Advanced nuclear energy systems and the need of accurate nuclear data: the n_TOF project at CERN

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To satisfy the world's constantly increasing demand for energy, a suitable mix of different energy sources has to be devised. In this scenario, an important role could be played by nuclear energy, provided that major safety, waste and proliferation issues affecting current nuclear reactors are satisfactorily addressed. To this purpose, a large effort has been under way for a few years towards the development of advanced nuclear systems with the aim of closing the fuel cycle. Generation IV reactors, with full or partial waste recycling capability, accelerator driven systems, as well as new fuel cycles are the main options being investigated. The design of advanced systems requires improvements in basic nuclear data, such as cross-sections for neutron-induced reactions on actinides. In this paper, the main concepts of advanced reactor systems are described, together with the related needs of new and accurate nuclear data. The present activity in this field at the neutron facility n_TOF at CERN is discussed.

Introduction

The constant increase in the world population, as well as a global improvement in life standards, is resulting in a continuously growing demand of energy. Due in particular to the economic growth in highly populated areas like Brazil, Russia, India and China, the demand of energy is expected to raise by 50% in two decades. At present, a large fraction of the energy needed is supplied by fossil fuels, which contribute more than 85% to primary energy and 75% to electricity production.[1] It is clear that the current situation cannot be sustained much longer. The use of fossil fuels, in fact, poses severe environmental problems, due to the large production of CO_2. Furthermore, the extraction of fossil fuels from natural reserves is expected to reach a peak in the near future, with a consequent increase in the final costs. It is therefore becoming urgent to find alternative safe, clean and possibly economic energy sources. An important role in the mix of energy sources for the future could be played by nuclear energy. It should be emphasized that the use of nuclear energy would make it easier to meet clean-air objectives since the

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Broader context

Nuclear fission is an important source of electricity production, and may still play an important role in the future, helping to reduce the dependence on fossil fuels and their impact on the world global climate. However, the sustainable production of energy using nuclear power relies on the development of advanced nuclear systems, whose main characteristics should be: optimized use of natural resources (particularly uranium), minimized production of radioactive waste to be disposed of in geological repositories, increased safety and reduced proliferation risks, as well as economic competitiveness. Accelerator driven systems, Generation IV fast reactors and new nuclear fuel cycles are some of the most promising options now being investigated.
The development of these advanced nuclear technologies requires high-quality and comprehensive basic nuclear data information. In particular, accurate neutron-induced reaction cross-sections are needed for actinides, fission products and structural materials. For the last few years, an intense experimental campaign has been performed at the neutron time-of-flight facility, n_TOF, currently operating at CERN, Geneva. Thanks to the exceptional characteristics of the CERN neutron beam and to state-of-the-art detection and acquisition systems, high-accuracy, high-resolution data are being collected, providing a fundamental contribution to the long-term sustainability and low environmental impact of nuclear energy.

emission of CO_2, including fuel extraction and preparation, is much smaller than for fossil fuels, and comparable to renewable sources.[2] The importance of nuclear power may further increase if, together with its traditional application for electricity generation, it could be employed for large-scale hydrogen production, thus allowing the substitution of fossil fuels even in the field of transportation.

As of today, 439 nuclear power plants are operating, mostly in OECD (Organisation for Economic Co-operation and Development) countries, contributing approximately 6% to the total primary energy supply and 15% to the electricity production. Most of the currently operating reactors are Generation II reactors. Generation III reactors, with enhanced safety and efficiency, are already available while a more advanced class (the evolutionary Gen. III, or Gen III+) are expected to become available in a few years. Most of the world's currently operating nuclear power plants started operation in the 70′s and 80′s, so many of them will be decommissioned in the next few decades. Research is therefore now being carried out with the aim of developing revolutionary nuclear reactors, characterized by a high efficiency in the use of uranium resources and low production of nuclear waste, as well as enhanced safety, better economics and higher proliferation-resistance compared to current reactors.[3] R&D is required in many areas, such as the fuel cycle, fuel preparation and reprocessing, new materials for structural elements and for monitoring systems, risk assessment, *etc.* Among the necessary research, an important part concerns the improvement of basic nuclear data involved in reactor

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physics. The main concept of the new generation reactors and the rationale for the need of accurate nuclear data, in particular on cross-sections for neutron-induced reactions, are here discussed.

