

2.5. Transmutation chain created by the ORIP_XXI collection

For irradiation conditions, the transmutation scheme for ^{129}I and ^{127}I can be greatly simplified, because for nuclides with competing transition channels, due to the low neutron capture cross-sections, β decay occurs with overwhelming probability. The radiative capture cross sections of thermal neutrons by ^{129}I and ^{127}I are 27 and 6,2 barns, respectively; their resonance systems range from 50 eV to 4 keV and yield integrals of 36 and 147 barns. The burning rate of these isotopes is the same with the hardness of the spectrum [103].

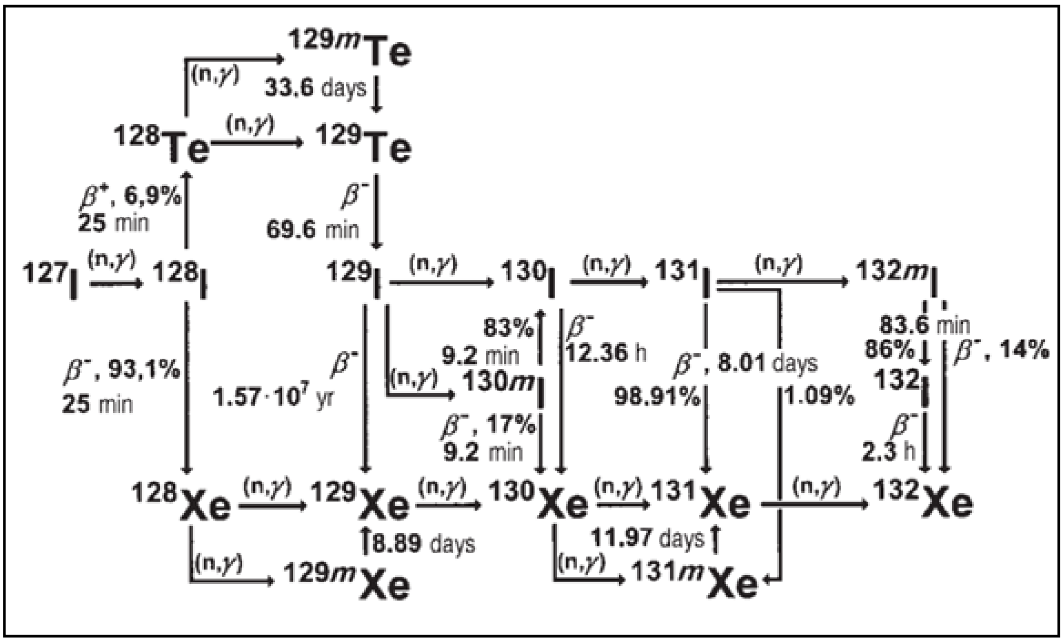


Fig. 4.8. Transmutation scheme for both ^{129}I and ^{127}I in the literature.

The ChainSolver 2.34 code is widely used to perform simulations of transmutation of samples during irradiation in nuclear reactors. This code calculates a temporal evolution of the nuclide density by including nuclear reaction, decay and accumulation. These calculations are extremely tedious because at each step of an irradiation duration, it is necessary to take into account the effective cross-sections of fast, epithermal and thermal flux of the reactions and the time of an irradiation. This code uses information from the complex data file of the ORIP_XXI program and is based on the solution of the Bateman equation [79]. The iodine transmutation chain created by the ChainSolver 2.34 code is shown in Figure 4.9.

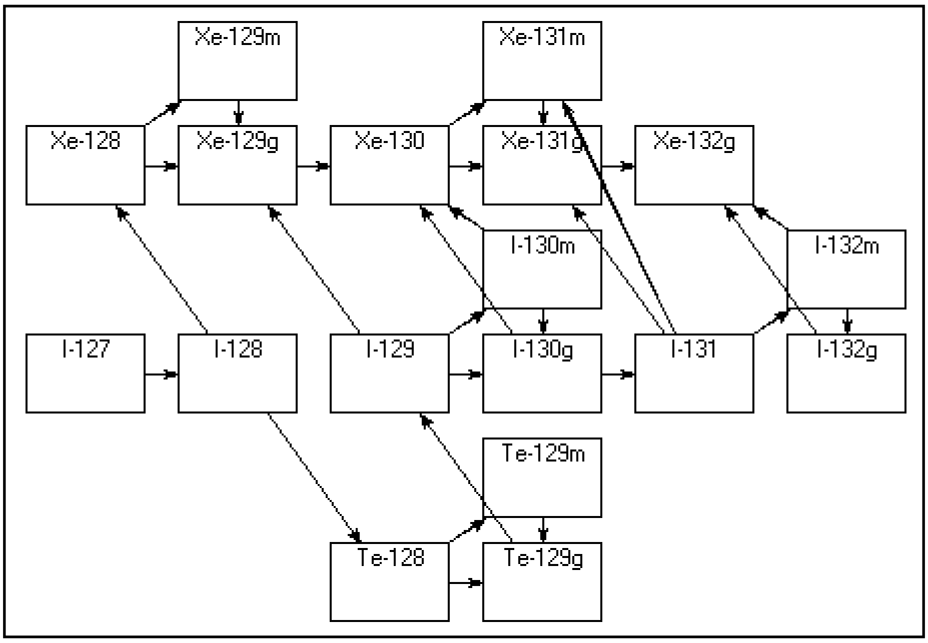


Fig. 4.9. Transmutation Chain of both ^{129}I and ^{127}I , created with ChainSolver code.

3. Efficiency of transmutation

3.1. Parameters influencing transmutation

The transmutation rate of LLFPs by neutron capture which corresponds to the ratio of its transmutation mass to the initial mass can be defined by the following formula [104] :

$$Transmutation\ Rate = 1 - \exp(-\sigma_{n,\gamma}.\Phi.t) \text{ ....(Eq. 4.1)}$$

Where "$\sigma_{n,\gamma}$" is the LLFP neutron capture cross-section in cm^2, "Φ" is the average neutron flux inside the iodide pin in 'n.cm^{-2}.s^{-1}' and 't' is the irradiation time in 's'. If we increase any of the three $\sigma_{n,\gamma}$, Φ, t; the transmutation rate will increase. The transmutation rate as a function of time is an increasing exponential function.

3.2. Calculated parameters of transmutation

Several calculated parameters have been cited in several previously published papers, we cite the transmutation rate TR or R in % [105], transmutation amount C in kg/GWt.y which is the same as g/MWt.y [98], transmutation support ratio TSR in y^{-1} [106], average transmutation acceleration A in atoms.s^{-1} [107], and effective transmutation half-life in years [62, 108].

