Transmutation of high-level radioactive waste - Perspectives

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Abstract
In a fast neutron spectrum essentially all long-lived actinides (e.g. Plutonium) undergo fission and thus can be transmuted into generally short lived fission products. Innovative nuclear reactor concepts e.g. accelerator driven systems (ADS) are currently in development that foresee a closed fuel cycle. The majority of the fissile nuclides (uranium, plutonium) shall be used for power generation and only fission products will be put into final disposal that needs to last for a historical time scale of only 1000 years. For the transmutation of high-level radioactive waste a lot of research and development is still required. One aspect is the precise knowledge of nuclear data for reactions with fast neutrons. Nuclear reactions relevant for transmutation are being investigated in the framework of the european project ERINDA. First results from the new neutron time-of-flight facility nELBE at Helmholtz-Zentrum Dresden-Rossendorf will be presented.

1 Motivation for nuclear transmutation

In the European Union a significant share of the gross electricity generation (27.6% in 2011) [1] comes from its 136 nuclear power reactors (incl. Switzerland) with a total electric power of about 125 GW. For example, France has 58 operational power reactors, the United Kingdom 16 and Sweden 10. The spent nuclear fuel constitutes the high-level radioactive waste that is produced with a rate of approximately 2500 tons per year.

In a once-through fuel cycle the nuclear waste management has to rely on direct final disposal of spent nuclear fuel after a certain time of intermediate storage to reduce the decay heat. By nuclear reprocessing the uranium and plutonium can be separated and reused for power generation, while the fission products and minor actinides are vitrified and conditioned for final disposal in a deep geological site. The repeated use of mixed oxide fuel in current power reactors can lead to a reduction of the plutonium inventory by about 25-45% [2].

In order to assess the radiological threat of spent nuclear fuel, we will now compare it with natural uranium. The ingestion radiotoxicity of 1 ton of spent nuclear fuel (4.5% UOX PWR with a burn up of 55 MWd/kg) is shown in Fig. 1. The long-lived radiotoxicity (> 10000 years is dominated by plutonium while below minor actinides are important. The short-lived radiotoxicity < 60 yrs is dominated by fission products. A common reference level is the ingestion radiotoxicity of the natural uranium required to produce 1 ton of enriched U-nuclear fuel (4.2% ^{235}U, 7.83 t nat. U) in equilibrium with its decay products: $1.47 \cdot 10^5$ Sv / tHM.

The mobility in the environment depends among other factors on the volatility of the chemical elements e.g. Cs, Sr, Tc, I, Se having a high volatility. This aspect is not included in Fig.1 but it is taken into account in the safety analysis of final repository concepts. The radiotoxicity of the long-lived fission products is lower than that of the long-lived actinides, but their activity is in the same order of magnitude.

From Fig. 1 the potential of partitioning and transmutation (P&T) as an alternative strategy to direct disposal of the long-lived waste is obvious: The long-term radiotoxicity will be reduced from geological to historical time scales of about 1000 years if all long-lived nuclides can be transmuted

