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Study of a low-power, fast-neutron-based ADS

M. Ripani[a], S. Frambati[b], L. Mansani[b], M. Bruzzone[b], M. Reale[b], S. Monti[c], M. Ciotti[d], M. Barbagallo[e], N. Colonnae[e], A. Celentano[f], M. Osipenko[f], G. Ricco[f], P. Saracco[f], C.M. Viberti[f], O. Frasciello[g], P. Boccaccio[g], J. Esposito[g], A. Lombardi[g], M. Maggiore[g], L. Piazza[g], G. Preteh[g], R. Alba[h], L. Calabretta[h], G. Cosentino[h], A. Del Zoppo[h], A. Di Pietro[h], P. Figuera[h], P. Finocchiaro[h], C. Maiolino[h], D. Santonocito[h], M. Schillacii[h], A. Kostyukov[i], A.Cammi[j], S. Bortot[j], S. Lorenzi[j], M. Ricottik[j], S. Dulla[k], P. Ravetto[k], A. Rebora[l], D. Chiesa[m], M. Clemenza[m], E. Previtali[m], M. Sisti[m], D. Alloni[n], A. Borio di Tigliole[n], M. Cagnazzo[n], R. Cremonesi[n], G. Magrotti[n], S. Manera[n], F. Panza[n], M. Prata[n], A. Salvini[n]

[a]*INFN, Sezione di Genova, Via Dodecaneso 33, Genova 16146, Italy*
[b]*Ansaldo Nucleare SpA, C.so F.M. Perrone 25,Genoa 16152, Italy*
[c]*ENEA, Via Martiri di Monte Sole, 4, Bologna 40129, Italy*
[d]*ENEA, Via Enrico Fermi, 45, Frascati (Rome) 00044, Italy*
[e]*INFN, Sezione di Bari, Via E. Orabona n. 4, Bari 70125, Italy*
[f]*INFN, Laboratori Nazionali di Frascati, Via Enrico Fermi, 40, Frascati (Rome) 00044, Italy*
[g]*INFN Laboratori Nazionali di Legnaro, Viale dell'Università 2, Legnaro (Padova) 35020, Italy*
[h]*INFN Laboratori Nazionali del Sud, via S.Sofia 62, Catania 95125, Italy*
[i]*Lomonosov Moscow State University, GSP-1, Leninskie Gory, Moscow 119991, Russian Federation*
[j]*Politecnico di Milano, Piazza L. da Vinci, 32, Milan 20133, Italy*
[k]*Politecnico di Torino, Corso Duca degli Abruzzi, 24, Turin 10129, Italy*
[l]*Università di Genova, Via allOpera Pia, 15, Genoa 16145, Italy*
[m]*Università di Milano Bicocca, Piazza dell'Ateneo Nuovo, 1, Milan 20126, Italy*
[n]*Università di Pavia and LENA, via Aselli 41, Pavia 27100, Italy*

Abstract

Within European Partitioning & Transmutation research programs, infrastructures specifically dedicated to the study of fundamental reactor physics and engineering parameters of future fast-neutron-based reactors are very important, being some of these features not available in present zero-power prototypes. This presentation will illustrate the conceptual design of an ADS with high safety standards, but ample flexibility for measurements. The design assumes as base option the 70 MeV, 0.75 mA proton cyclotron facility planned to be constructed at the INFN National Laboratory in Legnaro, Italy and a Beryllium target, with Helium gas as core coolant. Safety is guaranteed by limiting the thermal power to 200 kW, with a neutron multiplication coefficient around 0.94, loading the core with fuel containing Uranium enriched at 20% and a solid-lead diffuser. The small decay heat can be passively removed by thermal radiation from the vessel. Such a system could be used to study, among others, some specific aspects of neutron diffusion in lead, beam-core coupling, target cooling and could serve as a training facility.

