Annals of Nuclear Energy xxx (2013) xxx–xxx

Contents lists available at SciVerse ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Nuclear data for radioactive waste management

Enrico Sartori

Consultant, Retired from OECD/NEA 15 rue Georges Pitard, 75015 Paris, France

ARTICLE INFO

Article history:
Available online xxxx

Keywords:
Nuclear data
Radioactive waste
Spent fuel

ABSTRACT

The role nuclear data plays in determining the source term of radiation emitted by spent fuel and radioactive waste arising from nuclear activities is described. The isotopes most contributing to this source for different fuel cycles are identified. Current international activities aiming at improving the existing data bases, in particular as concerns data uncertainties are addressed.

1. Introduction

Nuclear waste management is a multi-disciplinary activity involving the full fuel cycle, from mining of fissionable minerals to final disposal of spent fuel or residual radioactive materials. It concerns diverse activities such as conversion of mass to different energy forms, for example electricity and heat, or the production of radioactive sources for medical, biological, materials applications, and irradiations (diagnostics, therapy, sterilization, etc.). The term nuclear waste is ambiguous: its meaning depends on ideological viewpoints – what for some is waste is for others a resource, in particular what concerns spent nuclear fuel. Most current fuel cycles exploit the energy potential of the order of a few percents only, the rest is often classified as waste although it still has a high energetic potential. It is a by-product of energy production/conversion both in fission and fusion systems, in medical and different industrial applications.

Nuclear waste needs to be disposed of in ways that ensure safe isolation from the contact with the biosphere for periods long enough to have it decayed to reach negligible effects on biological systems. Several paths are available, depending on policies in use in the different countries: it can go from direct disposal into deep geological repositories, transmutation through single or multiple recycling of spent fuel in closed fuel cycles and transmutation through specifically designed facilities such as accelerator driven systems (ADS). Final geological disposal will always be required; different fuel cycle options aim at the overall reduction of the amount to be stored away from the biosphere.

Waste management activities have to follow strict licensing procedures, all of which require predictive modelling of the performance of the system to ensure compliance with current regulation. Such models are based on algorithms describing the various physical and chemical phenomena occurring in the different processes, covering in some cases very long times into the future.

Human build predictive models are approximations that consider only the most relevant aspects for a given objective. In addition, waste management modelling involves multi-physics and multi-scales: it implicates diverse disciplines such as rock mechanics, fluid flow in the aquifer, heat transfer, chemical thermodynamics, nuclear and radiation physics, nuclear heat and decay, dosimetry, surface transportation as well as atmospheric transportation and a full modelling may even consider climate change such as future glaciations in the area of the final repository.

In all this, the most important component is the radiation source. The radioactive source of the waste has to be characterised with good precision, as its quantity will affect the modelling results throughout. In our practical applications the source is human made and therefore its characterisation is possible with relatively small uncertainty. Other parameters of a waste management system may have relatively high uncertainties, especially those concerning parts that are not human made: e.g. characterisation of the underground rocks, speed of flow of the aquifer, etc. In these cases the term variability rather than uncertainty is often used.

In order to quantify the source term, both measurements and predictive modelling are used. When modelling is carried out for practical applications, normally macroscopic phenomena are predicted that concern a full system or part of it (e.g. waste repository). In order to predict macroscopic phenomena there is a need to take into account the underlying microscopic phenomena that together result in macroscopic effects. The equations that predict such macroscopic phenomena are for instance the Boltzmann or transport equation in the case of radiation; the microscopic phenomena are in this case described by the nuclear data (cross-sections, decay-constants, yields, etc.).

The nuclear data themselves are derived through an evaluation process involving differential measurements of basic particle interaction processes and theory (Schrödinger or Dirac equations, sta-

E-mail address: esartori@noos.fr

0306-4549/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.anucene.2013.02.003

tistical models, pre-equilibrium and intra-nuclear cascade models). At each scale, different physical equations dominate the behaviour; in this case at microscopic level e.g. the Schrödinger equation, at macroscopic level the Boltzmann equation. These equations describe the average behaviour of a population of particles and it is assumed that the average, or expectation value, is the most significant value to describe the population. This is not generally true, only when the distributions of values follow a Gaussian or normal distribution. Gaussian distributions have nothing to do with physics, they are derived from statistics through the central limit theorem or in information theory, by assuming a distribution with the highest entropy or with minimal information. This is a very widely used (and sometimes abused) distribution as it is very convenient and requires only carrying along a minimal amount of information in the uncertainty propagation of different modelling parameters. In a general waste repository system simulation procedure, many parameters used cannot be assumed as having normal distributions. For such parameters specific uncertainty analysis methods are required. As to the basic nuclear cross-section data, we assume that a normal distribution of their values is justified and that the first (average) and second moment of the distributions (standard deviations and correlations) are adequate for our practical purposes.

Nuclear waste management involves many different technical questions, but above all the most difficult aspect concerns public opinion, acceptance, siting, planning and overseeing the implementation of repositories.

This article is an overview of the nuclear data required to correctly predicting the source term of radioactive wastes, the radiation doses in the different activities of manufacturing, production, handling, transport, recycling, transmuting, and storing of radioactive/or fissionable materials. It is based on information generally available to the public.