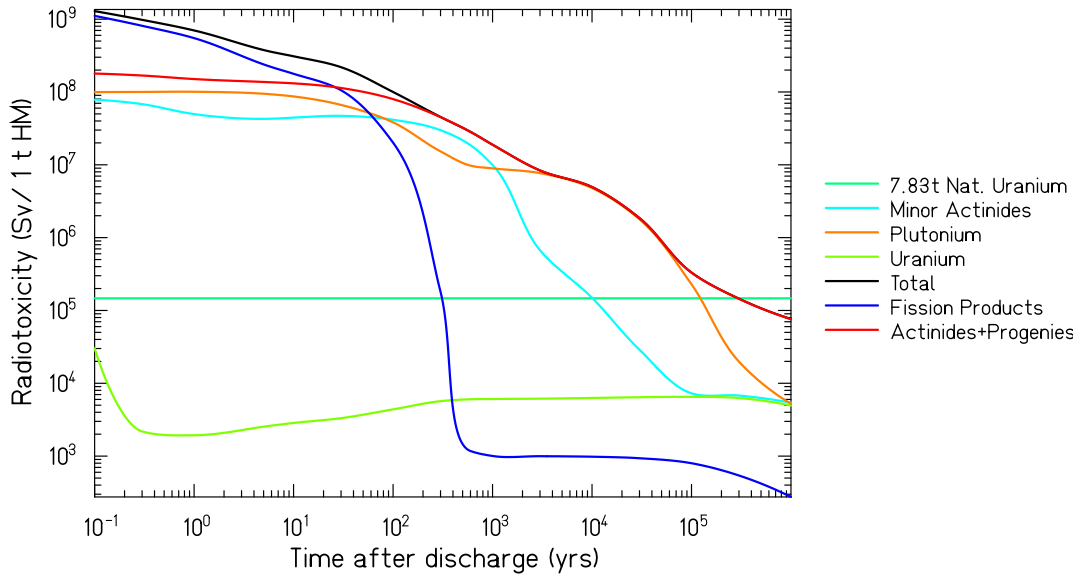

Fig. 1: Radiotoxicity of spent nuclear fuel in Sv per 1 ton of heavy metal as a function of time after discharge from the reactor. The radiotoxicity of plutonium and other (minor) actinides decay to the natural level of the uranium ore required to produce the fuel after 10-100 thousand years and thus their removal by transmutation is especially important. The reference level of the natural uranium required to produce 1 ton of enriched U-nuclear fuel is a horizontal line at $1.47 \cdot 10^5$ Sv / tHM.

into shorter lived fission products. The physics of transmutation is discussed in many review articles, e.g. [3, 4]. Among the expected benefits of P&T the transuranium elements in the waste will be strongly reduced. The long term radioactive inventory, decay heat and peak dose rate from the final repository will be strongly reduced, see [5] and references therein.

2 Transmutation technologies and options

The development and safety of Partitioning and Transmutation is a subject of current research in many european and national projects [6]. In Generation IV reactor concepts a closed nuclear fuel cycle is foreseen that will also include nuclear transmutation to significantly reduce the amount of long-lived actinides. The reactor design can be based on modern supercomputers where detailed thermo hydraulic and neutron-transport coupled simulations in realistic geometry are to be done. Fundamental simulation of the processes on the atomic level in parts of the reactor core are possible. Especially in the fast neutron range precise nuclear data for neutron induced reactions are required. Many such nuclear data experiments have been supported by the FP6-EFNUDAT, FP7-ERINDA and FP7-CHANDA projects. In the following paragraphs different technologies for transmutation are introduced, this is done on a basic level, that must be incomplete due to the limited space. References for further information are included. From the arguments given, one should not be tempted to give preference to either one technology, because this will mostly depend on the waste management or nuclear fuel cycle strategy the technology is being developed for.

Based on french legislature from 2006 the Advanced Sodium Technological Reactor for Industrial Demonstration (ASTRID) is being developed by a consortium lead by CEA. ASTRID is planned to be a 600 MWe sodium-cooled fast reactor and it is the most advanced generation IV reactor project. The preliminary design phase II has already been started. Sodium cooled breeder reactors have been developed and operated over many decades giving this technology a solid basis. The main motivation of

ASTRID is to recycle plutonium without limitation. A fuel cycle to transmute and manage americium is also an important topic under study. Homogeneous fuel with a content of 2% Am or heterogeneous fuel with up to 10 % Am in a blanket can be used for transmutation [7]. In principle, the minor actinide content in a fast reactor fuel is limited by the criticality control. The criticality is influenced by the delayed neutron fraction and by the Doppler broadening of the neutron resonances. The corresponding reactivity coefficients get less favourable for fuels containing high concentration of minor actinides.

In an accelerator driven system (ADS) a high-power proton accelerator is coupled to a spallation target that is surrounded by a subcritical reactor core. To allow for a fast neutron spectrum for transmutation the ADS needs to be liquid-metal cooled. If the effective neutron multiplication factor of the reactor core (k_{eff}) is kept below 1 the system cannot have a self-sustained chain reaction. The spallation target is a strong source of neutrons that leads to fission reactions in the reactor core. The accelerator driven system concept was developed at Los Alamos [8] for a thermal spectrum and at CERN [9] for a fast spectrum nearly 20 years ago.

To demonstrate the accelerator driven system technology, i.e. the coupling of the three components (accelerator, spallation target and subcritical reactor) at realistic power level to draw conclusions on the industrial realisation the Multi-purpose hYbrid Research Reactor for High-tech Applications (MYRRHA) project has been established at SCK-CEN, Mol Belgium [10]. A Pb-Bi eutectic cooled fast reactor shall be built that can be operated in critical and subcritical mode. The thermal power is in the range of 65-100 MW. With a high-power proton linac of 600 MeV, 4mA and a subcritical core configuration ($k_{eff} = 0.95$) MYRRHA shall operate with mixed oxide U/Pu fuel. The subcritical layout of the ADS removes limitations due to the reduced fraction of delayed neutrons from fuels rich in minor actinides.