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Keywords: Accelerator Driven Systems; Fast nuclear reactors; Radioactive waste; Transmutation; Neutrons; Accelerators

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1. Introduction

Operation of the widespread thermal nuclear reactors results in the accumulation of important quantities of highly radioactive, highly toxic, long-lived nuclear materials, in the form of plutonium, minor actinides and long-lived fission products. In the European Union, annually, about 2500 tons of spent fuel are produced, of which about 25 tons are plutonium isotopes, 3.5 tons are minor actinides and about 3 tons are long lived fission products. In some countries, spent fuel is reprocessed and used for the fabrication of mixed oxide (MOX) fuels, however with limited impact on the continuous build-up of these materials in storage sites. Different solutions have been proposed and minimization of waste has become an important aspect in the development of innovative nuclear energy systems. In fact, many Minor Actinides (MA) produced in reactor operation like ^{237}Np, ^{241}Am, ^{244}Cm, have a fission cross section threshold of about 0.5 MeV. Therefore a fast spectrum can lead to burning MA via fission quite effectively due to the high density of energetic neutrons. Here fast refers to the fact that, on the contrary of what happens in a thermal reactor, neutrons are not slowed down by a moderator inside the reactor core, thus maintaining a higher average energy (i.e. a higher velocity). The more energetic neutron spectrum is also achieved by using as a coolant a medium or heavy element, such as sodium or lead, or a low density gas. As a result, in fast reactors much of the neutron spectrum is above several hundred keV. For this and other reasons specifically related to the choice of cooling medium, among innovative nuclear reactor concepts, lead-cooled fast systems are of particular interest and many international research projects are focused on this technology: ELSY [Cinotti L. et al. (2007)][Alemberti (2009)][Cinotti (2011)], CDT [De Bruyn (2010)] and LEADER [LEADER (2009)].

However, in recent years it has become clear that safety aspects of fast critical reactors do not allow to load the core with Minor Actinides beyond certain limits, due to the relatively small fraction of delayed neutrons (essential for reactor control) in the actinide fission process [Nifenecker (1999)]. Accelerator-Driven Systems (ADS) could instead be particularly well-suited to maximize the transmutation rate, while still operating in a highly safe regime [NEA (2002)]. The main difference between a nuclear reactor and an ADS is that in the first case the effective multiplication coefficient is kept equal to unity, thereby ensuring that the fission chain reaction is self-sustained, while ADS are characterized by an effective multiplication coefficient smaller than unity (typically 0.95-0.98). Thus, to maintain a steady state condition of operation in a ADS, additional neutrons must be supplied by an external neutron source, by using an accelerated proton or deuteron beam impinging on a neutron production target. Most ADS designs are based on a fast reactor core, modified to obtain multiplication coefficients less than unity.

Another important aspect in the framework of innovative nuclear energy system projects is the opportunity to use infrastructures for research and study where experimental tests on new concepts can be performed, in order to validate measurement methodologies, simulation codes and data libraries, as well as to improve our understanding of the complex dynamic and kinetic effects in fast-neutron heavy-metal-cooled sub-critical systems. Therefore also in ADS design, lead technology is present in several projects: PDS-XADS [Cinotti et al. (2003)], IP-EUROTRANS [Rimpault et al. (2010)][Bruyn et al. (2010)][Granget et al. (2010)], Guinevere [Billebaud et al. (2009)], MYHHRA [At Abderrahim et al. (2010)] and EFIT [Mansani et al. (2012)]. The Guinevere setup at SCKCEN in Mol, Belgium [Billebaud et al. (2009)], based on a D+T accelerator providing nearly monochromatic 14 MeV neutrons embedded in a solid Lead matrix started operations in 2011.

Among the basic R&D requirements are the capability to test and develop experimental methods for the on-line measurement of sub-criticality in ADS systems and the need for hands-on experience on the kinetic and dynamic behavior in fast systems. Such an experience is essential in order to validate our theoretical understanding of the main processes and parameters underlying fast neutron systems, but it is also fundamental to assess the potential impact of these effects on control and safety parameters. It is of paramount importance to develop and build facilities powerful enough to fulfill a majority of these requests, but sufficiently low-power to not overcome the zone of comfortable, high-safety operation.