These parameters are defined as follows:

$$R(t) = 100 \times \frac{m_0 - m_t}{m_0} \quad \text{(Eq. 4.2)}$$

$$C(t) = \frac{m_0 - m_t}{P \times t_{irr}} \quad \text{(Eq. 4.3)}$$

$$TSR(t) = \frac{m_0 - m_t}{m_0 \times t_{irr}} \quad \text{(Eq. 4.4)}$$

$$A(t) = 5{,}40 \times 10^{16} \times \frac{m_0 - m_t}{t_{irr}} \quad \text{(Eq. 4.5)}$$

$$T_{1/2}^{tr} = \frac{t_{irr} \times ln2}{\ln[\frac{N_i(0)}{N_i(T)}]} \quad \text{(Eq. 4.6)}$$

$$T_{1/2}^{tr} = 3.171 \times 10^{-8} \frac{ln2}{\sigma \times \phi} \quad \text{(Eq. 4.7)}$$

Where m_0 is the amount of iodine-129 loaded into the core in g, m_t is the amount of iodine-129 discharged from the core in g, P is the thermal power of the core in MWt and t_{irr} is the duration of the operating cycle (years), σ is the effective neutron capture cross-section in barns, ϕ is the neutron flux in $n.cm^{-2}.s^{-1}$. The unit of $T_{1/2}$ (tr) is 'years'.

3.3. Evaluation of iodine-129 transmutation

The neutron flux and thermal power data used in these calculations for the Petten, BR2, SM3 and JOYO high flux reactors are summarized in the following table. The irradiation time t_{irr} was taken equal to 300 effective days at full power (EFPD).

Reactor HFR	Thermal power (MW_{th})	Neutron flux ($n.cm^{-2}.s^{-1}$)		
		Fast	Epithermal	Thermal
Petten (Netherlands)	45	1.36×10^{14}	1.0×10^{14}	3.75×10^{14}
BR2 (Belgium)	100	4.20×10^{14}	4.00×10^{13}	6.00×10^{14}
SM3 (Russia)	Up 100	8.80×10^{14}	5.10×10^{14}	5.40×10^{14}
JOYO* (Japan)	140	9.95×10^{14}	4.84×10^{14}	6.34×10^{13}

* *Moderating material considered is beryllium (90 %).*

Tab. 4.1. Neutron fluxes and thermal power data of the core.

With these values of neutron flux, thermal power, and irradiation time, the iodine-129 transmutation rate R, transmutation amount C, transmutation support ration TSR, average transmutation acceleration A and transmutation half-life $T_{1/2}$(tr) are calculated with the ChainSolver 2.34 code.

The total mass of iodine of 12 g (^{129}I presents 10.08 g which is 84 %) is considered in the four reactors studied for the present work; Petten, BR2, SM3 and JOYO with the corresponding data of fluxes, core thermal power and irradiation time to perform the calculations of (R, C, TSR and A). The total mass produced of xenon m_{Xe} and the mass of remaining iodine m_I as a function of time are calculated with the ChainSolver 2.34 code. The resulting mass is used to calculate R, C, TSR and A. In the present work, we are interested in the calculations of iodine-129 because iodine-127 is a stable nuclide.

The results of I-129 transmutation rate R, transmutation amount C, amount of Xe-130 produced masse m_{Xe}, amount of I-129 remaining mass m_I, transmutation support ratio TSR and average transmutation acceleration A over time are shown in Figure 4.10, Figure 4.11, Figure 4.12, Figure 4.13 and Figure 4.14 for the four reactors considered in this study.

At the end of the 300 EFPD, the final values reached for parameters already calculated (m_{Xe}, m_I, R, C, TSR and A) are presented in Table 4.2, Table 4.3 and Table 4.4.

HFR Reactor	Xe-130 mass(g)	I-129 mass (g)	R (%)
Petten	0.78	9.27	08.05
BR2	2.26	7.50	25.58
SM3	4.58	3.65	63.77
JOYO	3.69	6.16	38.88

Tab. 4.2. Xe-130 produced mass, I-129 remaining mass, I-129 transmutation rate.

HFR Reactor	C (kg/GWt.y))	TSR (y^{-1})	A (atoms/s)
Petten	01.8×10^{-2}	0.080	1.46×10^{14}
BR2	02.6×10^{-2}	0.256	4.64×10^{14}
SM3	06.4×10^{-2}	0.638	1.16×10^{15}
JOYO	02.8×10^{-2}	0.389	7.06×10^{14}

Tab. 4.3. Transmutation amount, transmutation support ratio, average transmutation acceleration.

3.3.1. Evaluation of transmutation rate R

From the code calculations and using equation 4.2, the transmutation rate R can be calculated.

The obtained transmutation rates in Petten, BR2, SM3 HFRs and Joyo fast reactor for 300 EFPD are 08.05 %, 25.58%, 63.77 % and 38.88 % respectively.

Higher transmutation rate R is registered in SM3 reactor (Figure 4.10), with a value eight times higher than that in Petten reactor, and two and a half times higher than that in BR2 reactor. Table 4.2 presents the produced mass of Xe-130, remaining mass of I-129, transmutation rate after 300 EFPD.

The transmutation rate R does not reflect xenon-130 mass compared to the initial loaded mass of iodine-129, because additional isotopes of iodine (I-127, I-128, I-130g, I-130m, I-131, I-132g, I-132m), xenon (Xe-128, Xe-129g, Xe-129m, Xe-131g, Xe-131m, Xe-132g) and tellurium (Te-128, Te-129m, Te-129g); 'g' means ground state and 'm' means metastable. These isotopes are shown in Figure 4.9.

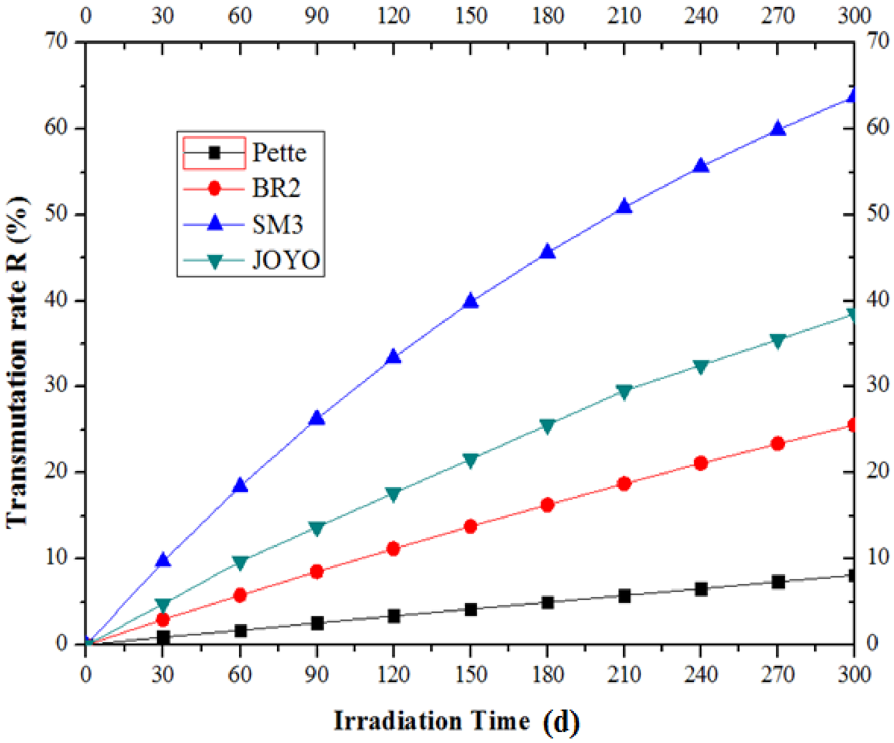


Fig. 4.10. Temporal Iodine-129 Transmutation Rate in HFR.

