SOME RECENT CONTRIBUTIONS OF BASIC NUCLEAR SCIENCE TO NUCLEAR WASTE TRANSMUTATION

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ABSTRACT

Nuclear waste transmutation aims at alleviating some long-term risks associated with actinides and with some long-lived fission products. Proposals of using accelerator driven system (ADS) to efficiently burn actinides in uranium free fuels have revitalised some basic researches in the field of nuclear and reactor physics. This is the case for high intensity accelerator in the ADS context and for the neutron source which relies to a large extent on basic nuclear physics related to spallation. There is also an experimental program called MUSE at Cadarache to study the sub-critical reactor physics with regard to its neutronics. A second area where basic research is involved is the measurement of new or more reliable neutron cross sections specific to transmutation and also to the thorium fuel cycle considered as a long-term option for "clean" energy production with reduced actinide production. This second area will possibly be covered by a new facility called n-TOF developed at CERN.

Keywords: Transmutation, High intensity accelerator, Spallation, Sub-critical reactor, Thorium

1- GENERAL BACKGROUND

Technically sound and socially acceptable management of the risks associated with nuclear wastes represents a major challenge nuclear energy has to face today. It is well known that almost all the radioactivity generated in the nuclear fuel cycle is concentrated in the spent fuels unloaded from reactors and represents a potential source of many types of risks for mankind.

There exist world wide two strategies to deal with these spent fuels which, for a typical burn-up of 33000 MWd/t, contain about 96 % of uranium, 1% of plutonium and 3% of fission products (FP) and minor actinides (MA: neptunium, americium and curium). The first strategy is to separate plutonium and to recycle it once in the same type of reactor (PWR), whereas uranium is put in interim storage and the 3% left (FP and MA) embedded in block of borosilicate glasses. In the second strategy, spent fuel is stored in interim storage and finally considered as the ultimate nuclear waste. In both cases, a deep geological repository is necessary for the high level wastes (vitrified or spent fuels). Concerns have raised about the future impact of these products on mankind, due to the very long lifetimes of the minor actinides (or of their daughters) and of some long lived fission products (LLFP) such as I-129, Cs-135, Tc-99 .... Indeed, MA are characterised by a very high radiotoxicity whereas some LLFP may be very mobile in the environment.

Nuclear transmutation has been considered since the 70' [1]as a way to lower these long-term risks and the associated uncertainties, by reducing the initial inventory of long-lived radionuclides to be put in geological disposal. Such reduction can only be achieved by transforming such a radionuclide to one (or more) other nuclide(s) through nuclear reactions. Photon or charged particle are ineffective in doing so for fundamental reasons related to their high non-nuclear interaction cross sections (electron pair creation for the first one, electronic stopping power for the second) with the consequence of an overall excessive energetic cost for transmutation Because it carries no electric charge, neutron is the only particle able to efficiently induce transmutation. Moreover, several moles of neutrons may be yearly available in nuclear reactors to transmute weighable quantities of long-lived products. This can be achieved by cumulative fissions for actinides and by capture for LLFP, ending up to stable nuclei within reasonable "historical" time span.

Many disciplines are concerned by transmutation techniques, which include the process itself and the associated nuclear fuel cycle: branches of nuclear science (reactor physics, nuclear physics and chemistry), material science as well as engineering. Transmutation has essentially been contemplated in critical reactors, especially in fast neutron one because their good neutron economy is in favour of transmutation. In this case, actinides are added up to few % to the standard U-Pu mixed oxide fuel or introduced separately in dedicated targets at a much higher concentration. In that case, the major needs in research concern measurements of transmutation yields and material behaviour of these new types of fuels. This very important program is pursuing since many years with different irradiation reactors such as for example Phenix and Osiris in France or HFR in Petten (The Netherlands). There are also important computer code calculations to simulate various impacts (safety, cycle lengths, fuel cycle, resources needed, waste production ...) of introducing such new products in a reactor. Finally, one has to carry out system and strategic studies to assess the gain achieved in term of risk reduction, especially in the long-term, and the various ways to implement transmutation on an industrial scale.

The renew of interest for transmutation since the end of the eighties has revitalised some basic nuclear researches, and we will concentrate in this paper on those specially related to nuclear and reactor physics. They can be divided in two categories.

