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PARTITIONING AND TRANSMUTATION OF ^{99}Tc IN FISSION REACTORS AND HYBRIDS SYSTEMS

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Abstract

Partitioning and transmutation of radioactive and long lived component from the highly radioactive waste stream in order to reduce or probably eliminate their radiotoxic inventory was the important option for the nuclear waste management. The important fission products that deserve most attention is the technetium. Technetium is present as a single isotopic species (^{99}Tc) can be transmuted by single neutron capture into the stable noble metal ruthenium (^{100}Ru). The technetium separation from spent fuel is possible with PUREX process. An, other chemical process was developed to separate a priory technetium with uranium is the UREX process. The transmutation of ^{99}Tc in thermal reactor such as LWRs will be difficult because of the long transmutation half-lives and the large inventory required. Better result can be obtained in fast reactors, or in accelerator driven height flux reactor.

Keywords: *"Technetium-99", "Chemical Separation", "Transmutation"*

I. Introduction

The long lived fission products (LLFPs) generated in fission reactors will remain as radioactive wastes for a long period if they are not transmuted into stable or short-lived nuclides. One of the most important fission products in the nuclear spent fuel is the long-living fission product ^{99}Tc with half life of 2.1×105 year. Technetium-99 is produced as a result of nuclear transformations in nuclear reactors. The cumulative yield for ^{99}Tc for thermal neutron fission of ^{235}U is 6%.[1] . After neutron capture the nucleus ^{100}Tc decay to the stable isotope ^{100}Ru with half life of 15.8s. This makes ^{99}Tc particularly interesting candidate for neutron transmutation. Also it has the ability to migrate in any kind of material, including the storage glasses, due to his high geochemical mobility[2]. Witch could in a worst-case scenario, lead to its release in the environment during long term storage. The chemical extraction of technetium from height level liquid waste HLLW is possible by the PUREX and UREX process. For the transmutation target the technetium metal can be adopted. The transmutation of ^{99}Tc in the fission reactors is possible but it is very difficult, so other systems like the ATW and the EA are envisaged in the future.

II. Chemical processing for separation of Technetium from spent fuel for transmutation:

During fuel dissolution, part of the technetium in metallic or oxide form does not go into solution. This fraction is estimated at 10 to 20 % of the total Tc of UO_2 fuel, accompanies the insoluble residues consisting of noble metals (Ru, Rh, Pd). In nitric medium and in the absence of a reducing agent the dissolved technetium is in its highest valency (VII) which is the most stable. It occur in anionic form in the state of pertechnetate ion TcO_4^-, this species can be extracted by tributil-phosphate (TBP) used by PUREX process as extractant. The PUREX (Plutonium Uranium Extraction) process which is universally employed in the irradiated fuel reprocessing industry is a wet chemical process on the use of tributil-phosphate (TBP) [$(RO)_3P=O$ with $R=C_4H_9$] diluted to 30% by volume with Kerosene[3]. The extraction of technetium proved to be serious hindrance because of its interference with the chemical mechanisms of the partitioning operation (U/Pu separation) in consequences the PUREX process limits the extraction of Tc. We modified the PUREX process in order that only > 99,9 % of the U and >95 % of the Tc are extracted. This Uranium Extraction is called UREX process. The UREX process use acetohydroxamic acid (AHA) [$C_{16}H_{16}N_2O_3$] as extractant agent[4]. Technetium can be separated as pertechnetic acid $HTcO_4$ using monoamide extraction molecule or active carbon absorption method. We can also use a precipitation method by reducing nitric acid for Tc separation from height level liquid waste HLLW.