3.3.2. Evaluation of ^{130}Xe mass produced and ^{129}I mass remaining

The mass of Xe-130 produced during irradiation (4.58 g) is higher in SM3 reactor than in JOYO (3.69 g), BR2 (2.26 g) and Petten (0.78 g) (Figure 4.11). This means that the irradiation time (300 EFPD) required to obtain 0.78 g of Xe-130 in Petten are equivalent to 87 EFPD in BR2 and to 26 EFPD in SM3 reactor, respectively. In the same way, 300 EFPD in BR2 reactor are equivalent to 85 EFPD in SM3 reactor, which almost the third irradiation time.

For the remaining I-129 mass after 300 EFPD for each reactor. Highlights the lowest remaining mass in SM3 (3.65 g) compared to Petten (9.27 g), BR2 (7.5 g), and JOYO (6.16 g) [Fig 4. 12].

This means that SM3 reactor achieves the highest xe-130 production, lowest remaining I-129 mass, and demonstrates the most efficient transmutation process based on the analyzed metrics.

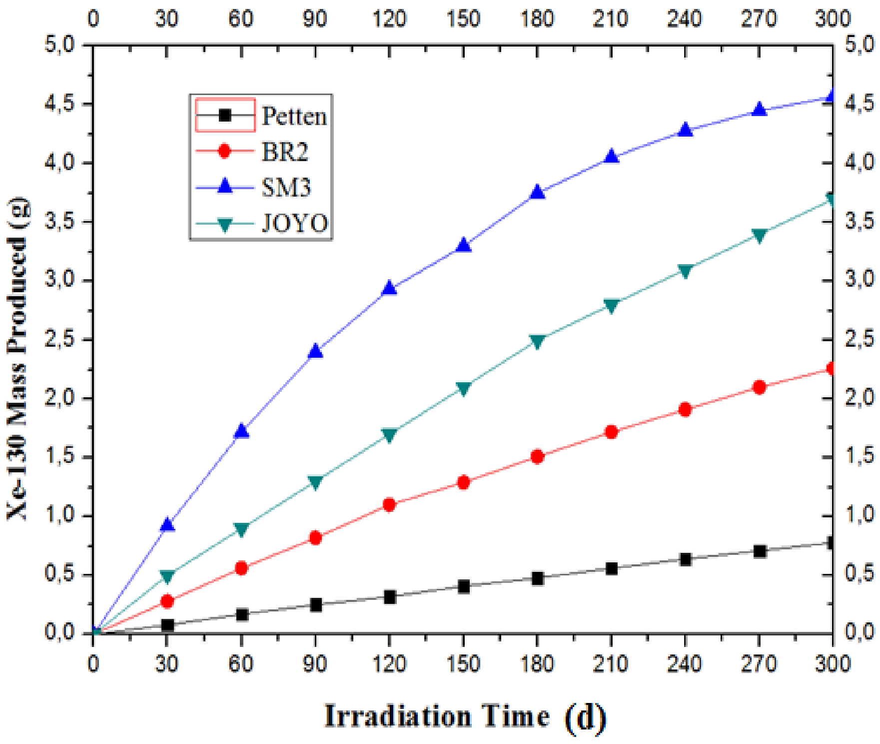


Fig. 4.11. Variation of the Xenon-130 concentration in HFRs as a function of EFPD.

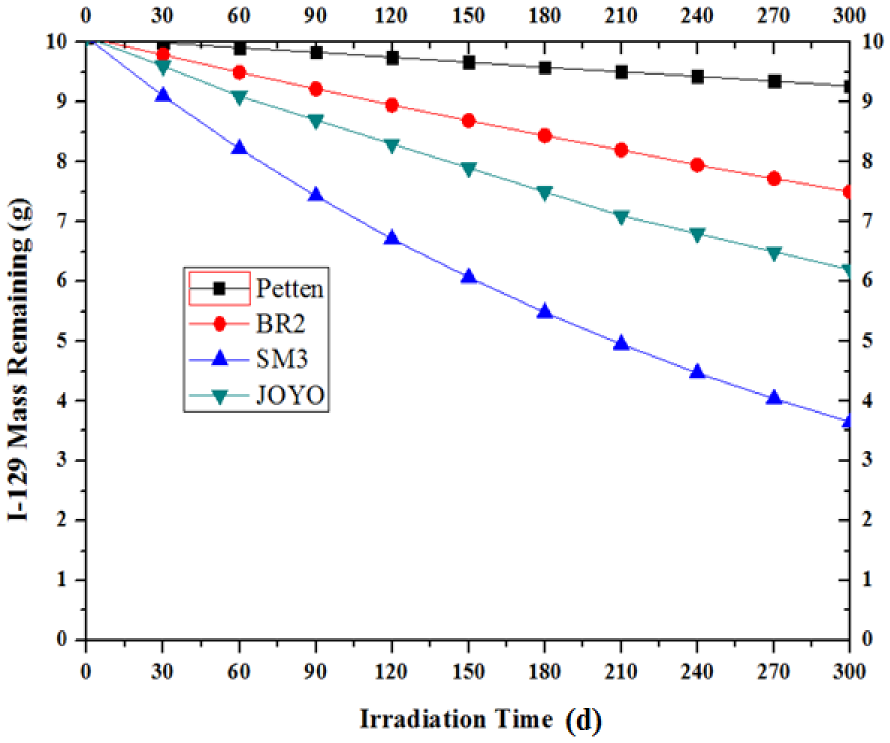


Fig. 4.12. Variation of the Iodine-129 concentration in HFRs as a function of EFPD.

3.3.3. Evaluation of transmutation amount C

From the code calculations and using equation 4.3, the transmutation amount C can be calculated.

At the end of the 300 EFPD, transmutation amounts in Petten, BR2, SM3 and JOYO are 1.8×10^{-2} kg/GWt.y, 2.6×10^{-2} kg/GWt.y, 6.4×10^{-2} kg/GWt.y, 2.8×10^{-2} kg/GWt.y respectively. The SM3 presents the highest value (Table 4.3 and Figure 4.13).

At the end of the 300 EFPD, the transmutation amount value in SM3 reactor is two and a half times greater than that of BR2 (HFR), and three and a half times greater than that of Petten (HFR). The final transmutation amount value after 300 EFPD in each reactor is presented in Table 4.3.