Following these requirements, a proposal was put forward by INFN, in collaboration with Ansaldo Nucleare, ENEA, Politecnico di Milano, Politecnico di Torino, LENA-Pavia [CDR (2012)]. The collaboration was recently extended to the University of Genova. Besides the motivations outlined above, the proposal got momentum from the availability in the very near future of the proton cyclotron purchased by INFN as driver for the SPES project on radioactive ion beams [Prete (2012)]. In this paper I will briefly describe the project.

2. Radioactive waste, issues and classification

The sustainability of nuclear power with innovative closed fuel cycles relies, among other aspects, on the availability of nuclear systems able to burn minor actinides, that is, high level radioactive waste from the discharge of power reactors that present the most radiotoxicity and long term disposal problems. ADS would offer the perspective to burn large quantities of minor actinides in relatively short times and could potentially reduce in a substantial way (from 10 to 1000 times) both the final amount of radioactive inventory to be stored in geological deposits and the storage times (from hundreds of thousands of years to a few centuries). The problem of closing the nuclear fuel cycle and, more generally, the problem of radioactive waste management in the industrial production of electricity could therefore find a definitive answer, avoiding, in perspective, the considerable burden on future generations presented by a final storage that must be guaranteed for geological periods.

In its 2009 document, the IAEA classifies radioactive waste as follows [IAEA (2009)]. Exempt waste (EW) has such a low radioactivity content, which no longer requires controlling. Very short-lived waste (VSLW) can be stored for a limited period of up to a few years to allow its radioactivity content to decrease by radioactive decay. It includes waste containing radionuclides with very short half-lives often used for research and medical purposes. Very low level waste (VLLW) usually has a higher radioactivity content than EW but may, nonetheless, not need a high level of containment and isolation. Typical waste in this class includes soil and rubble with low levels of radioactivity which originate from sites formerly contaminated by radioactivity. Low level waste (LLW) has a high radioactivity content but contains limited amounts of long-lived radionuclides. It requires robust isolation and containment for periods of up to a few hundred years and is suitable for disposal in engineered near-surface facilities. It covers a very broad range of waste and may include short-lived radionuclides at higher levels of activity concentration, and also long-lived radionuclides, but only at relatively low levels of activity concentration. Intermediate level waste (ILW), because of its radioactivity content, particularly of long-lived radionuclides, requires a greater degree of containment and isolation than that provided by near surface disposal. It requires disposal at greater depths, of the order of tens of meters to a few hundred meters. High level waste (HLW) presents levels of activity concentration high enough to generate significant quantities of heat by the radioactive decay process, or it contains large amounts of long-lived radionuclides that need to be considered in the design of an ad-hoc disposal facility. Disposal in deep, stable geological formations usually several hundred meters or more below the surface is the generally recognized option for disposal. Often surface and deep repository are designed together and comprise additional infrastructures, such as to form a High-Tech Campus.

In Italy, a more compact classification scheme that uses Categories based on activity was adopted [ENEA-DISP]. According to this scheme, First Category waste corresponds to IAEA VLLW, Second Category waste corresponds to LLW and ILW Short Lived and finally Third Category waste corresponds to A) LLW and ILW Long Lived, with less than 4000 Bq/g α-emitters and B) HLW with more than 4000 Bq/g α-emitters and more than 100 W/m^3 thermal emission. It has been estimated that in Italy, after decommissioning of nuclear installations (including old reactors and research facilities) and after reprocessing of old spent fuel there will be: 65000 m^3 Second Category waste, for which there appears to be the need for a definitive surface repository; 10000 m^3 Third Category waste, which is probably a quantity not relevant enough to require a deep geological repository, but which still requires an engineered (temporary) surface repository for long-term storage. It is worth noticing that current Italian law requires construction of a national repository, that should also become a High-Tech Campus.