In order to achieve nearly full transmutation of all long-lived actinides several fuel cycles are required in solid fueled nuclear reactors, as the change in reactivity in the core during the operation and also radiation damage to the fuel elements limits the achievable burn up. Partitioning of the spent nuclear fuel, i.e. chemical separation of minor actinides and fission fragments from uranium and plutonium will be necessary as well as refabrication of fuel elements that will be much more radioactive than conventional unirradiated enriched uranium oxide fuel elements. The neutron rate of curium is rather high and it can be advantageous to first transmute Pu, Am and let the curium decay first [3].

Using molten salt technology, e.g. ^{7}LiF or NaF, ZrF_4 together with (U,Pu,M.A.)F_4 a nuclear fuel is produced that is transparent and liquid at high temperatures $T \approx 600°$C. The chemical reprocessing can be simplified as solid fuel fabrication and refabrication does not occur. Also radiation damage in the solid fuel is not a limiting factor. A molten salt reactor exhibits large negative temperature and void coefficients of reactivity that improve the operational safety due to the thermodynamical properties of its liquid fuel. The technology has been realized once in the molten salt reactor experiment at Oak Ridge in the 1960s, where a Thorium fuel cycle was envisioned to breed ^{233}U in a thermal spectrum. In a molten salt reactor volatile fission products are continuously emitted from the fuel and need to be treated, e.g. gaseous fission products must be extracted with an off-gas system. The Molten Salt Fast Reactor (MSFR) is one of the generation IV concepts [11,12]. Fuel Salt chemistry for high burn up of actinides and online reprocessing as well as structural material aspects at high temperatures and neutron irradiations are being investigated, e.g. in the FP7-EVOL project. The transmutation potential has been investigated e.g. for the German phase out of nuclear power and more than 90% transmutation efficiency has been found by simulations [13].

3 nELBE A new time-of-flight facility for nuclear data

For the development of nuclear waste management strategies and technologies for transmutation precise nuclear data especially in the range of fast neutrons are required. Important reactions to be studied are for example the neutron-induced fission of plutonium isotopes and minor actinides as well as the inelastic

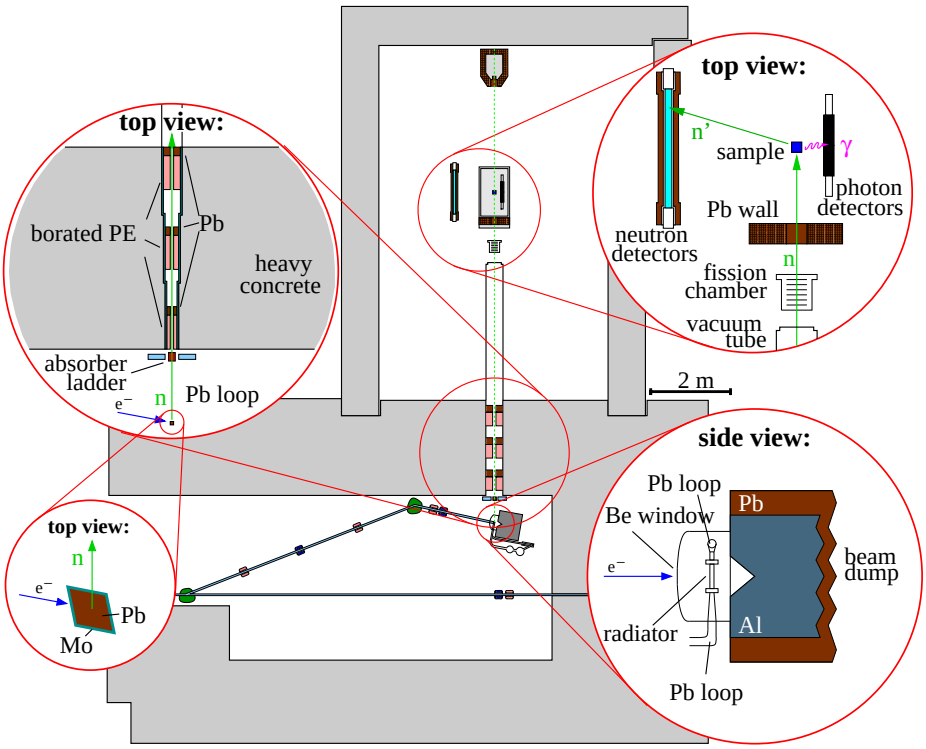

Fig. 2: Floorplan of the new nELBE facility at Helmholtz-Zentrum Dresden-Rossendorf. Insets show the neutron producing target, the collimator and the experimental setup in the time-of-flight hall. The maximum flight path is 11 m.

neutron scattering on structural, fuel and coolant materials, see e.g. [14].