2. Nuclear data

Nuclear data have been developed to take care of the microscopic phenomena of the multi-scale features of radiation interaction and transport. This removes the need to solve for each interaction the equations at the microscopic level (i.e. Schrödinger equation, etc.) and to concentrate on the interaction with engineering sized materials. This methodology was developed in the early days of nuclear physics, when computers were very slow and had only small storage capabilities compared to today. Modern simulation codes covering a wide range of interaction energy use a hybrid system: for very high particle energies cross-sections are computed on the fly using approximate nuclear models and for the lower energy part (normally below the threshold of pion emissions ($\pi^{0,\pm}$ - $\approx$ 150 MeV)) cross-section libraries are used. This is particularly the case for energies below 20 MeV. At lower energies the cross sections cover the unresolved and resolved resonance energy and thermal range, for which nuclear models are not capable of predicting cross-section values, but where these must be derived essentially from experimental measurement and interpolated with the theories of resonances.

A typical procedure for nuclear data evaluation is shown in Fig. 1:

Figs. 2 and 3 show for the most important plutonium isotopes both the evaluated neutron capture and the neutron induced fission cross-sections, both producing other radioactive isotopes. The different energy regions, requiring different methods in evaluating and interpolating data are shown. For isotopes most important for applications such data are derived from relative measurements and through interpretation from resonance theory (Hwang, 2010). In the higher energy part, called also the continuum, cross-sections display a rather smooth behaviour as a function of energy. That part is also based on experimental values and their interpretation through nuclear models (Index of Nuclear Models Computer Programs, http://www.oecd-nea.org/dbprog/nucmod.htm). If appropriately processed, they form the input data to computations for a wide variety of nuclear science and technology applications and source term estimation for waste management in particular.

Nuclear data and other basic data used for taking care of the microscopic phenomena in the modelling come as a rule in the form of a data library and are just one component of the overall simulation system. Fig. 4 shows the interaction of the different components in a process of simulation as well as verification and validation (V&V) of the whole system including the mechanism leading to model and data improvement.

Since about half a century a series of sets of basic nuclear data have been developed, available worldwide for the benefit of research, academia and industry, namely the following:

- **CINDA**, bibliographic information about nuclear experimental data, http://www.oecd-nea.org/cinda/cindaora.cgi.
- **EXFOR**, Experimental nuclear reaction data, http://www.oecd-nea.org/dbdata/x4/.
- **ENSDF**, evaluated nuclear structure data, http://www.nndc.bnl.gov/ensdf/.
- **AMC,** Atomic mass evaluation, http://ribll.impcas.ac.cn/ame/.

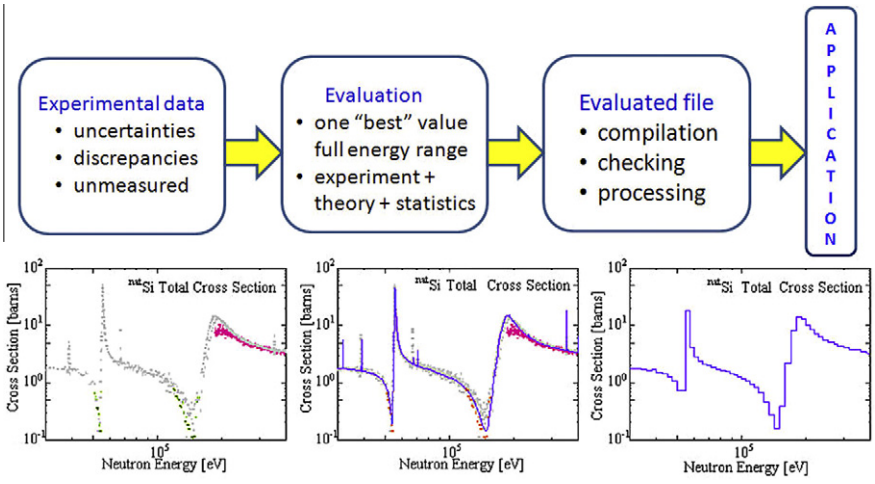


Fig. 1. Different steps in nuclear data evaluation and processing.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

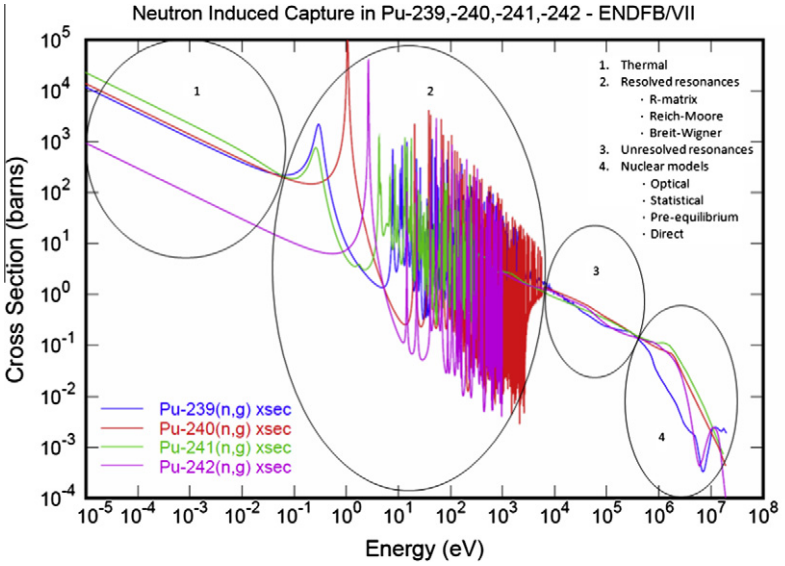


Fig. 2. Capture cross-section for different Pu isotopes.