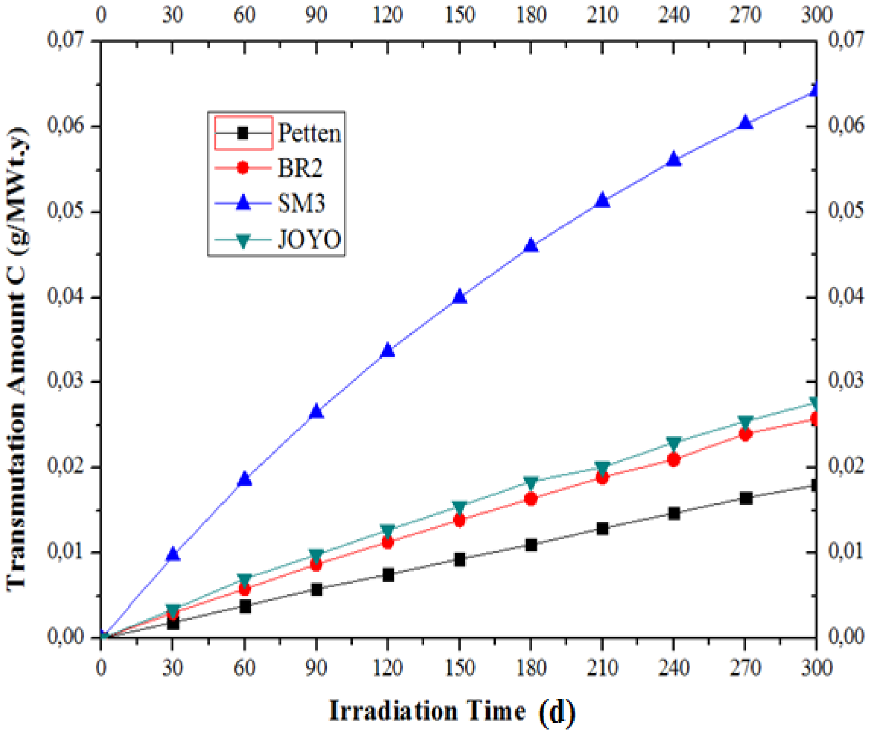


Fig. 4.13. Iodine-129 transmutation amount in HFRs as a function of EFPD.

3.3.4. Evaluation of transmutation support ratio TSR

From the code calculations and using equation 4.4, the transmutation support ratio TSR can be calculated.

At the end of the 300 EFPD, the transmutation support ratio in Petten, BR2, SM3 and JOYO are 0.08 y^{-1}, 0.256 y^{-1}, 0.638 y^{-1}, 0.389 y^{-1} respectively. The SM3 presents the highest value (Table 4.3 and Figure 4.14).

The SM3 reactor demonstrates a clear superiority in TSR compared to its peers. The SM3 reactor stands out with a TSR significantly higher than other reactors studied, suggesting an exceptional transmutation support capability.

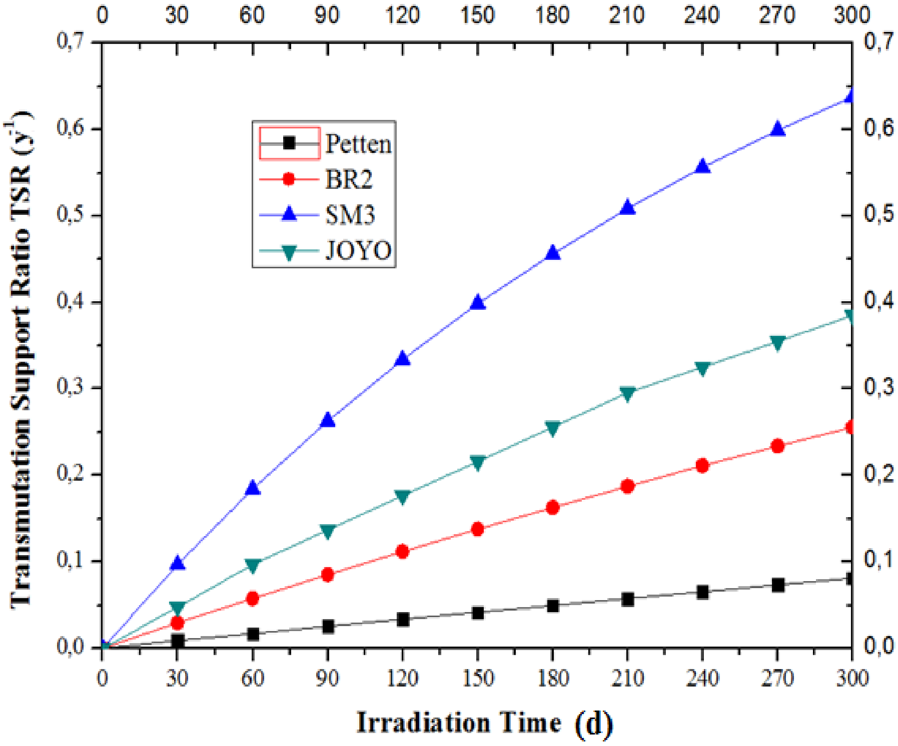


Fig. 4.14. Iodine-129 transmutation support ratio in HFRs as a function of EFPD.

3.3.5. Evaluation of average transmutation acceleration A

From the code calculations and using equation 4.5, the average transmutation acceleration A can be calculated.

At the end of the 300 EFPD, the average transmutation acceleration in Petten, BR2, SM3 and JOYO are 1.46×10^{14} transmuted atoms/s, 4.64×10^{14} transmuted atoms/s, 1.16×10^{15} transmuted atoms/s, 7.06×10^{14} transmuted atoms/s, respectively. (Table 4.3 and Figure 4.15). This indicates that SM3 reactor demonstrated the highest capacity to accelerate the transmutation process compared to the other reactors. The results clearly indicate that the SM3 reactor presents a notably superior performance, with an acceleration almost 3 times higher than that of the BR2 reactor, the second best performer.

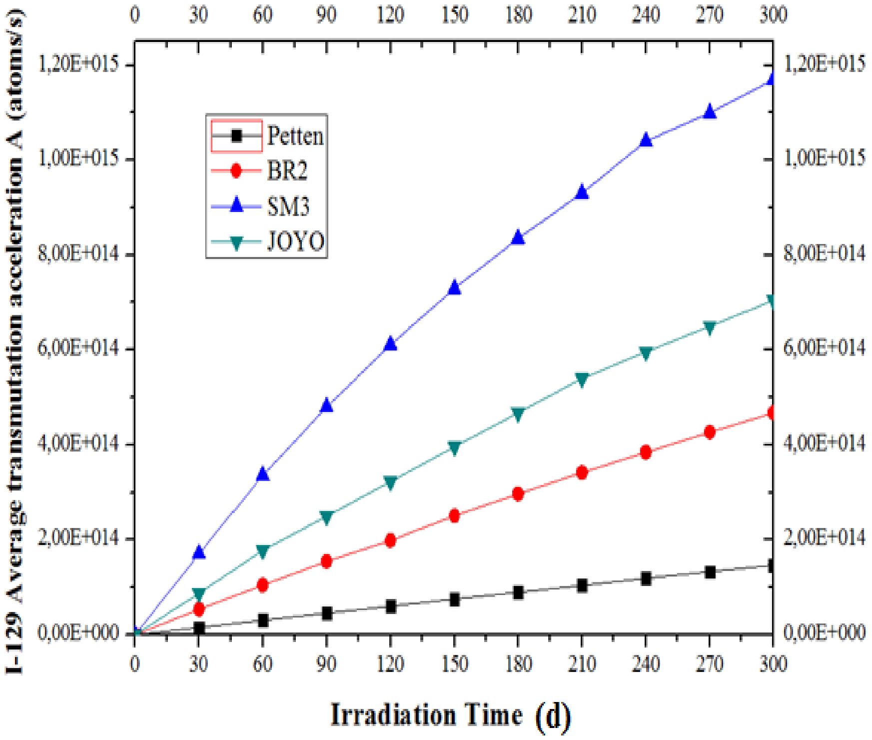


Fig. 4.15. Iodine-129 average transmutation acceleration in HFRs in time.