3. Nuclear waste transmutation

Transmutation (or nuclear incineration) of radioactive waste can take place due to neutron-induced reactions that transform long-lived radioactive isotopes into stable or short-lived isotopes. In the case of Long Lived Fission Fragments (LLFF) like e.g. ^{151}Sm, ^{99}Tc, ^{121}I, ^{79}Se, etc. transmutation can occur via neutron radiative capture (n,γ) like e.g. in the reaction $n + {}^{99}Tc\ (2.1 \cdot 10^5\ y) \longrightarrow {}^{100}Tc\ (16\ s) \longrightarrow {}^{100}Ru\ (stable)$. For Plutonium and Minor Actinides like e.g. ^{240}Pu, ^{237}Np, ^{241,243}Am, ^{244,245}Cm, etc., transmutation can occur via either neutron-induced fission (n,f) or neutron capture (n,γ). Apart for ^{245}Cm, minor actinides are characterized by fission threshold around the MeV neutron energy. Such isotopes can efficiently be burned in fast reactors, where the neutron spectrum typically ranges from 10 keV to 10 MeV. An important remark to be made is that, due to the sub-criticality of the system, in an ADS delayed neutrons are less relevant for reactor control [Bosio et al. (2001)], since the kinetics is dominated by prompt neutrons which follow the time-behavior of the external neutron source. Therefore a fast ADS offers a more ample capability in terms of adding Transuranic elements to the fuel and burning them.

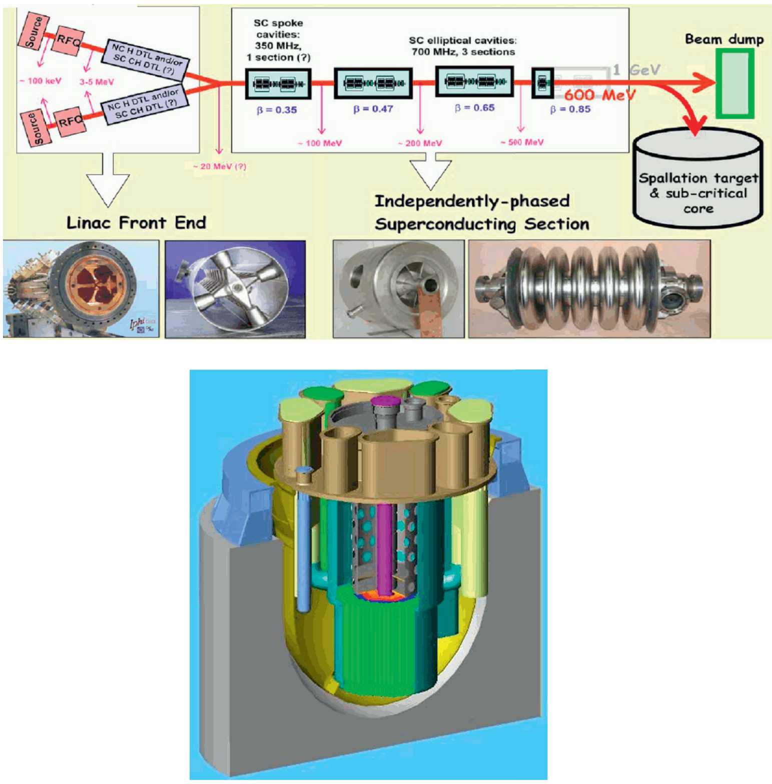

Fig. 1. Typical ADS layout, comprising an accelerator (in this case a proton accelerator), a beam transport system and a subcritical reactor core to which the beam is coupled. Pictures are taken from the EFIT project [Mansani et al. (2012)].

4. A low-power ADS

The typical layout of an ADS is shown in Fig. 1. Electrons, protons or other ions are accelerated by a specific particle accelerator. The accelerated beam is then transported to a subcritical reactor core, i.e. a core for which the effective neutron multiplication coefficient k_{eff} is less than unity. The fact that $k_{eff} < 1$ means that at each fission, the number of neutrons that will in turn produce another fission is less than unity, which implies that there is no self-sustained chain reaction. As a consequence, the reactor can be operated in steady state conditions only if additional neutrons are supplied through an external source. External neutron production is namely accomplished by driving the beam onto an appropriate target, where the beam is completely absorbed and undergoes a number of nuclear interactions resulting in neutron production.