The first one is related to the different steps (conceptual, demonstration ...) of designing a transmutation system. We will concentrate here on the so-called accelerator driven system (ADS), because of its strong links with nuclear and reactor physics. It is worth pointing out at this stage that other important innovative transmutation systems, based on critical reactors, are also proposed. But they raise specific issues in other areas and will therefore not be discussed in this paper. These areas are mainly chemistry or material science for molten salt reactor[2,3], or fuel technology for high temperature reactor[4].

The second area, largely independent from the transmutation device, concerns the need of reliable nuclear data for the various elements either to be transmuted (minor actinides, some long lived fission products), or entering in the composition of innovative fuel as matrix (e.g. thorium) or related to thorium fuel cycle. Indeed, thorium might become an attractive option for "clean energy" production due to a strong reduction of actinide production[5].

2- BASIC RESEARCHES IN RELATION WITH ADS.

There has been since the end of the eighties some new proposals of using a subcritical reactor either to produce energy[6] or as a way to efficiently transmute nuclear wastes[7]. Presently, although they are usually poorly funded, transmutation activities are world wide focusing essentially on ADS. This is for example the case for the P&T (Partitioning and Transmutation) program of the Key Action Fission of the 5th Frame Work Program of EURATOM[8]. A significative effort is now devoted in the 5th FWP to the preliminary design of an experimental ADS (PDS-XADS, 12 Meuros, 28 partners). The recent DOE project AAA (Advanced Accelerator Applications) is aiming at building within 10 years an Accelerator Driven Test (ADT) with an initial funding of 68 M$ for the FY-2001, based on proven technologies[9].

Such a sub-critical reactor needs to be driven by an intense external neutron source. This is practically achieved with an high-power accelerator (typically 1 to few 10 MW) which delivers an intense proton beam on a neutron spallation target, made of a large block of heavy metal (lead, lead-bismuth, tungsten).

Reasons for such proposals are multiple. In the beginning of the 90s, some physicists like C. Rubbia advocated sub-criticality as an attractive feature to produce nuclear electricity in a much safer way than in the present commercial critical reactors, likely to increase social acceptability with regard to reactor accident. Moreover, the use of thorium based fuels in such reactors added a second positive point with regard to proliferation and long lived nuclear wastes issues.

Since then, the arguments about ADS have largely evolved towards a more factual and scientific approach. As far as pure energy production is concerned, ADS is not considered as adding an important advantage with respect to present reactors in terms of safety. At the very most, one could perhaps take advantage from sub-criticality to produce energy within a more efficient breeding mode (this might be the case for a reactor based on a thorium fuel cycle). In fact, there is a consensus that ADS can play an important role for transmutation by giving more flexibility than critical reactors do. In terms of transmutation efficiency, a reactor loaded with uranium free fuels is obviously the best solution if one wants to avoid any actinide production. In this case, sub-criticality can compensate the overall safety degradation due to such efficient fuels (low proportion of delayed neutrons, decrease of the various safety coefficients). Because of the neutrons injected in a sub-critical reactor, the extra neutron availability can increase, allowing to transmute net neutron consumers nuclides like PFVL. This feature may be particularly interesting for thermal reactors. Finally, it should be possible to extend irradiation cycle by increasing the beam intensity in order to compensate the reactivity drop.

The design and the operation of an ADS have to be accompanied by researches on each of the 3 components: accelerator, spallation target and sub-critical reactor. We will review the lines of research and the main results by focusing on certain points.

2.1- The accelerator

High power linear proton accelerators in the region of 1000 MeV and in the range of about few tens of mA can in principle now be constructed. Present projects of powerful spallation neutron source use such accelerators. This is the case for the 2.65 MW super-conducting linac for SNS which is now under construction at at Oak Ridge[10] or the ESS project[11]. They are special requirements related to their coupling with a sub-critical reactor: very high duty cycle, minimisation of beam losses (maintenance) and of beam trips (reactor safety and long-term behaviour), highly reliable (if possible passive) devices to interrupt the beam if the reactor power (or its criticality level) increases. Obviously, such accelerator must reach a level of availability and reliability much higher than what has been achieved for fundamental researches. Because cyclotron have limited capacity (less than 10 mA) to produce intense beams, most of the projects, as mentioned above, consider a linear accelerator (usually supra).