3.3.6. Evaluation of transmutation half-life $T_{1/2}$ (tr)

After calculating the final masses at the end of irradiation using ChainSolver 2.34 code, as already quoted in the table 4.2, using the neutron fluxes and irradiation time values already indicated in table 4.1 and using equation 4.6; the transmutation half-life can be calculated in the four reactors; Petten, BR2, SM3 and JOYO (Table 4.4).

	Petten	BR2	SM3	JOYO
$T_{1/2}$(tr) [years]	8.685	2.376	0.685	1.418

Tab. 4.4. Transmutation half-life in the four reactors under study.

As the SM3 is the best reactor for its lower transmutation half-life and its high thermal neutron flux; if the neutron flux range between 10^{14} n.cm^{-2}.s^{-1} to 10^{16} n.cm^{-2}.s^{-1}, the transmutation half-life $T_{1/2}$ (tr) can be evaluated using equation 4.7 (Fig 4.16).

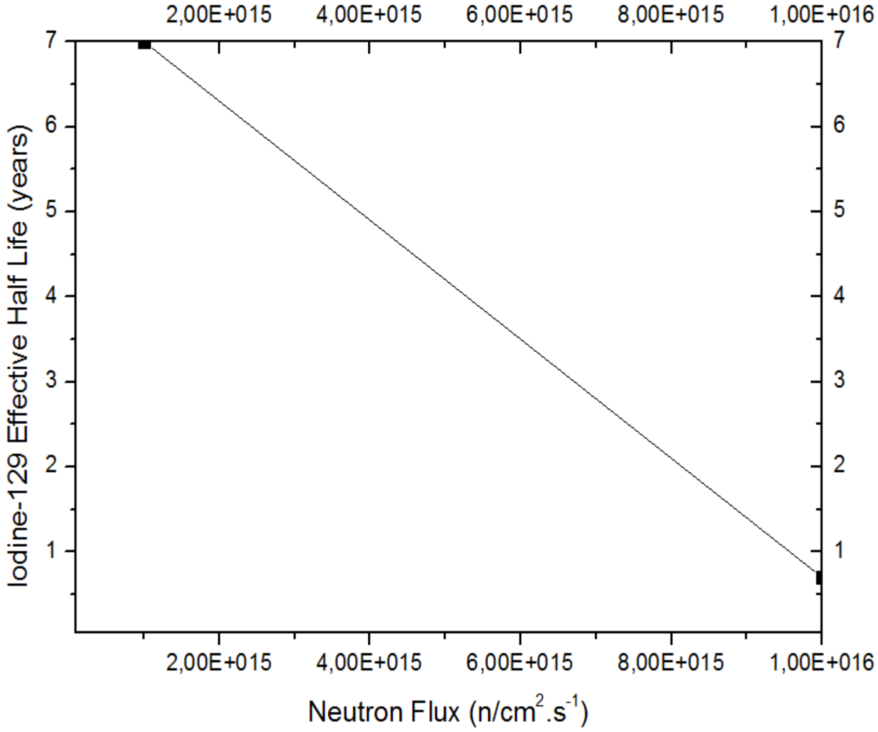


Fig. 4.16. Effective half-lives and neutron flux for Iodine-129.

The higher neutron flux values, the greater transmutation half-lives. If the of neutron flux is of order 10^{16} n.cm^{-2}.s^{-1}, the transmutation half-life will be of 0.7 year. The 10-fold increase in neutron flux, introduces a 10-fold change in transmutation half-life.

4. Conclusion

Based on the evaluation of iodine-129 transmutation rate R, we conclude that the SM3 reactor offers a better rate due to its high epithermal and fast flux than BR2 or Petten reactors. To accelerate the transmutation of iodine according to irradiation time in HFRs, the thermal and epithermal neutron fluxes must be as high as possible, this based on the neutron cross-section of Iodine-129 in the epithermal and fast region.

Conclusion

Transmutation rates (R), transmutation amounts (C), transmutation support ratio (TSR) and average transmutation acceleration (A) of iodine-129, with the corresponding xenon-130 produced masses and iodine-129 remaining masses, and transmutation half-life in the four reactors : Petten, BR2, JOYO and SM3 were numerically evaluated using the ChainSolver 2.34 code. Iodine-129 targets were irradiated for 300 days at full reactor power.

The transmutation rates in Petten, BR2, JOYO, and SM3 reactors were 08.05%, 25.58%, 33.88 and 63.77%, respectively. The transmutation amounts in Petten, BR2, JOYO, and SM3 reactors were 01.8×10^{-2} g/MWt.y, 02.6×10^{-2} g/MWt.y, 02.8×10^{-2} g/MWt.y and 06.4×10^{-2} g/MWt.y, respectively. The transmutation support ratios in Petten, BR2, JOYO, and SM3 reactors were 0.080 y^{-1}, 0.256 y^{-1}, 0.389 y^{-1} and 0.638 y^{-1}, respectively. The average transmutation acceleration in Petten, BR2, JOYO, and SM3 reactors were 1.46×10^{14} (atoms/s), 4.64×10^{14} (atoms/s), 7.06×10^{14} (atoms/s) and 1.16×10^{15} (atoms/s), respectively.

The SM3 reactor seems to be the best HFR that offers the highest value of Iodine-129 transmutation rate, transmutation amount, transmutation support ratio and average transmutation acceleration during the irradiation time, because of its higher thermal and epithermal neutron fluxes. To enhance the transmutation rate of iodine through irradiation in HFRs, thermal and epithermal neutrons must be highest as possible.

Based on this evaluation, we propose a new transmutation process based on multi-recycle transmutation process of iodine-129 in epithermal and fast spectrum. Further studies should examine the impact of possible moderator compositions and fuel configurations to optimize the neutron flux quality for more efficient transmutation purposes.

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References

[1] L'atome - Livret thématique, CEA, 2009.

[2] Guillaume Lumin et Franck Stevens; Les particules élémentaires; France 2014.

[3] Energies, Le nucléaire, quel intérêt pour la planète ? 85 questions à HENRI SAFA, Paris, 2008.

[4] La radioactivité – livret thématique CEA, 2010.

[5] Caroline Prouillac; Synthèse et évaluation de nouveaux composés organiques et phosphorés contre les effets des rayonnements ionisants. Etude de leur mécanisme d'action in vitro, OAI, thèse e doctorat, Université Paul Sabatier de Toulouse III, 2007.

[6] N. Amrani, "Destruction (transmutation) des produits de fissions a vies longues dans les réacteurs thermique : application Technétium-99", thèse de doctorat en science, université de Sétif, 2010.

[7] Systèmes nucléaires du futur : génération IV, clefs N° 55, CEA, été 2007.