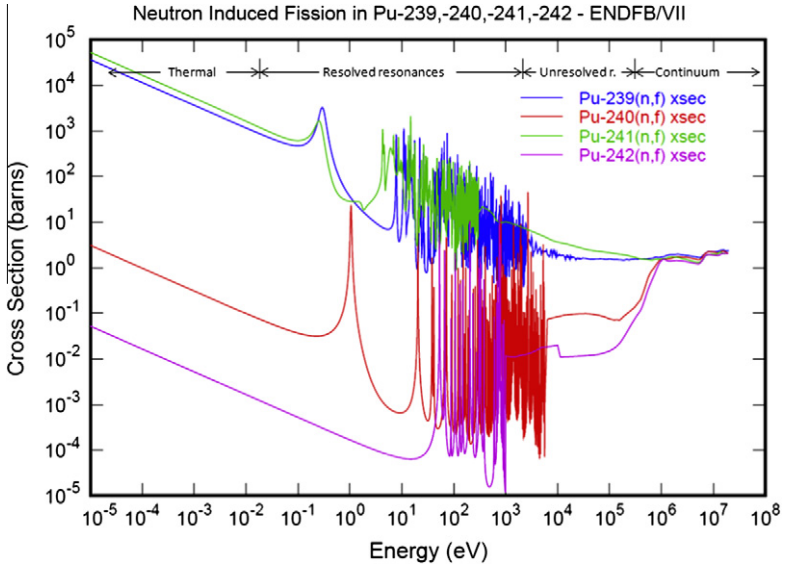


Fig. 3. Fission cross-section for different Pu isotopes.

– **RIPL,** Reference input parameter library for calculation of nuclear reactions and data evaluations, http://www-nds.iaea.org/ripl2/.
– **EVA**, Evaluated nuclear data, http://www.oecd-nea.org/dbdata/eva/.

These data-bases and the modelling codes are available from

– Nuclear Data Centres
  - OECD/NEA Data Bank Nuclear Data Service (Paris)
  - IAEA Nuclear Data Section (NDS Vienna)
  - US National Nuclear Data Center (BNL, Brookhaven)
  - Russian Center for Nuclear Data (CJD Obninsk)
  - JAEA Nuclear Data Center (Tokai-mura)
  - Nuclear and Atomic Data and Codes
  - TENDL-2011: TALYS-based Evaluated Nuclear Data Library
– Computer Code Centres
  - OECD/NEA Data Bank Computer Program Service
  - ORNL/RSICC - Radiation Safety Information Computational Center

Several evaluated nuclear data libraries have been developed over the last several decades. Those most widely used are shown in Table 1 in **bold** and can be retrieved on-line from the Nuclear Data Centres (Fig. 5).

Data evaluations available today have not all reached adequate precision for a number of advanced applications and planned developments. Further efforts are being carried out at national and international level to reduce data uncertainties where it is most critical. Table 2 lists a number of nuclides and reactions for which current precision does not yet meet the target accuracies. An international committee (Working Party on Evaluation Co-operation, http://www.oecd-nea.org/science/wpec) involving all major nuclear data producing centres meets yearly to monitor progress and to plan work for improving the most critical data. In support of this a High Priority Request List (HPRL, http://

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

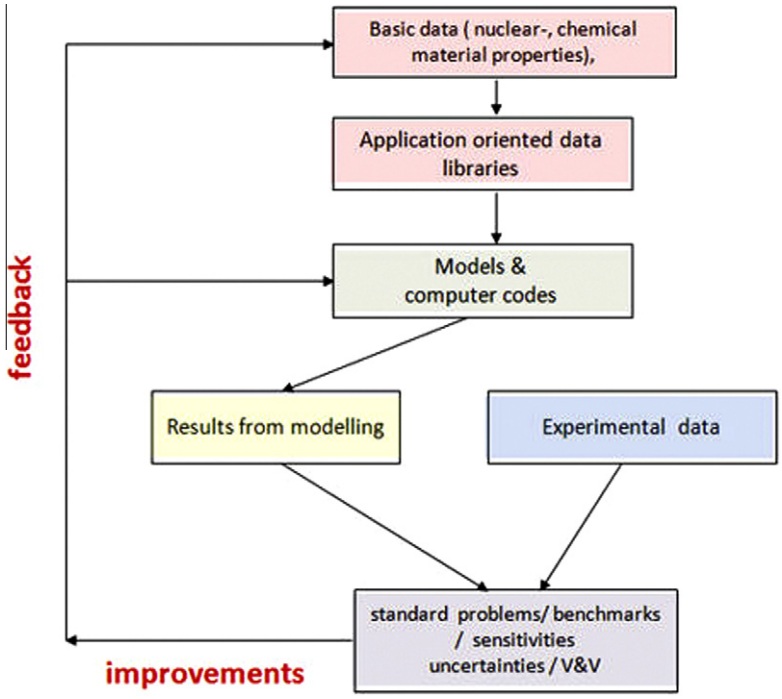


Fig. 4. Simulation, verification, validation and feedback procedure.

Table 1
Nuclear data available on-line.