Studies are carried out on high intensity injector, on optical (RFQ) and accelerating elements (DTL, elliptical superconducting cavities). They get the benefits of the various developments around big accelerators such as CEBAF[12] or KEK[13]. In France, the injector IPHI project[14] aims at producing 100 mA, 10 MeV proton beam. Recent measurements have shown a very high beam availability (99.96% during 103 hours with one beam trip only). Similar studies are carried out in Italy in the frame of the TRASCO project[15]

2.2- The neutron spallation source

The neutron spallation source needs a good description of all the nuclear phenomena which take place inside such a large heavy metal target (typically a cylinder of 60 cm length and 15-20 cm diameter). An accurate knowledge of the characteristics of the neutron produced (multiplicity e.g. the total number of neutrons produced per incident proton, energy and angular distribution) and of the nuclides generated inside the spallation target is obviously needed for any design. For example to determine the beam requirements for a certain reactor power level, for radioprotection set-up or to gather valuable data for thermal and material studies concerning the spallation target and the window which isolates the target from the accelerator.

Experimental researches on spallation have started in the 60' at intermediate and high energy and theoretical models developed since the pioneer work of H. W. Bertini[16]. This last decade, new type of data have been obtained, making the design of such spallation target much more reliable.

2.2.1.- Integral measurements of the multiplicity (n/p) versus beam energy and on various material are of crucial importance to calculate the reactor thermal power output for a certain beam intensity and sub-criticality level. Since about 10 years, extensive and accurate measurements have been carried out using large 4π neutrons detectors, such as ORION, with proton accelerator at CERN, COSY and SATURNE. The synthesis of this work is given in ref [17] and shown in Fig. 1.

2.2.2.- The results of differential neutron measurements (in energy and in emission angle) produced during the interaction of a proton with a thin heavy metal target (e.g. Pb, Fe, W) are essentially needed to improve [18] the INC nuclear models and therefore the reliability of the computing codes used for an engineer design. These INC models describe the way nucleons are ejected from a nucleus when hit by a high energy particle such as the incident proton. The most complete study has been performed at Saclay using the SATURNE accelerator before it was shut down by the end of 1997. Results with various beam energies (proton 0.8, 1.2 and 1.6 GeV) on different thin targets (Th, Pb, W, Fe, Al) have been obtained [19]. The possibility of direct applications engineering oriented, of these basic data, are discussed in ref [20].

2.2.3.- The same type of measurements have also been carried out for thick targets [21]in order to improve transport codes on one hand and to directly exploit the emitted neutron characteristics for radioprotection or material damage evaluation.

2.2.4.- One important issue which has been raised, specially within the nuclear data activities of the OCDE/NEA, is the validity of the intra-nuclear cascade model during the transport process when the particles are loosing more and