Current issues in nuclear energy production

Most of the presently operating nuclear reactors are based on the so-called "once-through" process, in which the U fuel, after preparation, goes through the reactor core only once. After the irradiation period the spent fuel is removed and has to be isolated from the environment (either in temporary or permanent repositories). The efficiency of the "once-through" cycle, in terms of uranium resources utilization, is quite low, because the spent fuel is still composed of more than 95% ^{238}U, non-fissioned ^{235}U, various plutonium isotopes and minor actinides (Np, Am, Cm, *etc*...) which in principle could still be exploited for energy production. The low burn-up efficiency of current generation reactors may lead to problems in the availability of U resources within 50–100 years. Other issues of current nuclear reactors are the high investment costs, long construction times, and proliferation concerns, *i.e.* the possibility to divert part of the fresh or spent fuel for military or terrorism purposes. However, the most important issue for current reactors, strongly affecting public acceptance of nuclear energy, is the production of large quantities of nuclear waste for which a suitable repository has to be found.

The problem of nuclear waste

The management of the high-level radioactive waste arising from nuclear power production is the major public environmental concern in the use of nuclear energy. Together with fission fragments, several transuranic isotopes are built up in a nuclear reactor, by a chain of neutron capture reactions and successive β-decays, starting at ^{238}U and, to a lesser extent, at ^{235}U. Many of these isotopes are α-emitters characterized by a very long lifetime. A list of the yearly production of high-level radioactive nuclear waste in a 1 GWe nuclear reactor is reported in Table 1. The most important contribution to the long-term radiation hazard comes from ^{239}Pu, whose half-life is 24110 years, from other Pu isotopes, as well as from the so-called "minor actinides" (MA), *i.e.* ^{237}Np ($t_{1/2} = 2.1 \times 10^6$ yr), ^{241}Am ($t_{1/2} = 432$ yr), ^{243}Am ($t_{1/2} = 7370$ yr) and ^{245}Cm ($t_{1/2} = 8500$ yr). Fig. 1 shows the radiotoxicity of the spent fuel as a function of time. Up to a few hundred years, the main component is due to the radioactive decay of fission fragments. Although Pu and MA represent only

Table 1 Material inventory of a 1 GWe reactor at loading and at discharge of the fuel, after one year of operation.[4]

Fuel and waste components	Initial loading mass/kg	Mass after discharge/kg
^{235}U	954	200
^{238}U	26328	25655
Long-lived Fission Fragments		63
^{239}Pu		156
Other Pu isotopes		110
Minor Actinides		20
Total mass	27282	27279

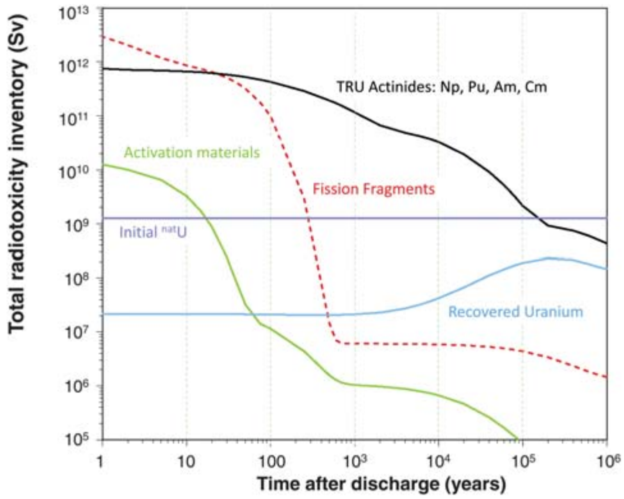

Fig. 1 Estimated radiotoxicity inventory of the spent fuel produced yearly in the world, for the various components of the nuclear waste.

1.5% of the waste volume, their radiotoxicity becomes dominant after approximately 300 years, and remains considerably high for hundreds of thousands of years, a period too long to guarantee a safe isolation from the environment by means of engineering barriers. Furthermore, actinides pose criticality and proliferation concerns. Therefore, their disposal requires deep underground geologically stable repositories. The volume of nuclear waste that needs to be stored, however, is so large that new geological sites with large storage capacity (~50000 tons of spent fuel) would need to be located and set up every few decades.

A possible solution to the problem of nuclear waste could come from transmutation processes in which long-lived radioactive isotopes are transformed into short-lived or stable ones. The most effective nuclear processes are neutron-induced reactions. In particular, long-lived fission fragments can be incinerated by means of neutron capture reactions.