The nELBE time-of-flight facility at Helmholtz-Zentrum Dresden-Rossendorf (HZDR) which – based on a superconducting electron accelerator – is dedicated to measurements in the fast neutron range has been extensively rebuilt. The time-of-flight path is now between 4 and 11 m. A much larger time-of-flight experimental hall allows us to reduce the background from scattered neutrons as all walls including ceiling and floor are at least 3 m away from the evacuated neutron beam line. The liquid lead circuit used as a neutron producing target has also been redesigned and rebuilt. Through the liquid-lead technology the neutron beam intensity is not limited by the heat dissipation inside the target. The technical design including thermomechanical parameters of the liquid lead circuit and the beam dump has been discussed in [15]. The neutron source strength at the nominal beam current of 1 mA has been calculated to be 10^{13} neutrons/s, [16]. The accelerator produces high brilliance electron beams with variable micropulse repetition rates and duty cycles. The electrons are accelerated up to 40 MeV in continuous wave-mode by superconducting radio frequency cavities. The maximum average beam current at a micropulse rate of 13 MHz is 1 mA. For typical time-of-flight measurements the repetition rate is reduced to 100-200 kHz resulting in a source strength of about 10^{11} n/s. The bunch length is about 5 ps, so that the time-of-flight resolution is not degraded and short flight paths can be used with a high resolution detection system. Figure 2 shows the floor plan of the new neutron time-of-flight facility in the Center for High Power Radiation Sources of HZDR.

The electron beam passes through a beryllium window mounted on a stainless-steel vacuum chamber and hits the radiator, consisting of a molybdenum channel confining the liquid lead. The channel has a rhombic cross section with 11 mm side length. The electrons generate bremsstrahlung photons which release neutrons in secondary (γ,n) reactions on lead. These leave the radiator almost isotropically, while

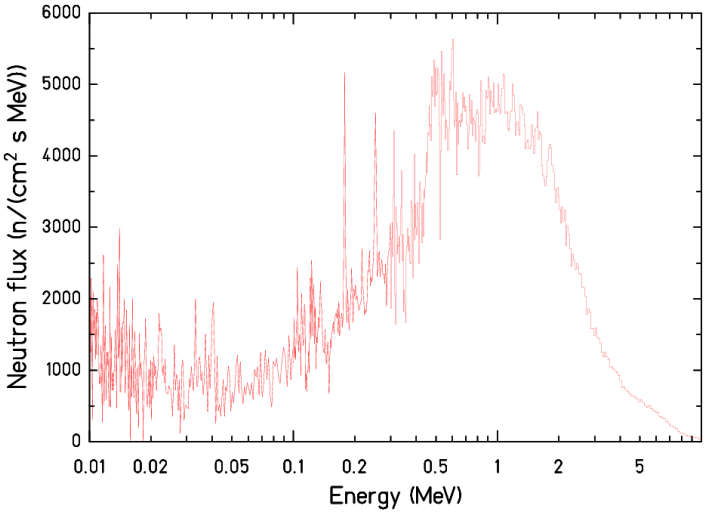

Fig. 3: Neutron spectrum measured with a ^{235}U fission chamber (H19, PTB Braunschweig) at a flight path of 815 cm. The electron beam energy was 30 MeV and the average current 6 μA with a measurement live time of 15 hours.

the angular distributions of electrons and photons are strongly forward-peaked. The collimator and the resulting neutron beam properties at the experimental area have been optimized using MCNP in order to maintain the correlation of time-of-flight and neutron energy. The collimator of 2.6 m length contains three replaceable elements of lead and borated polyethylene that are mounted inside a precision steel tube [16].

The redesigned neutron producing target and collimator have the same dimensions as before the extension of the facility, consequently a very similar neutron spectrum and spatial beam profile is expected. In a first beam time, the neutron spectral rate and the beam profile has been measured. In Fig. 3 the neutron spectral rate measured with a ^{235}U fission chamber is shown. At 30 MeV electron energy the usable neutron energy range extends from ca. 100 keV to 10 MeV. Absorption dips at 78,117, 355, 528, 722, 820 keV are due to ^{208}Pb scattering resonances. Pb is used as the neutron producing target, as collimator material and as bremsstrahlung absorber at the collimator entrance. Emission peaks at 40,89,179, 254, 314, 605 keV come from the near threshold photoneutron emission of ^{208}Pb (strong capture resonances of ^{207}Pb) [17]. The beam profile shown in Fig. 4 was measured by moving a plastic scintillator (length: 1 m, width: 11 mm) through the beam. The scintillator was read out on both ends by high gain photomultiplier tubes. By setting a gate on the time-of-flight spectrum the bremsstrahlung and neutrons can be separated. A fit assuming a rectangular beam profile with a sharp edge shows that the beam has some halo that might be attributed to a too small window at the exit of the neutron beam line. The beam profile measurement technique is described in [17].