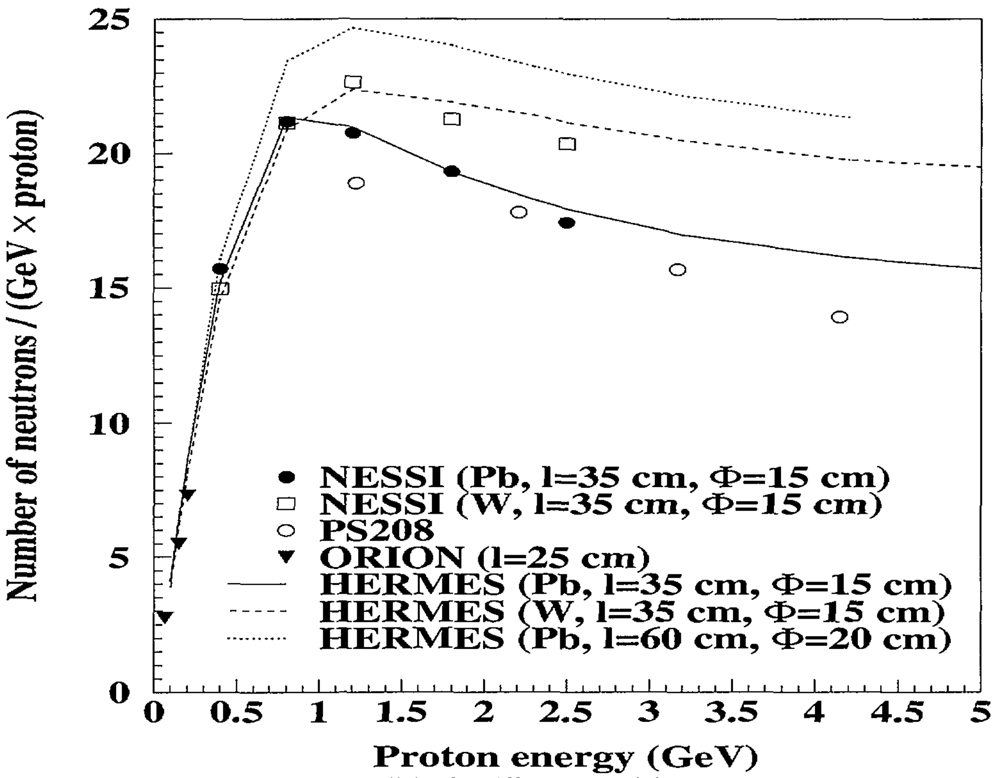

Figure 1: Neutron multiplicity for different materials and proton energy

more energy. Normally, the various cross sections are generated inside such transport codes (HETC, LAHET) using INC model until the energy raises a value as low as 20 MeV. Below, transport codes such as MCNP utilize evaluated cross-sections taken from a nuclear data file, like ENDF-B6 or JEF-2, which have been developed for the needs of fission critical reactors and fusion. A model working at 1000 MeV is likely to be inadequate at 20 MeV. Consequently, there has been propositions [22] to extend the 20 MeV upper limit of the present libraries to 150-200 MeV, where INC works. This can be done by deriving evaluated cross-sections from nuclear reactions and optical models which need to be constrained by some inclusive experimental data.

Such experimental programme has been initiated in different European laboratories (France, Belgium, The Netherlands and Sweden), using the cyclotrons CYCLONE (Louvain-la-Neuve), TSL (Uppsala) and AGOR (Groningen). These measurements are part of the HINDAS programme supported in the 5th Euratom FWP [23,24] and a synthesis of the programme is given in Tab. 1.

2.2.5.- An other important activity related to the spallation target is the identification of the isotopic composition of the spallation residues created inside the target during its irradiation by the proton beam. The standard method based on high energy resolution γ spectroscopy is limited to decay products and often hampered by the complexity of the γ spectra to be analysed. Developed at GSI with the FRS facility, the inverse kinematics method is a very powerful tool to overcome these drawbacks, although it is limited to thin targets. Therefore its main application lies in the improvement of INC models to predict the residues isotopic composition. There is one important exception where the results of such experiment can directly been used. That is the direct identification of the residues produced by the proton beam in the window which isolates the spallation target from the accelerator, with the inverse kinematics reaction Fe(1 GeV/A) on a H_2 target. These direct information's are very relevant to study corrosion effects due to the contact with the liquid target (lead or lead-bismuth) which is strongly dependent on small impurities (such as Ca or S). The inverse kinematics method is presented in this conference [25] and will not be furthermore discussed in this paper.

Table 1: Inclusive nuclear reactions (20-200 MeV) programme in the European Community					
Incident Particle	Energy (MeV)	Target	Detected particles	Status and date	Accelerator
n	30-65	^{nat}Pb	p, d, t, α	completed 1998	CYCLONE
p	65	^{208}Pb	p, d, t, ^{3}He, α	completed 1999	CYCLONE
p	65	^{208}Pb	n	completed 1999	CYCLONE
n	100	^{208}Pb	p, d, α	completed 1999	TSL
n	100	^{56}Fe	p, d, α	completed 2000	TSL
p	135	^{208}Pb	p, d, t, ^{3}He, α	completed 2000	AGOR
p	65	^{59}Co	p, d, t, ^{3}He, α	in progress2001	CYCLONE
p	65	^{59}Co	n	in progress2001	CYCLONE
p	65	^{238}U	p, d, t, ^{3}He, α	in progress2001	CYCLONE
p	65	^{238}U	n	in progress2001	CYCLONE
n	100	^{238}U	p, d, α	in progress2001	TSL
n	100	^{208}Pb	n	in project 2002	TSL
p	135	^{59}Co	p, d, t, ^{3}He, α	in project 2002	AGOR
p	135	^{238}U	p, d, t, ^{3}He, α	in project 2002	AGOR