For transuranic elements, *i.e.* Np, Pu, Am, and Cm, neutron-induced fission is in principle a much more efficient process, since it leads also to energy gain and a surplus of neutrons. However, as shown in Fig. 2, the fission cross-section of many actinides is characterized by thresholds of a few hundred keV, which hinders the possibility to use conventional thermal reactors for their transmutation. The possibility to incinerate the highly radiotoxic component of nuclear waste is therefore linked to the availability of nuclear systems characterized by a fast neutron spectrum (*i.e.* with energies ranging from a few keV to several MeV).

Several possibilities are being considered for nuclear waste incineration. One possibility is the use of an accelerator driven system, a device obtained by combining a sub-critical nuclear reactor with a proton or deuteron accelerator that provides, through the spallation process on a heavy material, the neutron excess necessary to sustain the chain reaction. The main advantage of such a device resides in the fact that it is intrinsically safe, and the neutron spectrum could be optimized for nuclear waste incineration. Another possibility is the use of the so-called fusion-fission transmuters, in which the intense neutron fluxes needed for transmutation are produced in fusion systems. Most of the efforts in the field of emerging nuclear technology are however now being devoted towards the development of

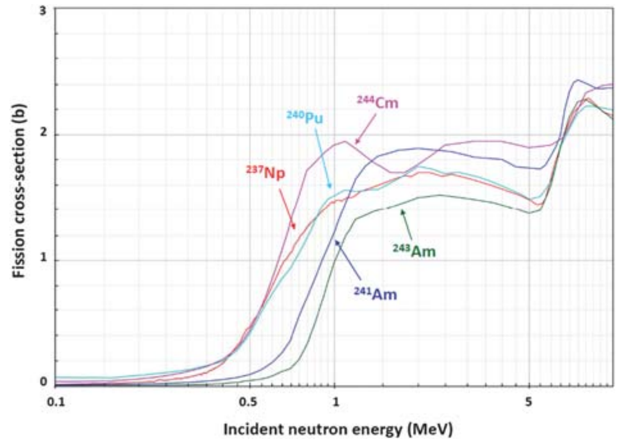

Fig. 2 Neutron-induced fission cross-section as a function of the neutron energy for long-lived minor actinides present in nuclear waste. A recycling of these isotopes in the fuel cycle can only be achieved in advanced nuclear systems with a fast neutron spectrum.

advanced critical systems based on a closed fuel cycle, in particular the so-called Generation IV (or Gen IV) nuclear reactors.[3]

Advanced nuclear systems

The main concept of Generation IV systems is based on the possibility of recycling a large fraction of the long-lived actinides, thus minimizing the final inventory of high-level nuclear waste. The use of fuel mixtures that include part of the waste would lead to the double advantage of burning highly radiotoxic isotopes while producing energy. Contrary to the once-through process, recycling would on one hand allow a reduction in the volume of nuclear waste to be permanently stored in geological sites, and on the other hand it would better exploit the uranium resources, therefore ensuring long-term sustainability of the nuclear energy option.

Together with a much more efficient use of uranium, and a smaller environmental impact of nuclear waste production due to their recycling, key objectives of Gen IV reactors are higher proliferation resistance, better economics, and enhanced safety, combined with physical protection even against acts of terrorism. Finally, future reactors should ensure a large scale production of hydrogen, in order to substitute fossil fuels in transportation. In order to meet these objectives, fast breeder reactors are needed, as well as very high temperature thermal reactors. At present, sodium-, lead- or gas-cooled systems are the best candidates for Gen IV fast nuclear reactors.

A final remark regards the possibility to develop innovative fuel cycles. In particular, a topic of great interest is the use of the Th/U fuel cycle in either critical or subcritical systems. This cycle is based on the fertile ^{232}Th isotope, which breeds the fissile ^{233}U by neutron capture and subsequent β-decay of ^{233}Th and ^{233}Pa, according to the following scheme:

$$^{232}\mathrm{Th}(n,\gamma)^{233}\,\mathrm{Th} \xrightarrow{\beta^-,\ t_{1/2}=22.3\mathrm{m}} {}^{233}\,\mathrm{Pa} \xrightarrow{\beta^-,\ t_{1/2}=27\mathrm{d}} {}^{233}\,\mathrm{U}$$

An interesting advantage in using this cycle, as compared to the conventional U/Pu cycle, is the much lower production of high-level radioactive waste, due to the absence of ^{238}U which acts as a seed for the build-up of heavy transuranic actinides in conventional reactors. As a consequence, the radiotoxicity of the spent fuel in reactors based on the Th/U fuel cycle is significantly reduced. Another advantage of this cycle is the large availability of the fuel. In fact, thorium is 3 to 4 times more abundant than uranium in the Earth's crust, and is widely distributed in nature as an easy mining resource in many geographical areas. Finally, natural thorium is entirely constituted of the fertile ^{232}Th isotope, so that there is no need for isotopic separation in the preparation of the fertile component of the fuel.