III. Transmutation of the long lived fission product ^{99}Tc:

Nuclear transmutation can be induced by any particles or quanta enable to penetrate nuclei and to interact with nucleons. The most effective nuclear process that can be used for transmutation of radiotoxic isotope ^{99}Tc, is the neutron absorption[5]. After capturing neutron ^{99}Tc becomes ^{100}Tc, usually in excited state.

$$^{99}Tc \quad (n,\gamma)\, ^{100}Tc \xrightarrow{T_{1/2} = 15.8\ s} {}^{100}Ru$$

This state decays very fast –typically in few ns into ^{100}Tc ground state, by emitting the so-called prompt photons. The ^{100}Tc with half life of 15,8s decays by β^- transition into ^{100}Ru, about 7% of the cases into excited state of this isotope. Then, these ^{100}Ru excited states decay very fast, in less than 1ns, into the ^{100}Ru ground state, emitting so-called delayed photons[6]. Photon – neutron reaction on the other hand have relatively small cross section for transmutation of ^{99}Tc. Both (γ, n) and (γ, 2n) result in ^{98}Tc and ^{97}Tc respectively which have even longer half- lives than ^{99}Tc. Only with (γ, 3n) reaction is ^{96}Tc produced with a short half-life of 4, 28d. Unfortunately the cross section for this last reaction is very small [7]. The fabrication and irradiation of targets for transmutation of Tc has been studied by the EFTTRA group [8]. The technetium metal can be used as target material. It can be obtained by reacting its compounds with hydrogen, ammonia, carbon monoxide or ethanol vapor; by thermolysis of double Tc(VII) halides pertechnetate M_2TcHal_6 (M is an alkali-metal ion or ammonium, Hal=Cl, Br, I) with organic cations; and by electrolysis of technetium solution [9].the metallic target of Tc have been irradiated in high flux reactors (HFR) in petten. The first test has

shown that technetium metal has a good irradiation behavior and no microstructural change[10].

IV. Transmutation reactors:

The neutronic consummation in transmutation reactions is very large. This neutronic excess may be obtained from:

1- The principal actual source of neutrons which is the fission reactors with its two big families, thermal and fast reactors. The transmutation of ^{99}Tc, in the pressure water reactor PWR is very difficult because it can not give the neutronic excess needed for this kind of reaction, excepted if we increase significantly the Uranium enriching rate[11].

2- Hybrid systems: a hybrid system witch also called Accelerator Driven System (ADS) is a combination of an accelerator, a sub-critical reactor and spallation target. The number of neutrons produced in the core from fission reactions is insufficient to maintain the fission reactions so we must produce continually neutrons from an external source (spallation source)[12].

The ATW (Accelerator Driven System for Transmutation of wastes) or the ADT (Accelerator Driven System for Transmutation) is special kinds of ADS destined principally for transmutation of wastes. The attractive solution for the spallation target in ATW system is the Liquid Lead Bismuth Eutectic LBE for coolant a liquid lead or LBE became a primary choice of many preconception designs. Several option of ATW fuel has been also proposed varying from solid oxide fuel (e.g. Pu +AM) to nitrides fuels. In the longer time the perspective Th-based fuel cycle is considered as particularly attractive for the ATW. In 1993 Carlos Rubbia developed a new concept of hybrid system called Energy Amplifier (EA). The Energy Amplifier is an ADS with Th—based fuel cycle and lead as coolant, slowing down neutrons and spallation target. One of the objectives of an EA is to destroy the radiotoxic waste with the concept of Adiabatic Resonance Crossing[13].

V. Nuclear consideration for Technetium Transmutation:

The time needed to incinerate a half initial mass of the nucleus j is called transmutation rate T_j^{transm} [14]. The number of transmuted nucleus during the interval time dt is proportioned to the neutron flux ϕ ($n/cm^2.s$) and the neutron capture cross section σ^j (barns) is given by :

$$dN = \sigma^j . \phi . N' . dt$$

Where N' is the number of initial nucleus of ^{99}Tc.
The number of transmuted nucleus after time t is:

$$N(t) = N' \exp(-\sigma^j . \phi . t)$$

For $t = T_j^{transm}$

$$N(T_j^{transm}) = N'/2 = N' \exp(-\sigma^j . \phi . T_j^{transm})$$

Then :

$$T_j^{transm} = \frac{\ln 2}{\sigma^j \times 10^{-24} \times \varphi}(s)$$

The transmutation rate of a j nucleus is given, as function of the neutron capture cross section σ^j (barns) and the neutron flux ϕ (n/cm^2.s) by, the following relation:

$$T_j^{transm} = \frac{\ln 2}{\sigma^j \times \varphi \times 3{,}16 \times 10^{-17}}(years)$$