BROND-2.2	BROND-2004.1	CENDL-2.0	CENDL-2.1
CENDL-3.1	EFF-2.4	ENDFB-6.2	ENDFB-6.3
ENDFB-6.4	ENDFB-6.5	ENDFB-6.6	ENDFB-6.7
ENDFB-6.8	**ENDFB-7.1**	ENDFHE-6.2	ENDFHE-6.4
FENDL-2.1	IRDF-2002.0	IRDF-90.2	JEF-2.2
JEFF-3.0	JEFF-3.1	**JEFF-3.11**	JENDL-2004.1
JENDL-2005.0	JENDL-2007.0	JENDL-2008.1	JENDL-3.2
JENDL-3.3	**JENDL-4.0**	PADF-2007.0	STANDARDS-2006.0
UKEF-1.1	UKHEDD-2.1	UKHEDD-2.2	UKHEDD-2.4
UKHEDD-2.6	UKPADD-3.0	UKPADD-6.1	UKPADD-6.10
UKPADD-6.5	UKPADD-6.6	UKPADD-6.8	UKPADD-6.9
TENDL-2011			

www.oecd-nea.org/dbdata/hprl/) is maintained, guiding work for measurement nuclear theory and evaluation programmes.

In order to facilitate users the interpretation of nuclear data, user-friendly software has been developed for their visualisation and manipulation. Several exist and can be used on line (e.g. JANIS, http://www.oecd-nea.org/janis/).

The most widely used computer code for interpreting and processing evaluated nuclear data libraries is NJOY (McFarlane and Muir, 1994). It is a comprehensive nuclear data processing system designed to convert physics data in the Evaluated Nuclear Data Files (ENDF) (McFarlane, 1998a) format into forms useful for nuclear applications (Fig. 6). It is used to prepare data, in particular cross-sections, for particle transport codes using both deterministic multi-group and continuous energy Monte Carlo methods. The code also makes some physics additions to the basic ENDF data, including radiation heating and damage, gas production, and thermal scattering from liquid and solid moderator materials. NJOY is also useful for supporting data work, both experimental

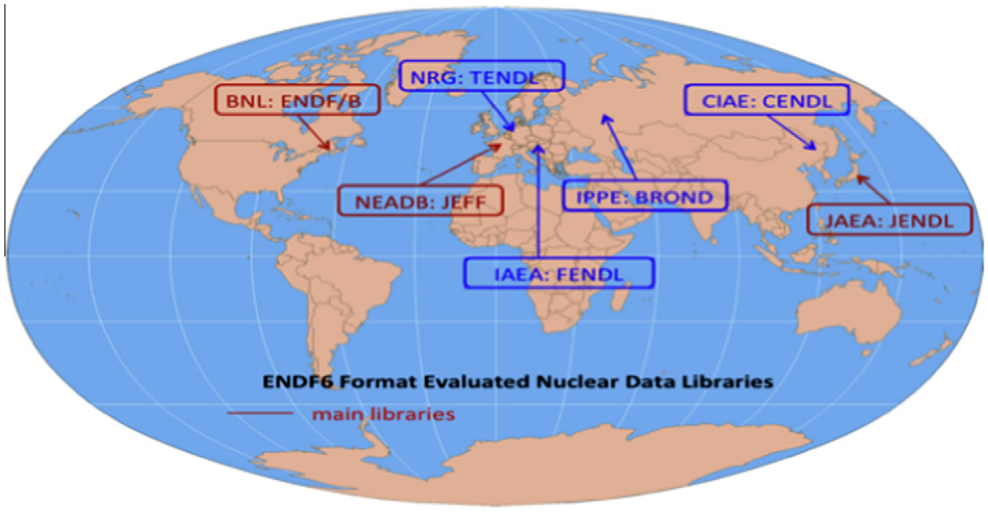


Fig. 5. Nuclear data centres.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

Table 2
Example of available precision today and required precision.

Nuclides	Reactions	Energy region	Currant accuracy (%)	Target accuracy (%)
^{238}U	Inelastic	0.498–6.07 MeV	10–20	2–3
	Capture	2.04–24.8 keV	3–9	1.5–2
^{241}Pu	Fission	454 eV–1.35 MeV	8–20	2–3 or 5–8
^{239}Pu	Capture	2.04–498 keV	7–15	4–7
^{240}Pu	Fission	0.498–1.35 MeV	6	15–2
	Number of neutrons per fission	0.498–1.35 MeV	4	1–3

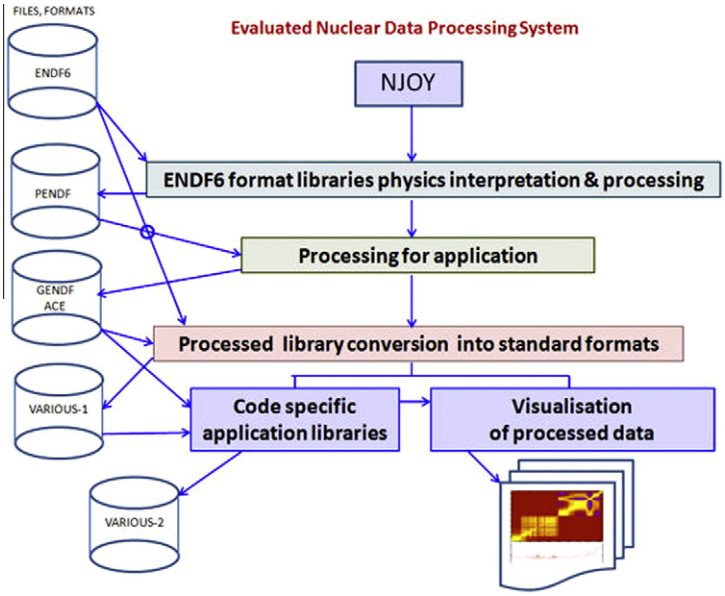


Fig. 6. The NJOY evaluated nuclear data processing system.

and in evaluation, through the analysis of experiments, display, and sensitivity studies.