2.3- Physics and kinetics of a sub-critical medium

It is important to extend the present computer codes used for critical reactors to sub-critical reactor. This is the aim of an important program called MUSE initiated at Cadarache with the mock up reactor MASURKA set in a sub-critical configuration. The external neutron source is provided by short pulsed 14 MeV neutron (d,t) generator developed at CNRS and called GENEPI. Because such a neutron source is intense and pulsed, multiplication factor k and kinetic behaviour can be studied with a low neutron noise background. After preliminary tests with a californium neutron source and a commercial neutron source[26], the MUSE-4 experiments are expected to start in autumn this year using MOX fuel configuration with sodium. The perspective of MUSE experiments is to study other configurations with new types of fuels (fertile free, thorium based fuels) or with new unusual coolants (lead based alloys, gas).

3.- NUCLEAR DATA ACTIVITIES

This paragraph refers to nuclear data specific to transmutation needed for critical or sub-critical reactors, that is to say below 20 MeV. Most of the nuclear data activities focus on neutron induced fission and capture cross-section measurements, which are the prominent nuclear reactions in this energy region. Three categories of nuclides are studied in this frame. First the minor actinides produced in the present reactors loaded with uranium or plutonium based fuels: Np-237, Am-241, Am-243, Cm-243 and Cm-244. Because some PFVL are also considered for transmutation, neutron capture measurements are undertaken for I-129, Tc-99. Finally, one has also to consider some new measurements for thorium fuel cycle related actinides such as Th-232, Pa-231, Pa-233 and U-233.

Often, such experimental data are either lacking or old and poorly reliable, although they may appear in nuclear data libraries as evaluated data. One must therefore be cautious in using libraries for these nuclides for any transmutation assessment. In some instance, evaluated cross-sections may also substantially differ from one library to an other. One example of this is the Am-242(n,γ)Am-243 thermal cross-section which was measured at ILL by G. Fioni et al.[27] in order to resolve the observed discrepancy between the values given by JEF-2 (5511 barn) and ENDF-B6 (252 barn). The measured value (200 barn) is in agreement with ENDF-B6 and indicates that Am-241 could in principle be transmuted in a thermal spectrum through a direct double capture process for a flux greater than 2 10^{15} n/sec/cm^2 and a net production of neutrons.

Different experimental methods, sometimes new, are considered to carry out cross-section measurements. Monoenergetic neutrons are produced and selected by time of flight in current electron accelerators like GELINA at Geel for high resolution neutron total cross-section measurements. Recent Np-237 data have been obtained in the energy range 0.3 eV – 2 keV[28] and measurements on I-129 are underway. It is worthwhile to point out that I-129 is one of the most offending radionuclide in an accidental scenario, like a well drilling, occuring in a deep geological repository. Therefore, iodine transmutation might become necessary in case its dilution in sea from a reprocessing plant like la Hague will be no more allowed.

Because most of these nuclides are difficult to obtain and are sometimes highly radioactive (especially in the case of Cm isotopes or Pa-233), new experimental ideas have emerged such as to need only little amounts of material (in the order of few milligrams). Three new types of experimental set-up match this requirement and are able to speed up the experimental data acquisition:

- the new facility n-TOF
- the use of transfer reaction
- the use of a slowing-down spectrometer

3.1.- The n-TOF facility at CERN

It has been proposed by C. Rubbia and collaborator to build an intense neutron spallation source using the pulsed proton beam delivered by the 24 GeV CERN PS accelerator[29]. Intense pulsed neutron bursts (2 10^{16} in 4π and at each neutron pulse of 10 ns) offers the possibility to measure neutron cross sections using a time of flight (TOF) basis of 180 meter long and on a large energy interval from 1eV to 250 MeV. The testing phase of this facility at 20 GeV is now completed and has confirm these expected characteristics[30]. A fission parallel plates fission detector with localisation is now in operation and has been used for testing the facility by measuring the U-235 fission cross-sections versus neutron energy. The agreement with the ENDF-B6 library is very satisfactory as shown in Fig. 2. This fission detector is intended to be used for some minor actinides fission cross-sections measurements[31].