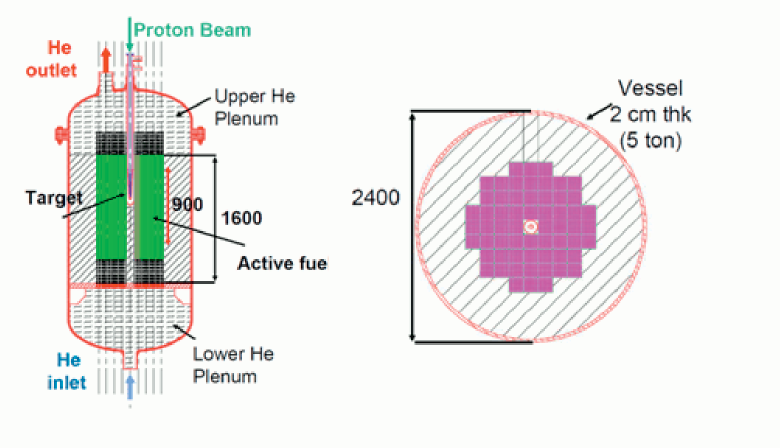

Fig. 2. Overview of the proposed fa cility. The lead radial reflector is not to scale in order to improve image readability. Left: side view of the facility, with the core (160 cm of height, 90 cm of which containing active fuel), lower and upper helium plena and inlet/outlet channels, axial and radial reflectors. The proton beam enters from the vessel upper head. Right: plane view of the active core, with 60 active elements, the radial reflector and the central convertor position.

The idea of the project outlined in this paper was originated by the availability of a 70 MeV, 0.75 mA proton cyclotron purchased by INFN as driver for the SPES project on radioactive ion beams at the INFN National Laboratory of Legnaro (LNL) [Prete (2012)]. For the core design, UO2 with 20 wt% 235U was chosen as the fuel, to avoid security issues related to handling Pu. The fuel rods and assemblies are arranged in such a way to guarantee a neutron multiplication coefficient ~ 0.95, which is the limit for waste storage facilities. The resulting thermal power was required to be in the order of 150-200 kW, sufficiently low to limit safety issues but sufficiently high to study some aspects of dynamics. The corresponding core temperature should not exceed 300^oC, which allows to use a solid Lead matrix being well below the melting point (however, to ensure mechanical stability, the Lead matrix would be embedded in a steel structure). This simplifies engineering aspects further as no liquid metal circulation system has to be designed. Cooling of the core would be performed by circulating He gas through small channels in between the fuel elements. As convertor to produce the neutrons from the incident 70 MeV proton beam, Beryllium was chosen, as the presence of a weakly bound neutron provides abundant neutron production at this relatively low beam energy, in the order of 0.1 neutrons per incident proton. A dedicated measurement of the neutron yield from Beryllium at the similar energy of 62 MeV was performed by an INFN team at the Laboratori Nazionali del Sud in Catania, Italy [Alba et al. (2012)][Schillaci (2012)].

Fig. 3 shows a side and top view of the vessel containing the active core, with the convertor target situated approximately at the center of the core and the inlet for the incident proton beam. The geometry of the beryllium target was been derived from the conical shape already used for the TRADE project [Rosa (2005)]. The conical surface allows the distribution of the beam power on a wide surface, thus helping target cooling. Beryllium was chosen both because it provides a very good proton-to-neutron conversion factor (mainly due to (n,xn) reactions on the ^{9}Be isotope), and due to its high thermal conductivity, about 6 times that of lead. The total thermal power deposited on the target by the 70 MeV, 0.75 mA proton beam is about 50 kW. The beryllium converter is also cooled by Helium gas, with a bayonet-shaped cooling channel allowing helium to enter and exit the cooling channel from the top of the reactor vessel.