3. Data required for determining the source term (McFarlane, 1998b)

The basic data and tools required for estimating the radioactive source intensity are essentially the following:

1. Activation cross sections library
   - Good capture cross sections for about 200 important fission-product nuclides (^{31}Ga–^{71}Lu).
   - Capture, fission, and higher energy threshold reactions for a large number of actinide isotopes.
   - For accelerators all reaction channels are needed, for all targets with significant lifetimes, for energies up to ~150 MeV (threshold for pion $\pi^{\pm,0}$ formation), for both neutrons and protons. This involves more than 10 000 important reactions, many of which have never been measured.
2. Fission product yields library
   The two fission fragments are normally not equal in size. The result is a distribution of possible fission fragments known as a fission-product yield distribution, which has two peaks (Fig. 7). These distributions are very important for the operation of fission power reactors; they affect parasitic absorption, on-power decay heat, decay heat after shutdown or emergency scram, and delayed neutrons (which are important factors for reactor kinetics and control). Because of their importance for both operations and safety, fission-product yield distributions are made available in computer files for reactor calculations and characterisation of spent fuel.
3. Decay data library
   - The lifetimes and modes of decay of radioactive nuclei have been studied intensively over the history of nuclear physics. Their evaluations provide a consistent set of "best" data for each nuclide, stored in the Evaluated Nuclear Structure Data File, ENSDF.
   - The decay-data sub-libraries in ENDF6 format contain sections that describe the decay modes, mode lifetimes, decay energies, and spectra of decay products (neutrons, α, β^+, β^-, γ, and X rays). Table 3 shows at the bottom left the long lived natural radioactive isotopes. Np has long disappeared

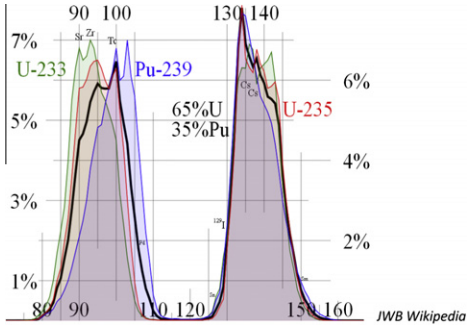
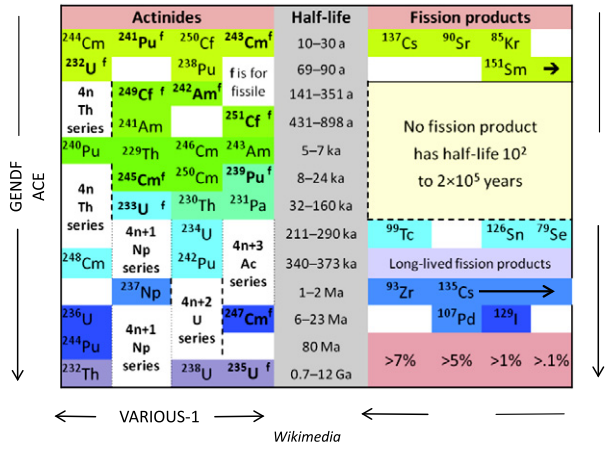


Fig. 7. Fission product yields by mass for thermal neutron fission of ^{233,235}U and ^{239}Pu.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

Table 3
The four main decay chains (thorium, neptunium, uranium, and actinium).



from the earth crust but reappears in the current industrial activities. Some of the isotopes are very short lived and disappear after a relatively short 'cooling' time, which can be engineered through isolation from the biosphere. This is the case for most fission products and some 'minor' actinides. A major issue concerns, how to manage the longer lived radionuclides.

4. Inventory codes

The inventories of radionuclides can be calculated as a function of time for a system exposed to a flux of radiation (neutrons or protons, usually) with use of the basic data just described. This calculation requires solving a set of coupled differential equations (Bateman) providing the balance between production (by a reaction, or as a fission product) and destruction (by a reaction, or by decay) of each possible nuclide. The most widely used inventory codes, http://www.oecd-nea.org/tools/abstract/list/category/d, are the following: ORIGEN, FISPACT, EASY, CINDER, CARL, ANITA.

4. Radioactive waste types

Low-level-waste (LLW) arises in the normal operation of nuclear power plants and fuel cycle facilities, as well as from the use of radioactive isotopes in medicine, industry and agriculture, the half-lives in the waste are short enough that effective disposal is achievable by deposition in supervised near-surface vaults, whilst decay takes place.

Intermediate-level-waste (ILW) is short lived (⩽30 a) and long lived (>30 a) and requires special handling to limit radiation exposures. Some forms require long-term isolation because of the long-lived radionuclides that they contain. Typical examples of ILW are filter ion-exchange resins, filter sludges, precipitates, evaporator concentrates, incinerator ash and fuel cladding.

High-level-waste (HLW) refers to the highly radioactive wastes requiring shielding and permanent isolation from the biosphere; it arises as a by product of nuclear power generation, typically the spent nuclear fuel needing long-term cooling.