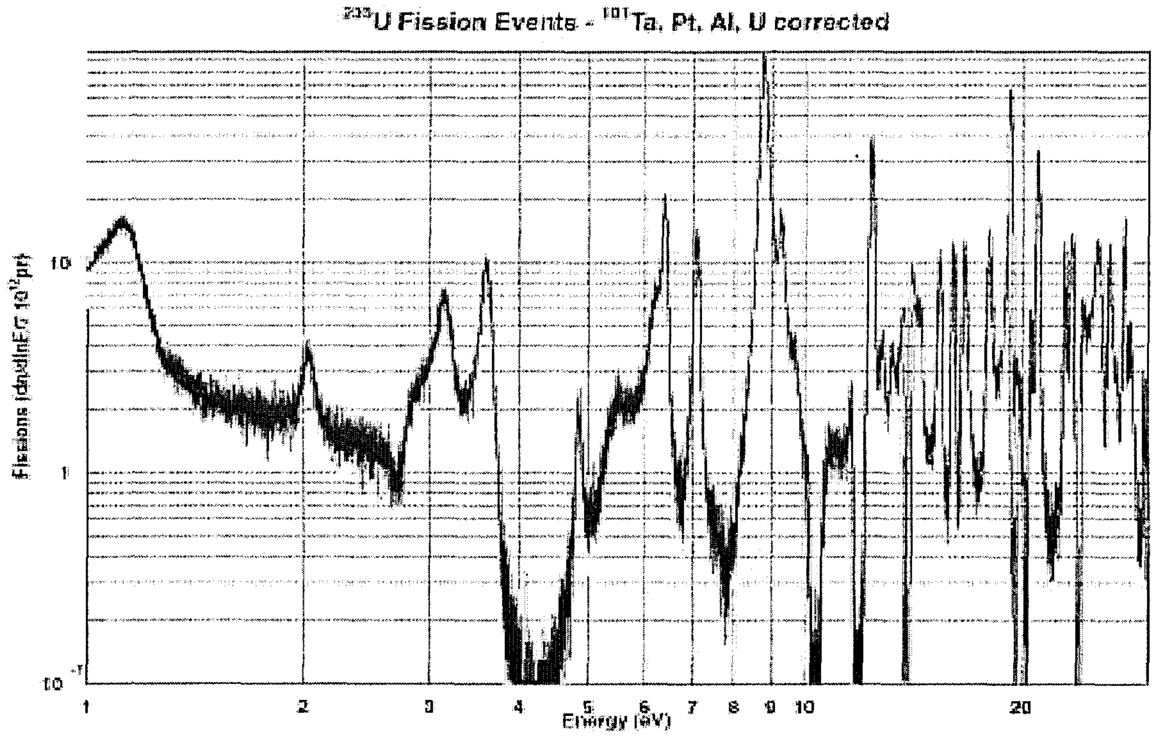

Figure 2 : U-235(n,f) versus energy; the continuous line is the ENDF-B6 cross sections

3.2.- Thorium related nuclear data

The neutron capture cross-section on Th-232 has been measured from 60 keV to 2 MeV, a region where nuclear libraries show large discrepancies. This has been done using monoenergetic neutrons produced in a two body nuclear reaction induced by protons or deuterons delivered at different energies by the 3 MV Van de Graaf electrostatic accelerator of Bordeaux (France)[32]. At lower energy, between 0.1 eV and 2 keV, measurements have been carried out with 5% energy resolution using a slowing-down time lead spectrometer associated with the neutron source GENEPI and small quantities of thorium (100 milligram). Comparisons with data base is underway[33]

A nice way to measure the Pa-233 high energy fission without having to deal with the scarcity and the high radioactivity of a Pa-233 target (27 days half life) is to select the excitation energy of the compound system Pa-234 (n+Pa-233) by measuring the energy of the proton emitted in the transfer reaction Th-232(He-3,p)Pa-234. The fission probability is then obtained by detecting all the fission events in coincidence with the proton. It is then easy to get the (n,f) cross-section by multiplying this fission probability with the compound nucleus cross-section calculated with an optical model in a reliable way[34]. The experimental results obtained between 1 and 10 MeV are in agreement with JENDL-3.

4.- CONCLUSION

From the time when the basic phenomena and the reactor theory were discovered, the nuclear physics community, including the accelerator one, became more and more separated from the reactor community. In a more general way one observes since the 50' such fissyparity between areas which use to belong to the so-called nuclear science: radiochemistry, nuclear chemistry, high energy physics. Nuclear energy became more and more a matter for engineers. It is interesting to see that the waste issue and more specifically ADS and basic new nuclear data activities gives an opportunity for the nuclear physics community to be involved with their colleagues from the reactor. No doubt that these links between basic research and nuclear energy technology and the corresponding organisations will play a role to explore and propose new ideas. This open approach may finally increase the credibility among the public opinions of the solutions proposed to deal properly with the nuclear wastes.