[8] Energie nucléaire, Jean-Louis Basdevant, James Rich et Michel Spiro, les éditions de l'école polytechnique, 2002.

[9] The history of nuclear energy, U.S department of energy, office of nuclear energy, Science and technology, 1994.

[10] Le fonctionnement d'un réacteur nucléaire, livret thématique, CEA, 2002.

[11] Le nucléaire expliqué par des physiciens, EDP sciences, 2002.

[12] Paul Reuss, l'épopée de l'énergie nucléaire une histoire scientifique et industrielle, EDP sciences, février 2007.

[13] Le fonctionnement d'un réacteur nucléaire – livret thématique CEA, 2002.

[14] Claude Le Sech, Christian Ngô, nuclear physics; "from quarks to applications", *DUNOD*, 2020.

[15] https://www.irsn.fr/FR/connaissances/Mediatheque/Pages/ListPicture.aspx

[16] Jacques Bouchard, Jean-Paul Deffain, Alain Gouchet, Introduction au génie atomique, MINES PARIS, les presses Paris-Tech, mai 2007.

[17] Gilbert NAUDET et Paul REUSS, « énergie, électricité et nucléaire », EDP sciences, collection génie atomique, septembre 2008.

[18] Le cycle du combustible nucléaire – livret thématique CEA, 2002.

[19] LOUIS PATARIN, le cycle du combustible nucléaire, EDP sciences, janvier 2002.

[20] X. Doligez, Influence du retraitement physico-chimique du sel combustible sur le comportement du MSFR et sur le dimensionnement de son unité de retraitement, thèse, Institut polytechnique de Grenoble, 2010.

[21] CLEF du CEA – N ˚46 – Printemps 2002.

[22] R. GUILLAUMONT, Déchets radioactifs – Gestion. Technique de l'ingénieur. BN 3660, 2010.

[23] Déchet de haute activité à vie longue; Article de Wikipedia, l'encyclopédie libre; le 16 février 2023.

[24] Les déchets radioactifs – livret thématique CEA, 2005.

[25] Olivier DERUELLE, "Mise au point d'un système de spectroscopie pour mesurer des sections efficaces neutroniques applicables à un possible développement du nucléaire comme source d'énergie", thèse de doctorat en sciences, université de Paris XI, 2002.

[26] N. Amrani; "La transmutation du déchet nucléaire technétium-99", thèse de magister en physique, université de Sétif, Algérie; 2003-2004.

[27] Perspectives de l'énergie nucléaires, 2008, Agence pour l'Energie Nucléaire (AEN).

[28] Hubert Doubre, les déchets de la fission nucléaire, IN2P3/CNRS et Université Paris-Sud CSNSM, 91405 Orsay Cedex, 2000.

[29] Bernard Bonin, Les déchets nucléaires : États des lieux et perspectives, EDP SCIENCES, Juillet 2011.

[30] IRSN, Institut de radioprotection et de sureté nucléaire, La gestion des déchets radioactifs : collecter, trier, traiter, conditionner, entreposer et finalement stocker pour se protéger, France, 2013.

[31] Catalogue descriptif des familles, inventaire national des matières et déchets radioactifs, 2009, ANDRA.

[32] Gibilaro Mathieu, Co-réduction électrochimique de l’aluminium et des lanthanides en milieu fluorures fondus ; application au traitement pyrochimique des effluents nucléaires, thèse doctorat, université de Toulouse III – Paul Sabatier, 2008.

[33] https://laradioactivite.com/dechets_radioactifs/perspectives_dechets_radioactifs.

[34] Panorama international des recherches sur les alternatives au stockage géologique des déchets de haute et moyenne activité à vie longue; rapport IRSN/2019-00318s.

[35] Actinide and fission product partitioning and transmutation; 6th Information Exchange Meeting Madrid, Spain, 11-13 December 2000.

[36] M. Houas; “Transmutation des actinides mineurs dans un réacteur à haut flux: application Américium-241”, thèse de doctorat en science; université de Sétif, Algérie; 2017.

[37] Thomas LEMESLE; "Etude de matrices vitreuses alumino-phosphates pour les conditionnement de l'iode radioactif", thèse de doctorat; Université de Lille1; 2013.

[38] Fabienne AUDUBERTS; "Mise au point d'une matrice apatitique pour le confinement de l'iode 129; Institut national polytechnique de Toulouse; 1995.

[39] Haller W. A,; Perkins R. W. (1967) Organic iodine-131 compounds released from a nuclear fuel chemical processing plant, Health Phys., 13, p733.

[40] Serne R.J. Relyea J.F. (1981) Status of radionuclide sorption-desorption studies performed by the WRIT Program, PNL-SA-9787.

[41] Véronique LACOSTE; "Transmutation de produits de fission à vie longue dans un grand massif de plomb"; thèse de doctorat; université Bordeaux I; 1998.

[42] Antoine COULON; "Incorporation d'iode dans des phosphates de calcium de structure apatitique"; thèse de doctorat; université de Limoges; 2014.

[43] Lionel CAMPAYO; "Incorporation du césium dans des phosphates de structure apatitique et rhabdophane. Application au conditionnement des radionucléides séparés"; thèse de doctorat; université de Limoges; 2003.

[44] Jean-Baptiste Clavel; "Etude de systèmes et scénarios électronucléaires double strate de transmutation des actinides mineurs en ADS"; thèse de doctorat; université Nantes Angers Le Mans; 2012.

[45] Anne-Lise CHABAUTY; Conditionnement de l'iode radioactif dans des verres de phosphate d'argent; thèse de doctorat; université de Lille; France, 2018.

References

[46] Lorrain B. (1992); L'iode dans le procédé. Comportement et rejets, réunion DCC-DPR.

[47] Tait J. C., Gauld I. C., Wilkin G.B (1989) Derivation of initial radionuclide inventories for the safety assessment of the disposal of used CANDU fuel, Atomic Energy of Canada Limited Report, AECL-9881.

[48] Hebel W., Cottone G. (1982) CEC research activities into the immobilization of volatile radionuclides from reprocessing, IAEA-SM 261/47, 245.

[49] Hak Z., Bancrioft (1980) Sorption of iodide on copper, Environ. Sc. Technol., 14(9), p1106.

[50] D. Jackson, S.R. Jones, M.J. Fulker, N.G.M. Coverlade; Environmental monitoring in the vicinity of Sell afield following deposition of radioactivity from the Chernobyl accident; Journal of Society for Radiological Protection; 2000.

[51] Bernard BOULLIS; Perspectives d'évolution du procédé PUREX; Revue Générale de l'Electricité S.A. 1996; pp. 39-42.

[52] C. Madic, J. Bourges, J.F. Dozol; The international conference on accelerator-driven transmutation technologies and applications; Las Vegas, Nevada (USA), 1994.

[53] G. Uchiyama, T. Asakura; International Topical Meeting; Demonstrating the practical achievements of nuclear waste management and disposal, 1996.

[54] R. Malmbeck, C. Apostolidis; Radioactive Waste Management Strategies and Issues; 5th European Commission; Luxembourg, 2000.

[55] C. Hill, D. Guillaneux; Les recherches scientifiques sur l'aval du cycle pour le 21éme siècle; CEA Marcoule France, 2012.