The IAEA has developed a Safety Guide (IAEA, 2004) providing values of activity concentration for bulk amounts of material containing radionuclides of artificial origin, derived using the exemption concept (e.g. effective doses to individuals 10 μSv or less in a year, etc.). It lists the following 225 radionuclides + 32 isomers for clearance of LLW:

^{3}**H**, ^{7}**Be**, ^{14}**C**, ^{18}**F**, ^{22,24}**Na**, ^{31}**Si**, ^{32,33}**P**, ^{35}**S**, ^{36,38}**Cl**, ^{42,43}**K**, ^{45,47}**Ca**, ^{46,47,48}**Sc**, ^{48}**V**, ^{51}**Cr**, ^{51,52,52m,53,54,56}**Mn**, ^{52,55,59}**Fe**, ^{55,56,57,58,58m,60,60m,61,62m}**Co**, ^{59,63,65}**Ni**, ^{64}**Cu**, ^{65,69,69m}**Zn**, ^{72}**Ga**, ^{71}**Ge**, ^{73,74,76,77}**As**, ^{75}**Se**, ^{82}**Br**, ^{86}**Rb**, ^{85,85m,87m,89,90,91,92}**Sr**, ^{90,91,91m,92,93}**Y**, ^{93,95,97}**Zr**, ^{93m,94,95,97,98}**Nb**, ^{90,93,99,101}**Mo**, ^{96,96m,97,97m,99,99m}**Tc**, ^{97,103,105,106}**Ru**, ^{103m,105}**Rh**, ^{103,109}**Pd**, ^{105,110m,111}**Ag**, ^{109,115,115m}**Cd**, ^{111,113m,114m,115m}**In**, ^{113,125}**Sn**, ^{122,124,125}**Sb**, ^{123m,125m,127,127m,129,129m,131,131m,132,133,133m,134}**Te**, ^{123,125,126,129,130,131,132,133,134,135}**I**, ^{129,131,132,134,134m,135,136,137,138}**Cs**, ^{131,140}**Ba**, ^{140}**La**, ^{139,141,143,144}**Ce**, ^{142,143}**Pr**, ^{147,149}**Nd**, ^{147,149}**Pm**, ^{151,153}**Sm**, ^{152,152m,154,155}**Eu**, ^{153,159}**Gd**, ^{160}**Tb**, ^{165,166}**Dy**, ^{166}**Ho**, ^{169,171}**Er**, ^{170,171}**Tm**, ^{175}**Yb**, ^{177}**Lu**, ^{181}**Hf**, ^{182}**Ta**, ^{181,185,187}**W**, ^{186,188}**Re**, ^{185,191,191m,193}**Os**, ^{190,192,194}**Ir**, ^{191,193m,197,197m}**Pt**, ^{198,199}**Au**, ^{197,197m,203}**Hg**, ^{200,201,202,204}**Tl**, ^{203}**Pb**, ^{206,207}**Bi**, ^{203,205,207}**Po**, ^{211}**At**, ^{225,227}**Ra**, ^{226,229}**Th**, ^{230,233}**Pa**, ^{230,231,232,233,236,237,239,240}**U**, ^{237,239,240}**Np**, ^{234,235,236,237,238,239,240,241,242,243,244}**Pu**, ^{241,242,242m,243}**Am**, ^{242,243,244,245,246,247,248}**Cm**, ^{249}**Bk**, ^{246,248,249,250,251,252,253,254}**Cf**, ^{253,254,254m}**Es**, ^{254,255}**Fm**.

Doses emitted by such nuclides need thus to be determined for radiation protection and waste classification purposes; in order to

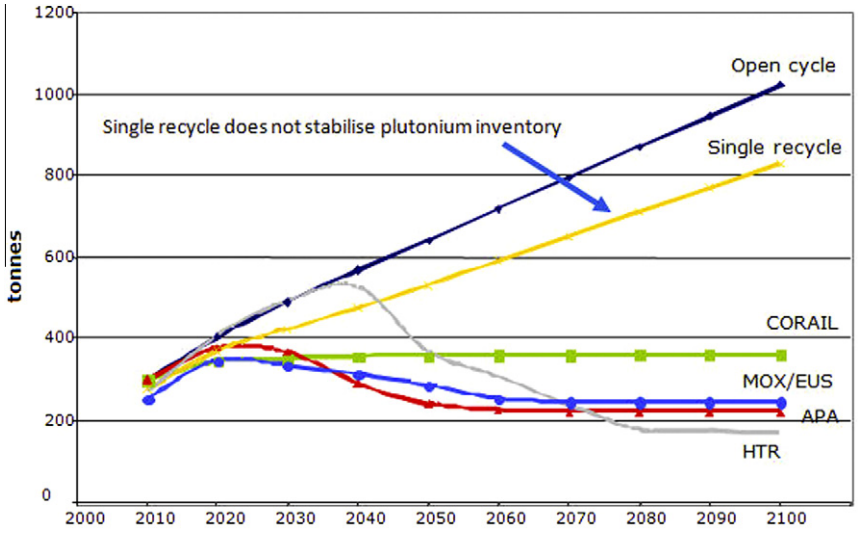


Fig. 8. Limitations of current plutonium recycling options.

Table 4
Main spallation nuclides with long half-lives in a Pb–Bi alloy.