REFERENCE

[1] - H.C. Clairborne, ORNL-TM-3964 report, 1972
[2] - contributions 197 and 199 of J. Vergnes et al. at this conference
[3] - contributions 197 and 199 of Alekseev et al. at this conference
[4] - contribution 236 of F. Carré et al. at this conference
[5] - Thorium as a Waste management Option, EUR-19142 EN Report, 2000
[6] - F. Carminati et al., CERN/AT/93-47 (ET), 1993
[7] - C. D. Bowman et al., Nuclear Instruments and Methods, A320 (1992) 336-367, 1992
[8] - Contribution 011 of M. Hugon at this conference
[9] - Contribution 327 of G. Van Tuyle et al. at this conference
[10] - see http://www.sns.gov/
[11] - R. Wagner et al., The European Spallation Neutron Source (ESS) : status of R&D programme, Physica B 276-278(2000) 38-44
[12] - see http://www.jlab.org/
[13] - see http://www.kek.jp
[14] - J-M. Lagniel et al., "IPHI, the Saclay High-Intensity Proton Injector Project", Proc. PAC1997, Vancouver, Canada, 1997
[15] - see http://trasco.lnl.infn.it
[16] - H. W. Bertini et al., Phys. Rev. 131 (1963) 1801
[17] - A. Letourneau et al., NIM B170 (2000) 299-322
[18] - J.Cugnon, Nucl. Phys. A462 (1987) 751; J.Cugnon et al., Nucl. Phys. A620 (1997) 475
[19] - X.Ledoux et al, Physical Review Letters **82** (1999) 4412 ; S. Leray et al, to be published
[20] - T.Terrien et al., Impact of fundamental measurements on the spallation process for the design of a hybrid system, Global 99, Jackson Hole, USA, Aug-Sept 1999
[21] - S. Ménard, thesis, Orsay (France), 1998 and C.Varignon, thesis, Caen (France), 1999
[22] - A.J. Koning, ECN-C-93-005 report, Petten, January 1993

[23] - J.P.Meulders et al, High and intermediate energy nuclear data for accelerator-driven systems - the HINDAS project, Sixth OCDE/NEA Information Exchange Meeting on Actinide and Fission Product Partinioning and Transmutation, Madrid, Spain,11-13 December 2000

[24] - M.Kerveno, thesis, Nantes (France), 2000

[25] - Contribution 090 of L. Audouin et al. at this conference

[26] - Invited paper 169 of G. Alibert et al. at this conference

[27] - G. Fioni et al., Transmutation of Am-241 in a high thermal neutron flux, American Institute of Physics Conference Series 447 (1998) 43-51

[28] - V. Gressier et al., « The investigation of the total neutron cross section of Np-237 », Actinide and Fission Product Partitioning and Transmutation, Fifth International Information Exchange Meeting, Mol, Belgium, 25-27 November 1998.

[29] - C. Rubbia et al, A High Resolution Spallation Driven Facility at the CERN-PS to Measure Neutron Cross Sections in the Interval from 1 eV to 250 MeV, CERN/LHC/98-02 (EET) 1998

[30] - P. Cennini, private communication

[31] - C. Stephan, private communication

[32] - D. Karamanis et al., Neutron Capture Cross Section of Th-232 in the Energy Range from 0.06 to 2 MeV, 6th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Madrid, Spain, 11-13 December 2000 and article submitted for publication in Nuclear Science and Engineering

[33] - A. Billebaud et al., Recent Capture Cross Section Measurements on Th-232 from 0.1 eV to 20 keV and Self Shielding Effect Evaluation, 6th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Madrid, Spain, 11-13 December 2000

[34] - M. Petit et al., First determination of the neutron fission cross section for Pa-233 from 0.5 MeV to 5 MeV using the transfer reaction Th-232(He-3,p)Pa-234, XVth International Workshop on Fission Physics, Obninsk, 2-6 October 2000 and 6th OECD/NEA Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation, Madrid, Spain,11-13 December 2000