The need of nuclear data for advanced nuclear systems

In order to reduce uncertainties in the design and operation of new generation reactors, high precision data on the cross-section for neutron-induced reactions on a variety of isotopes are required, from thermal energy to several tens of MeV. In particular, a pressing need exists for new measurements on capture and fission reactions for the main isotopes involved in the Th/U fuel cycle, as well as for long-lived Pu, Np, Am and Cm isotopes. Finally, data are required for long-lived fission fragments involved in transmutation projects, and for structural materials. A "Nuclear Data High Priority Request List"[5] has been compiled by the Nuclear Energy Agency (NEA) of the OECD.

The present knowledge of neutron cross-sections is largely inadequate for new applications in the field of emerging nuclear technologies, and needs to be updated with new experimental and/or theoretical information. Sensitivity studies performed by means of Monte Carlo simulations indicate that for most long-lived fission fragments and minor actinides, the present uncertainties are much larger than needed for the reliable design and safe operation of advanced nuclear reactors. Table 2 lists the current and needed accuracy for the cross-sections of various reactions involved in the design of advanced nuclear systems. The table applies to different types of Gen IV fast reactors, as well as to ADS, and has been obtained for different core designs. The need to improve the current knowledge of nuclear data is evident.

A sensitivity analysis performed for Gen IV fast reactors, as well as for ADS and other systems for nuclear waste incineration,[6,7] has indicated the need of new nuclear data for a long list of isotopes. Capture and fission cross-sections are needed for fertile and fissile isotopes involved in the Th-cycle, in particular

Table 2 Current and needed accuracy on neutron cross-sections required for the design of Generation IV fast reactors.[6]

Isotope	Reaction	Energy range	Current accuracy (%)	Needed accuracy (%)
^{238}Pu	Fiss	0.2–1.4 MeV	17	3–5
^{239}Pu	Capt	2.0–500. keV	7–15	4–7
^{241}Pu	Fiss	450 eV–1.4 MeV	8–20	2–5
^{242}Pu	Fiss	0.5–2.2 MeV	19–21	3–5
^{241}Am	Fiss	2.2–6. MeV	9	2
^{243}Am	Fiss	0.5–6. MeV	12	3
^{244}Cm	Fiss	0.5–1.4 MeV	50	5
^{245}Cm	Fiss	67–183 keV	47	7

^{232}Th, ^{231}Pa, ^{233}U, ^{234}U and ^{236}U. Similarly, reliable experimental data on capture, fission and inelastic cross-sections in a wide energy range are required for transuranic isotopes, in particular ^{237}Np, ^{238,240,241}Pu, ^{241,243}Am and ^{244,245}Cm. Furthermore, the incineration scheme of long lived fission products requires accurate capture cross-sections for ^{79}Se, ^{99}Tc, ^{129}I, ^{135}Cs, ^{151}Sm, *etc.* Finally, data on structural materials used for reactor components or as coolant, are still far from being accurately known. New measurements on stable isotopes are therefore needed as well.

The n_TOF project

With the aim of fulfilling some of the requests of nuclear data for advanced reactor systems, as well as for collecting fresh new data of interest for nuclear astrophysics, a neutron facility was built a few years ago at CERN: n_TOF.[8] It consists of a time-of-flight installation based on a spallation neutron source. The neutron beam is produced by 20 GeV/c protons from the CERN Proton Synchrotron accelerator, impinging onto a massive Pb block, surrounded by a water layer acting both as coolant and moderator of the neutron spectrum. A scheme of the facility is shown in Fig. 3. The main features of the neutron beam are the very high instantaneous neutron flux and the wide energy spectrum, covering over 9 orders of magnitude, from thermal to approximately 1 GeV neutron energy, as shown in Fig. 4. Another important feature is the high resolution in neutron energy (reconstructed from the time-of-flight), thanks to the 200 m long flight path used in the measurements. n_TOF represent one of the two major neutron facilities in Europe, the other one being GELINA, at the European Commission Joint Research Center, Institute for Reference Materials and Measurements, (JRC-IRMM), in Geel, Belgium. An intense experimental program is carried on by an international collaboration of 120 researchers, from 40 universities and research institutes, mostly from Europe. Several detection systems and an innovative data acquisition system based on flash ADCs have been set up at n_TOF. Among them, a 4π total absorption calorimeter made of 40 BaF_2 crystals was built for measuring capture reactions, while fission measurements are carried out with a fast ionization chamber and parallel plate avalanche counters. The activity of the n_TOF collaboration has been supported by the European Commission, within the Fifth, the Sixth and currently the Seventh Framework Program for Research.