Transmutation of the toxic element in nuclear reactors or sub critical system has a sense if the rates of nuclear interactions with neutrons are much higher than rates of natural decays witch are defined by half time $T_{1/2}$. The transmutation under neutron flux can be reasonable if $T_{1/2} >> T_j^{transm}$. The transmutation of the radiotoxic fission product ^{99}Tc in fast neutron reactors with standard flux $\varphi = 10^{15}$ (n/cm^2.s).The neutron capture cross section $\sigma^j = 0.2$ barns for neutrons energy E_n= 0.2 MeV. The residence time in the reactor is T_j^{transm} =110 years. In the thermal reactor, standards flux is $\varphi = 10^{14}$ (n/cm^2.s).The neutron capture cross section σ^j = 4.3 barns for neutrons energy E_n= 1eV. The residence time in the reactor is T_j^{transm} =51 years.

In fact, it is helpful to use two types of neutron consumption definitions, depending on the choice of units, -D* (neutron/transmutation) and -D (neutron/fission). D can be obtained as the product of D* and of the yield of a nuclide per fission, Y. If a LLFP transmuter is fed constantly with a group of nuclides, then the D value of this group is the sum of Y × D* of the group components.

The total neutron consumption needed to incinerate all "transmutable" and "questionable" long lived isotopes of fission products:

$$D(^{99}Tc) = \sum D_j^* Y_j \approx 0.22 \text{ (neutron/ fission in NP)}$$

This value defines the total neutron consumption for incineration of all LLFP presented above, if preliminary isotope separation of fission product in LWR discharge has been realised. To incinerate all Tc without isotopic separation, one needs about 0.15 (neutron/fission in nuclear park). Isotopic separation of ^{99}Tc allows reducing this neutron consumption to 0.08 (neutron/fission in NP). To transmute elements such as Tc, in a fast spectrum transmuter, one needs to know the neutron surplus G available and to the fraction (f) of these transmuter in a Nuclear Power Park (NPP).Taking into account a "standard" value of the neutron parasitic capture (CM) and the neutron leakage (L) as CM + L = 0.3 neutron/fission (which is valid for a fast reactor of an intermediate size and traditional composition), one gets for the neutron surplus (for example in a sub-critical system):

$$G = -\sum \varepsilon_j \times D_j - (CM + L) + \mu \quad ,$$

Where ε_j is a fraction of J-nucleus in fuel, μ is a neutron spallation source ($\mu \cong 0{,}15$ neutron/fission if keff = 0.95). Finally, the maximum rate of transmutation (RT measured in number of transmutations per fission) of any isotope in a given transmuter can be easily evaluated if the neutron surplus G available for LLFP transmutation is known:

$$RTj = \frac{G(\text{neutron/fission})}{D_j^{*}(\text{neutron/transmutation})}$$

One can then calculate the maximum rate max RTj of transmutation of a given isotope J per GWth × year of a transmuter:

$$R_j^{Max}(Kg/GW \times ans) \approx 1.6 \times Aj \times RTj$$

Where A_j is the atomic number of isotope J.

For ^{99}Tc D_j is equal to 1.01 and, in a fast spectrum sub-critical transmuter based on TRU from LWR-UOX, G = 0.96. Then, $R^{max}j \sim 160$ kg/ GWth × year if all neutron surplus is devoted to ^{99}Tc transmutation.

VI. Conclusion

One of the most important fission products in the nuclear spent fuel is the long-living fission product ^{99}Tc with half life of 2.1×105 year. His chemical separation is possible with success using PUREX (Plutonum Uranium Refining by EXtraction) process based on the tributil-phosphate (TBP) as extractant agent. Better results are obtained by the UREX (URanium EXtraction) process. After neutron capture the nucleus ^{100}Tc decay to the stable isotope ^{100}Ru with half life of 15.8s. This makes ^{99}Tc particularly interesting candidate for neutron transmutation. The metallic technetium can be used as target material. The transmutation of ^{99}Tc in the fission reactors is possible but other systems like the accelerator driven for transmutation of waste (ATW) and the energy amplifier (EA) are envisaged in the future.

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