[56] J. M. Adnet, L. Donnet; 5th international conference on recycling, conditioning and disposal; France 1998.

[57] C. Guy, F. Audubert, J.E. Lartigue; New conditioning for separated long-lived radionuclides; Applied Physics (2002) pp.827-837.

[58] K. Konashi, B. Tsuchiya; Tenth International Conference on Emerging Nuclear Energy IENES 2000; Petten (Netherland); pp.244-250.

[59] K. Abrahams, J. L. Kloosterman; Recycling and transmutation of nuclear waste; ECN-RX-95-067; 1995.

[60] Neutron-driven nuclear transmutation by adiabatic resonance crossing TARC; Project report; Nuclear Science and Technology; European Commission; CERN; 1999.

[61] A. Zaetta, F. Varaine, Ch. Chabert; La faisabilité scientifique de la transmutation des déchets à vie longue; CEA; 2005.

[62] W. S. Tang, Y. Kim, R. N. Hill, T. A. Taiwo, and H. S. Khalil; Long Lived Fission product Transmutation studies; Nuclear Science and Engineering; 146, 291-318 (2004).

[63] S. Pillon; Synthèse des études sur les combustibles et cibles de transmutation (fabrication, conception, comportement sous irradiation et dissolution) réalisées dans le cadre de la loi Bataille; Note technique DEC/DIR 04-002; CEA; décembre 2004.

[64] Damien PRIEUR; Elaboration de combustible à base d'oxydes d'Uranium et d'Américium; modélisation thermodynamique et propriétés des matériaux; thèse de doctorat, université de Limoges; 2011.

[65] A Status Report, "Physics and Safety of transmutation Systems"; Nuclear Energy Agency; Nuclear Science; ISBN 92-64-01082-3, OECD 2006.

[66] Yoshiro Shirasu and Kazuo Minato; Selection of chemical forms of iodine for transmutation of ^{129}I; Journal of nuclear materials; Vol 320, Issues 1-2, PP.25-30, July 2003.

[67] R. J. M. Konings; Transmutation of iodine: results of the EFTTRA-T1 irradiation test; Journal of Nuclear materials 244 (1997) 16-21.

[68] R. J. M. Konings, E. H. P. Cordfunks; Irradiation tests of transmutation of the fission products technetium and iodine; CEC Joint Research Centre, Institute of advanced materials, the Netherlands, 1997.

[69] T. Aoyama, S. Maeda, Y. Maeda, and S. Suzuki; Transmutation of Technetium in the Experimental Fast Reactor "JOYO"; Journal of Nuclear and Radiochemical Sciences, Vol. 6, No. 3, pp. 279-282, 2005.

[70] D. W. Wootan and J. V. Nelson; Transmutation of Selected Fission products in a Fast Reactor; American Nuclear Society; Waste Disposal Options Conference; Washington, 1993.

[71] Baetsle, L. J., Raed, Ch. D. E. and Delbrassine, A. (1992) "Transmutation capability of the high flux reactor BR2 for minor actinides and long lived fission products; Proceedings of the Second International Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation, Argonne, USA.

[72] Baetsle, L. H, "Impact of fission product partitioning and transmutation of Np-237, I-129, and Tc-99 on waste disposal strategies"; Proceedings of the Second

International Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation, Argonne 1992, USA.

[73] Myung Chan Lee, Hee Sung Shin, Duk-Jun Koh and Boris Kochurov; Analysis of transmutation rates of long-lived radionuclides in the Korean multi-purpose research reactor; Korea Atomic Energy research institute and Institute of theoretical and experimental physics, Russia, 1994.

[74] Frédéric Varaine; Processus et faisabilité de la transmutation des déchets nucléaires à vie longue; Département d'étude des réacteurs, Service de physique des réacteurs et duc cycle, CEA Cadarache, 2006.

[75] Anne-Marie BAUDRON et all; Les méthodes de la neutronique; Département de modélisation des systèmes et structures; 2016.

[76] Leslie D. Galloway et al; Bateman Equation Adaptation for Solving and Integrating Peak Activity into EPA ELCR and Dose Models; ORNL/TM-2020/1780; Oak Ridge National Laboratory; 2020.

[77] Jerzy Cetnar; General solution of Bateman equations for nuclear transmutations; Annals of Nuclear Energy 33 (2006) 640-645; 2006.

[78] Olivier BRINGER; Mesures des sections efficaces de capture et potentiels d'incinération des actinides mineurs dans les hauts flux de neutrons: Impact sur la transmutation des déchets; thèse de doctorat; Institut National Polytechnique de Grenoble; 2007.

[79] E. G. Romanov, V. A. Tarasov, E .Z. Vahetov; ORIP_XXI Computer Programs for Isotope Transmutation Simulations; Mathematics and Computation, Supercomputing, Reactor Physics and Nuclear and Biological Applications; Palais des Papes, Avignon, France, 2005.

[80] V. Berthou, C. Degueldre, J. Magill; Transmutation characteristics in thermal and fast neutron spectra: application to americium; Journal of Nuclear Materials 320 (2003) 156-162.

[81] K. M. Feng, J.H. Huang; Transmutation of the actinide neptenium-237 with a hybrid reactor; Fusion Engineering and Design 29 (1995) 64-68.

[82] Hanan N.A, Pond R.B a,d Matos J.E (2002); "Revisiting the HFR-Petten LEU conversion study", *24th International Meeting* on Reduced Enrichment for Research and Test reactors, 3-8 November, San Carlos de Bariloche, Argentina.

[83] Ponsard. B, Srivastava. S.C, Mausner. L.F, Knapp (Russ),F.F, Garland M.A and Mizadeh S. (2009) "Production of Sn-117m in the BR2 high-flux reactor", *Journal of Applied Radiation Isotopes*, Vol.67, pp.1158-1161.

[84] Amrani N and Boucenna A, (2001); "Transmutation of the radiotoxic isotope ^{99}Tc under irradiation in the BR2 high flux reactor", *Journal of Annals of nuclear energy*; Vol 38, PP 1347-1350.

[85] Houas M, Amrani N and Boucenna A, (2016), "Evaluation of the Americium transmutation performance in high flux reactors", *Journal of Annals of nuclear energy*, Vol 97, pp 198-203.

[86] Tarasov V.A, Romanov E.G, Toprov, Yu G, Radchenko V.M, Rotmanov K.V, Lebedeva L.S, Kozar A.A and Peretrukin V.F; 2007; "Transmutation of ^{99}Tc and preparation of artificial stable ruthenium: I. Transmutation of metallic ^{99}Tc in a high-flux SM reactor"; *Journal of Radiochemistry*; Vol 49; pp 289-292.

[87] Naima Amrani and Ahmed Boucenna; 2007; "Evaluation of technetium transmutation in the experimental fast reactor JOYO"; *Journal of Annals of nuclear energy*; Vol 34(9); pp 703-706.

[88] Cho W.J and Kim G.Y; 2016; "Reconsideration of thermal criteria for Korean spent fuel repository"; *Journal of Annals of nuclear energy*; Vol 88; pp 73-82.