To study the inelastic scattering a first test with $LaBr_3$ scintillators has been made. The target was a disk of ^{nat}Fe with a diameter of 7 cm and thickness 0.80 cm. The $LaBr_3$ detector was 30 cm away from the target under an angle of 159 degrees. Despite the short measurement time the 847 keV and 1238 keV gamma rays from the inelastic scattering ^{56}Fe(n,n') are clearly visible. Fig. 5 shows the time-of-fight to gamma-ray energy correlation matrix measured with the $LaBr_3$ scintillator. The time-of-flight was calibrated using the bremsstrahlung peak shown as a horizonal line. The gamma ray line at 1468 keV is from intrinsic radioactivity of the ^{138}La and can be used to monitor the PMT gain. It is planned to study the inelastic scattering with several $LaBr_3$ scintillators under different angles

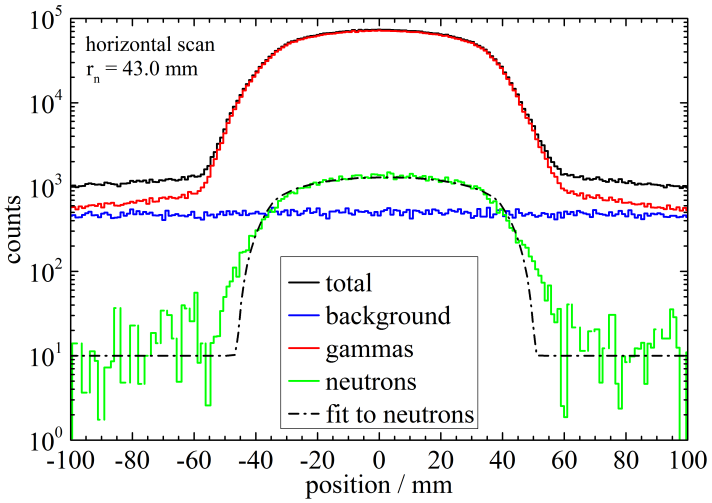

Fig. 4: Time-of-flight beam profile measured with a plastic scintillator moved through the beam in horizontal direction. A time-of-flight gate was used to determine the bremsstrahlung and neutron beam profile separately.

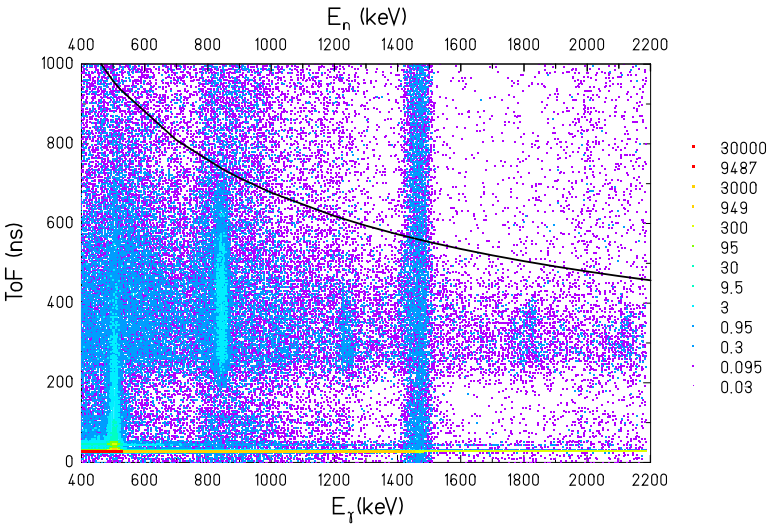

Fig. 5: Energy vs. time-of-flight spectrum of gamma rays emitted from ^{56}Fe(n,n') measured with a 3*3" $LaBr_3$ scintillator with an angle of 159 degrees to the beam direction. The flight path is $\approx$ 936cm. The black line corresponds shows the time-of-flight curve of neutrons with kinetic energy E_n (upper abscissa). The live time of the measurement is 1291 s.

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