Isotope	Half-life	Liq.-gas T^0 (°C)	Predominant radiation (MeV)
^{210}Po	0.38 a	254–962	α 5.3
^{209}Po	103 a	254–962	α 4.88
^{208}Po	2.93 a	254–962	α 5.11
^{208}Bi	0.37 Ma	271–1564	EC-γ 2.6
^{207}Bi	30 a	271–1564	β+ 0.56
^{204}Tl	3.77 a	304–1473	β− 0.76
^{203}Hg	0.13 a	−39–357	β− 0.2
^{194}Hg	1.9 a	−39–357	EC 0.4

achieve this, cross-sections for producing these nuclides through activation, fission and other nuclear reactions need to be evaluated and stored in nuclear data libraries.

5. Plutonium recycling (OECD, various) in the medium (OECD, 2003) and long term

Whatever the viewpoint, as to whether plutonium be treated as a waste or as an energy resource, the stockpile of plutonium, either accumulated in spent fuel, or extracted from spent fuel for reuse as fuel in current or future reactors, increases steadily. Several technical options have been investigated for the management of the stockpile in the medium term, in particular its stabilisation. Fig. 8 shows the performance and limitations of different options investigated for the medium term. The open cycle (UO_2) case corresponds to the uppermost (blue) curve. The total inventory of plutonium accumulates at a rate dependent on the output of the power plant park. The single-recycle MOX case also accumulates plutonium steadily, though at a lower rate; in any case it does not stabilise plutonium inventory. The other options are all able to stabilise the total plutonium inventory at various levels between 200 and 400 tonnes. This demonstrates how effective these advanced options are at preventing the steady accumulation of the total plutonium inventory. The majority of the options considered are aimed at LWRs, reflecting the present dominance of LWRs and the likelihood that it will persist over the medium term. The option of single recycle as MOX in LWRs is already soundly established on a commercial scale and presents very low technical risk to utilities (essentially the same technical risk as UO_2 fuel in a once-through cycle) and requires the minimum of future development to support. Other options, especially those involving non-oxide fuels, represent a higher technical risk, and will require extended research and development programmes to establish on a commercial scale. However, many of these more advanced options have the potential to deliver significant benefits over the single MOX recycle approach.

The major observations on plutonium recycling studies and experience are as follows:

Multiple MOX recycle in standard PWRs is practical at least as far as the second recycle generation. The highly-moderated PWR overcomes problems with void coefficients and extends multiple recycle beyond two generations. It also shows a reduction of radiotoxicity in the waste stream and higher plutonium consumption. Thermal MOX recycle, with or without multiple recycle is compatible with later transfer to fast reactors/advanced systems and for maintaining future options.

For all investigations and practical implementations of plutonium recycling leading to a reduced waste stream, good quality nuclear data libraries as those described previously are required especially those relative to major and minor actinide isotopes.

Another aspect of waste minimisation and management that has been intensively investigated is technical approaches for minor actinide burning in thermal reactors, including LWRs, HWRs, and HTRs. The implications for enrichment, fuel fabrication, in-core fuel management, fuel cycle economics and irradiated fuel and waste management were assessed in terms of kg of minor actinides destroyed per TWhe electrical output and the irradiation timescales. Minor actinide burning R&D is currently still at the conceptual stage and has yet to progress beyond theoretical studies and small-scale laboratory sample sizes. To progress to commercial scale demonstration and eventually commercial deployment will demand a large commitment. The studies carried out have required the use the latest improved cross-section libraries. A consensus view on this issue is being finalised by (OECD, 2013).

6. Transmutation using accelerator driven systems (ADS) (OECD, 2002)

Different strategies have been investigated in recent years for transmuting in an evolutionary manner nuclear waste. These include plutonium utilisation in conventional reactors and confining the minor actinides to a smaller part of the fuel cycle. New

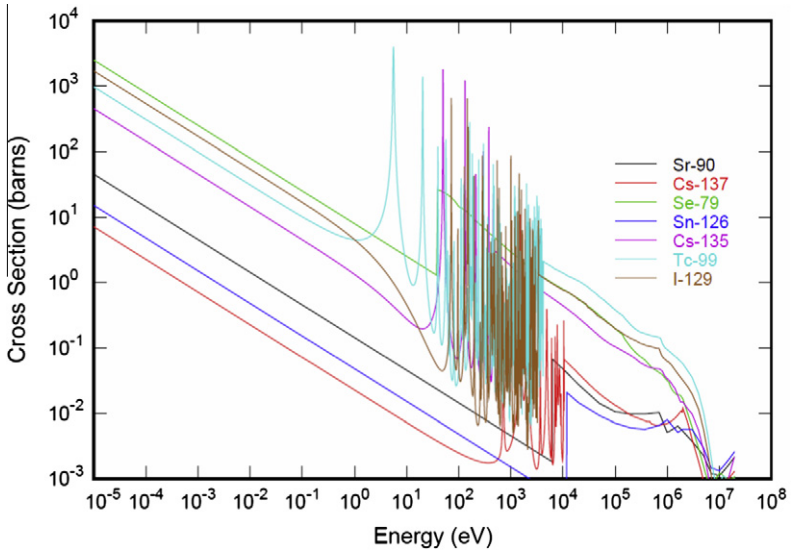


Fig. 9. Capture cross section for selected fission products.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