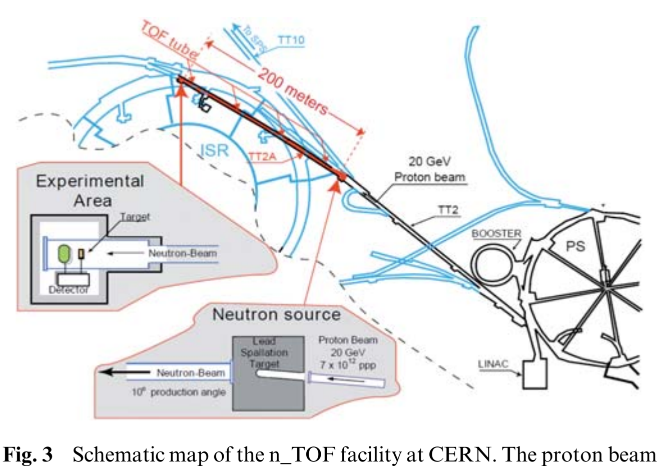

Fig. 3 Schematic map of the n_TOF facility at CERN. The proton beam from the PS accelerator complex impinges onto a massive water-cooled lead block.

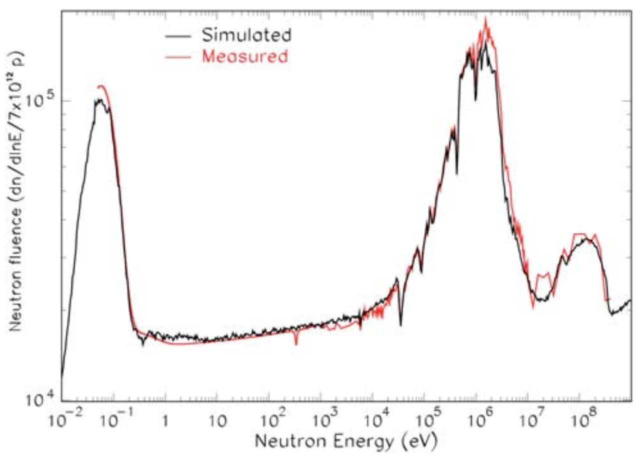

Fig. 4 Simulated and measured neutron fluence per proton pulse, available for measurements in the experimental area, at 200 m from the spallation target.

Recent results

The innovative features of the n_TOF facility, in combination with the high-performance experimental setups and state-of-the-art acquisition system allows one to collect, in some cases for the first time, accurate neutron cross-section data even on highly radioactive samples or on isotopes available only in small quantities.

In the first experimental campaign, data have been collected on several reactions of interest for the Th/U fuel cycle, for Generation IV fast reactors, and for accelerator driven systems mostly focusing on nuclear waste transmutation. A list of the cross-sections measured at n_TOF can be found in ref. 9. In most cases, the accuracy of 3 to 5%, required for the applications to emerging nuclear technologies, has been reached in the n_TOF data. Accurate capture cross-sections have been obtained in particular for ^{232}Th(n,γ)[10] and ^{233}U(n,f),[11] the two most important reactions involved in the Th/U fuel cycle. For the same application, the fission cross-sections of ^{232}Th and ^{234}U have also been measured and are now in the process of being released.