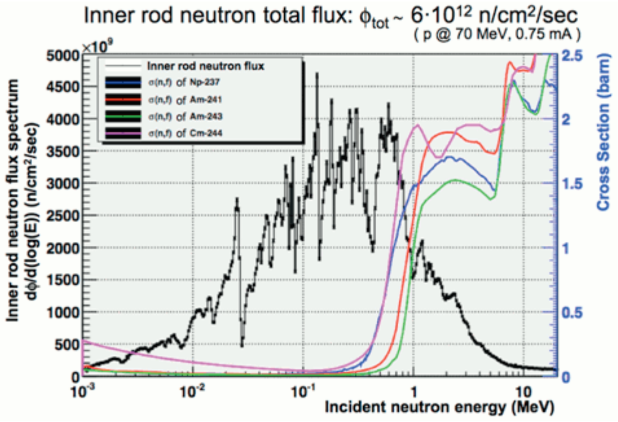

Fig. 3. Inner rod neutron flux spectrum compared with the fission neutron cross-section in MA. The integrated flux is around $6 \cdot 10^{12}$ n/cm^2/sec while the flux useful for fast fission, above 0.5 MeV, amounts to 30% of the integrated flux.

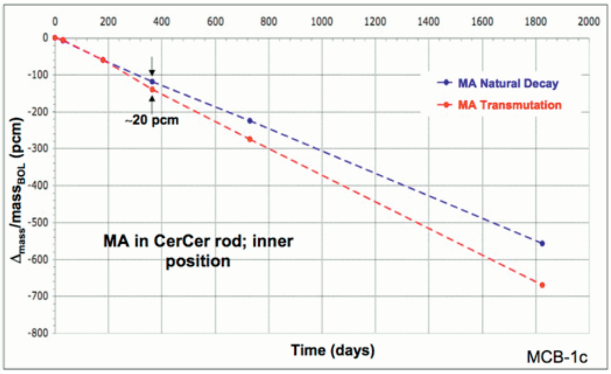

Fig. 4. Minor Actinides total mass relative change in CerCer rod respectively in the reactor (in red) and in the case of pure natural decay (in blue).

Fig. 4 shows the neutron spectrum simulated with the MCNPX code [Pelowitz (2008)], together with some representative fission cross sections for minor actinides. The initial value of k_{eff} is around 0.94. The total evaluated reactor power with its systematic error is $P_{tot} = (199 + 2910)$ kW$_{th}$, about 0.4 W$_{th}$/cm^3 . A preliminary estimate of the behaviour of k_{eff} as a function of time showed variations of the order of 0.3% (300 pcm) within the first six years of operation. As part of the system design, burn out of some test rods was investigated. In detail, an inner fuel rod was replaced with a CerCer rod type [Artioli et al. (2008)] containing a specific combination of minor actinides. The burn out simulations were performed with MCB-1c [Cetnar et al. (1998)] and are shown in Fig. 4, compared with the natural decay of the same materials. The comparison between the two curves shows that, after one year of continuous irradiation, the mass of minor actinides is reduced of more than 15% compared to the expected quantity due to natural decay (0.02% respect to the initial mass). Clearly, in order to observe experimentally these small differences (mass change of the order of 10 mg) and perform studies on transmutations with fast neutrons, a mass spectrometry system must be used, which is currently available at INFN-LNL. Therefore, these preliminary studies indicate that the proposed setup may be a demonstrator facility where to test the concepts and effectiveness of burn out of minor actinides in nuclear waste. Further studies are being conducted to assess in detail the capabilities of this apparatus and define its potentiality for an experimental program on lead fast systems and waste burn-out. An alternative version is also currently being studied, where the core would only contain plutonium-free actinides or spent fuel.

The realistic measurement of transmutation efficiency for industrial applications will however require, as a natural evolution, an improved system with higher power, in the MW range, and liquid lead cooling. This is one of the goals of the MYRRHA project at SCKCEN [At Abderrahim et al. (2010)]. The development of a very high power deuteron accelerator (5 MW) is also the goal of the International Fusion Materials Irradiation Facility (IFMIF) project [IFMIF (2012)], to which INFN-LNL gives an important contribution [Pepato et al. (2010)]. All these current technical developments and efforts can make full-scale ADS more realistic in the future.

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