[89] Hearne J.A and Tsvetko P.V; 2019; "Analysis of the transmutation of long-lived fission products using a charged particle beam"; *Journal of Annals of Nuclear Energy*, Vol 133, pp 501-510.

[90] Ingelbrecht C, Lupo J, Raptis K, Altzitzoglou T and Noguere G; 2002; "I-129 targets for studies of nuclear waste transmutation"; *Journal of Nuclear instruments and methods in physics research A*, Vol 480, pp 204-208.

[91] Rehman H, Lee J and Kim Y; 2019; "Photon and neutron hybrid transmutation for radioactive cesium and iodine"; *Journal of Annals of nuclear energy*, Vol 133, pp 527-537.

[92] A Takibayev, M Saito, V Artisyuk and H Sagara; 2005; "Fusion-driven transmutation of selected long-lived fission products; *Journal of Progress in nuclear energy*, Vol 47, pp 354-360.

[93] A comparative study; Nuclear development, Accelerator-driven systems (ADS) and fast reactors (FR) in advanced nuclear fuel cycles; *Journal of Nuclear Energy agency*; pp 263-269, 1999.

[94] Ichimura E, Takaki N, Schram R.P.C, Klein Meulekamp R and Bakker K; 2004; "Iodine transmutation studies using metal iodide targets"; *Journal of Nuclear material*, Vol 334, pp 149-158.

[95] Nakamura M, Yoshida H, Tani S and Inoue T; 1992; "Present status of the omega program in Japan", *Proceeding of the second international information exchange meeting on actinide and fission product separation and transmutation, Argonne, Illinois*, USA.

[96] Beeckmans A, Rixian Z and Diercks J; 1992; "BR2 reactor fission inventory", *Technical note* ABW/130/R0574, 20 May.

[97] Hebel J.A and Cottone G; 1987; "Management modes for iodine-129", *radioactive Waste Management Series*, Vol 7.

[98] Zheng M, Tian W, Zhang D, Qiu S and Su G; 2015; "Minor actinides transmutation in a board type sodium cooled breed and burn reactor core", *Journal of Annals of nuclear Energy*, Vol 81, pp 41-49.

[99] Kora K, Nakaya H, Matsuura H, Goto M, Nakagawa S and Shimakawa S; 2016; "A study on transmutation of LLFPs using various types of HTGRs"; *Journal of Nuclear Engineering Design,* Vol 300, pp 330-338.

[100] Lu H, Ishiwatari Y and Oka Y; 2011; "Study on the LLFPs transmutation in a super-critical water-cooled fast reactor"; *Journal of Nuclear Engineering Design*, Vol 241, pp 395-401.

[101] Wakabayashi T, Takahashi M, Chiba N and Tachi Y; 2020; "A fast reactor transmutation system for 6 LLFP nuclides"; *Journal of Nuclear Engineering Design*, Vol 363, 110667.

[102] Lee M.C, Shin H.S and Koh D.K; 1994; "Analysis of transmutation rates of long-lived radionuclides in the Korean multi-purpose research reactor"; *Proceeding of the third international information exchange meeting on actinide and fission product partitioning and transmutation*, OECD Nuclear Energy Agency, Cadarache, France.

[103] A.A. Kozar; 2001; "Increasing ^{129}I transmutation efficiency"; *Atomic Energy*, Institute of physical chemistry, Russian Academy of Sciences. Vol 91-2, pp 667-674.

[104] Kairat Ismailov, Kenji Nishihara, Masaki Saito and Hiroshi Sagara; 2013; "Optimization study on accelerator driven system design for effective transmutation of iodine-129"; *Journal of Annals of Nuclear Energy*, Vol 56, pp 136-142.

[105] Naima Amrani and Ahmed Boucenna; 2008; "Transmutation of Technetium into stable Ruthenium in high flux conceptual research reactor"; *Journal of Nuclear Instruments and Methods in Physics Research B*, Vol 266, pp 926-928.

[106] Zhong Chen, Zijia Zhao, Dongmei Pan, Baoxin Yuan and Hua Li; 2016; "Preliminary design of inner radius of spallation target of an accelerator driven subcritical reactor for waste transmutation (ADS-NWT)"; *Journal of Progress in Nuclear Energy*, Vol91, pp 217-222.

[107] Ohki S, Naganuma M, Okubo T and Tanaka K; 2013; "Fast breeder reactor core concept for heterogeneous minor actinide loading"; *Journal of nuclear science and technology*, Vol 50, pp 59-71.

[108] Huseyin Yapici; 2003; "Study on transmutation of minor actinides discharged from high burn-up PWR-MOX spent fuel in the force-free helical reactor"; *Annals of Nuclear Energy*; Vol 30, Issue 4; pp 413-436.

Abstract - Résumé - ملخص

Abstract

Nuclear wastes are a major obstacle to the use of the nuclear power with safety. A lot of scientific efforts are devoted to find definitive solutions to this problem. One of the ways envisaged is Separation followed by Transmutation, whose principle is based on the transformation of the long-lived radioactive element by a nuclear reaction to another less radioactively element or stable.

The objective of this work is to study the destruction of fission products in four high flux nuclear reactors. It takes as an example of application Iodine-129. Its transmutation efficiency is obtained from the calculation code ChainSolver 2.34. The results are then compared to choose the best reactor to destroy iodine.

Keywords: Nuclear waste; Fission product; Transmutation; Iodine-129; ChainSolver.

Résumé

Les déchets nucléaires constituent un obstacle majeur à l'utilisation de l''énergie nucléaire en toute sécurité. De nombreux efforts scientifiques sont consacrés à la recherche de solutions définitives à ce problème. L'une des vois envisagées est la Séparation suivie de la Transmutation, dont le principe repose sur la transformation de l'élément radioactif à vie longue par une réaction nucléaire en un autre élément moins radioactif ou stable.

L'objectif de ce travail est d'étudier la destruction des produis de fission dans quatre réacteurs nucléaires à haut flux. On prend comme exemple d'application l'iode-129. Son efficacité de transmutation est obtenue à partir du code calcul ChainSolver 2.34. Les résultats sont ensuite comparés afin de choisir le meilleur réacteur pour détruire l'iode.

Mots-clés: Déchet nucléaire; Produit de fission; Transmutation; Iode-129; ChainSolver.

ملخص

تعتبر النفايات النووية عقبة رئيسية أمام استخدام الطاقة النووية بأمان. يتم تكريس الكثير من الجهود العلمية لإيجاد حلول نهائية لهذه المشكلة. إحدى الطرق المتوخاة هي الفصل الذي يتبعه التحويل، والذي يعتمد مبدأه على تحويل العنصر المشع طويل العمر عن طريق تفاعل نووي إلى عنصر أخر أقل إشعاعية أو مستقرا.

الهدف من هذا العمل هو دراسة تدمير نواتج الانشطار في أربعة مفاعلات نووية عالية التدفق. يأخذ كمثال على تطبيق اليود-129. يتم الحصول على كفاءة التحويل من برنامج الحساب ChainSolver 2.34 ثم تتم مقارنة النتائج لاختيار أفضل مفاعل لتدمير اليود.