Similarly, high accuracy data have been collected on the capture cross-sections for some of the most important transuranic isotopes involved in transmutation processes, in particular ^{237}Np, ^{240}Pu and ^{243}Am.[12] Finally, accurate neutron-induced fission cross-section measurements have been performed for the minor actinides ^{241}Am, ^{243}Am and ^{245}Cm.[13] As previously mentioned, the current accuracy on these isotopes is much poorer than requested for the reliable and safe design of Generation IV nuclear energy systems. In most cases, the n_TOF results, combined with advances in nuclear theories for fission, is providing the basis for new, more reliable cross-sections. A striking example is the case of the neutron-induced fission cross-section of ^{243}Am, shown in Fig. 5. The results of various measurements performed in the past on this reaction showed discrepancies of as much as 20%, with data mostly clustered in

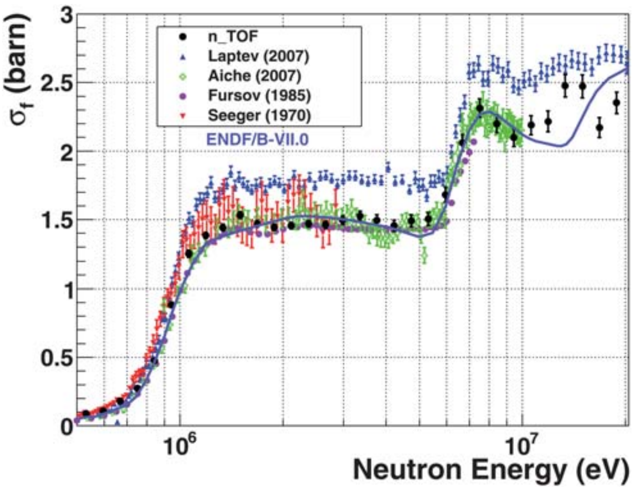

Fig. 5 Neutron-induced fission cross-section for ^{243}Am, compared with some previous measurements. The prediction of the Evaluated Nuclear Data File (ENDF-B/VII) is shown by the curve.

two groups. This situation is unacceptable for the new needs related to the development of Gen IV reactions. For this reason, a new measurement was performed at n_TOF, aiming at an overall uncertainty of approximately 3%. The results of the n_TOF measurement, represented by the black circles, confirm the current ENDF-B/VII evaluation and resolve the discrepancies between previous cross-section data. This represents an important step toward a significant improvement of actinide cross-sections for nuclear technology applications.

After the n_TOF facility had been refurbished with a completely new spallation target, with more efficient cooling and a ventilation system, the second experimental campaign has recently started. The aim of the new campaign, which is predicted to last several years, is to perform a new series of capture and fission measurements, in particular on plutonium isotopes and minor actinides, not measured so far, with high accuracy, matching the needs for Generation IV reactors. On a longer timescale, the construction of a second flight path at a shorter distance of 20 m is planned. Thanks to a much higher flux, the second experimental area will allow measurements on isotopes of very high specific activity and relatively low mass.

Conclusions

The fast growth in global energy demands, combined with the fast depletion of the exploitable reserves of fossil fuels, and the concerns on the severe environmental impact of the use of fossil fuels, in particular related to the production of CO_2 and associated climatic changes, have recently triggered a renewed interest in nuclear energy, and a reconsideration of its role in the mix of safe, clean and cost-effective energy sources of the future. At present, however, safety, proliferation, efficiency and, especially, waste management issues associated with current generation reactors, represent a limit to a further expansion in the use of this energy source. The development of new generation systems, based on a closed fuel cycle, is being pursued with the aim of overcoming major limitations of present generation reactors. In Generation IV systems, the total or partial recycling of actinides would result in a much more efficient use of the uranium resources and, more importantly, in a large reduction of the volume of long-lived nuclear waste to be stored in geological repositories.

The development of advanced nuclear reactors requires R&D in several fields. Among them, advances in the knowledge of basic nuclear data are needed. In particular, the design of fast reactors able to burn Pu isotopes and minor actinides (Np, Am and Cm), requires accurate neutron capture and fission cross-section data on these isotopes, as well as other information such as fission fragment distributions, neutron multiplicities, delayed neutron emission, *etc.* A large effort is therefore being devoted by the experimental and theoretical nuclear physics community to update the current knowledge of basic nuclear data, to meet the needs of future nuclear technologies. In this respect, an important contribution is being provided in Europe by the n_TOF project, in which high-accuracy measurements are being performed at the pulsed, white-spectrum neutron facility at CERN. Thanks to this effort by the European nuclear physics community, new and accurate data are now becoming available, which will contribute to more reliable cross-section databases for the design of advanced nuclear systems. The effort will continue in the future at the n_TOF facility, with a comprehensive experimental program already foreseen for the next few years.

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