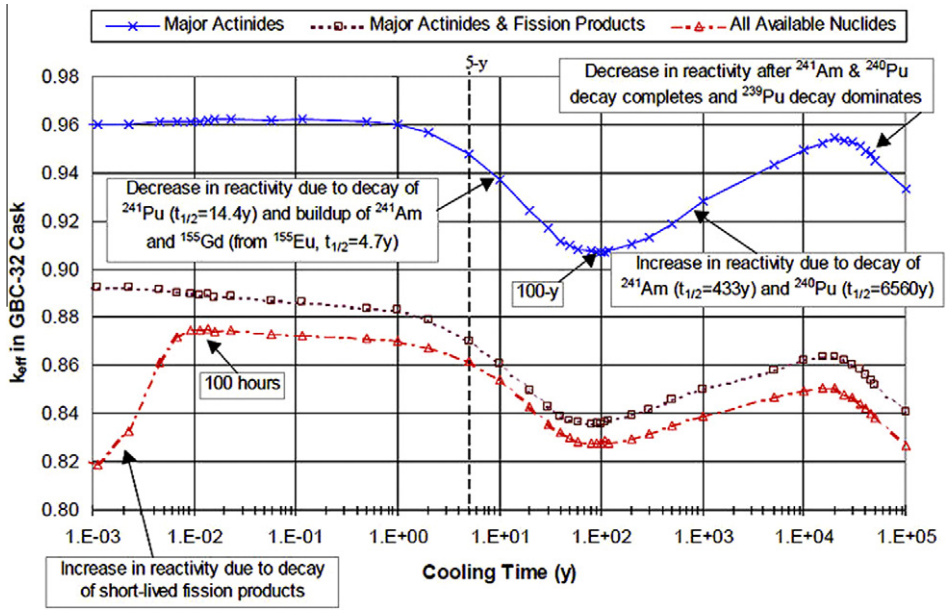


Fig. 10. Values of k_{eff} for an ∞ planar array as a function of database axial profiles for 38–42 GWd/MTU.

Table 5
Biologically significant long-lived radioisotopes in spent fuel.

Isotope	Half-life	Decay mode	Daughter product
^{90}Sr	0.07 a	β	^{90}Y
^{99}Tc	0.213 Ma	β	^{99}Ru
^{137}Cs	0.08 a	β	^{137}Ba
^{237}Np	2 .14 Ma	α	^{233}Pa
^{239}Np	0.006a	β	^{239}Pu
^{238}Pu	0.24 a	α	^{234}U
^{239}Pu	0.024 Ma	α	^{235}U
^{241}Pu	0.04 a	β	^{241}Am
^{241}Am	432 a	α	^{237}Np
^{242}Cu	0.446 a	α	^{238}Pu

transuranium isotopes burning strategies are considered in which plutonium and minor actinides are managed together to minimise the proliferation risk. These include fast reactors and accelerator driven systems (ADS). An accelerator-driven system combines a particle accelerator with a sub-critical fissionable core. The proton accelerator, delivers a continuous-wave beam with an energy around 1 GeV. High-power accelerators of several tens of MW are required for the purpose. The proton beam through interaction with a heavy-metal spallation target produces source neutrons for driving the subcritical core. The target is made of heavy metal in solid or liquid state emitting a few tens of neutrons per incident proton.

For the high energy part of protons interacting with the heavy target, cross sections, rather than extracting them from existing libraries, are produced on the fly when required at specific energies with nuclear models. Cross-section libraries for energies higher than the threshold of pion emissions ($\pi^{0,\pm} \approx 150$ MeV) are rarely produced because the number of reaction channels opening from that energy on is increasing drastically, leading to an unmanageable size of files and difficulty in maintaining them. The preferred heavy metal targets for proton projectiles are Pb or a Pb–Bi alloy. Table 4 shows the long-lived nuclides produced in the spallation process.

The neutron capture process is currently the only promising nuclear reaction for transmuting fission products. But capture removes neutrons and thus can be used in system with sufficient excess neutrons in the overall balance. The transmutation of a fission product makes sense only if the reaction rate (product of microscopic cross-section with neutron flux) is higher than the decay rate of the nuclide. With the currently achievable neutron fluxes, this condition cannot be met for the most abundant fission products ^{137}Cs and ^{90}Sr with half-lives of only about 30 years. It cannot be used either for the longer-lived ^{126}Sn. In fact these nuclides have capture cross sections that are 1–3 orders of magnitude smaller than those of other longer lived nuclides such as ^{129}I, ^{135}Cs, ^{99}Tc, and ^{79}Se (Fig. 9). In practice, only ^{129}I and ^{99}Tc can be transmuted and the radiological impact of the other fission products can be reduced only by special conditioning and confinement.

7. Spent fuel

Spent nuclear fuel is considered by some as a waste, by others as recoverable fissile and fissionable, breedable isotopes. Notwithstanding ideological or energy strategy considerations, modelling of the behaviour of these materials is a requirement for the purpose of licensing spent fuel management. It concerns such different industrial activities and safety considerations as:

- Handling (fabrication, reprocessing): criticality, radiation shielding, clearance (LLW).
- Transportation (road, rail, ship, plane): criticality, burnup-credit,[1] radiation shielding.
- Storage (wet, dry): criticality, burnup-credit, radiation shielding, decay heat.
- Repository (intermediate, geological): inventory, radiotoxicity, decay-heat, release to environment.

The investigations must ensure that such operations will not cause a criticality accident, that radiation doses to biological environment are well within permitted limits, that the hot spent fuel is properly cooled for the required length of time. The connected studies require good quality nuclear data for licensing purposes. The simulation of spent fuel behaviour requires the knowledge of

[1] Burn-up credit is a term that applies to the reduction in reactivity of burned nuclear fuel due to the change in composition during irradiation.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

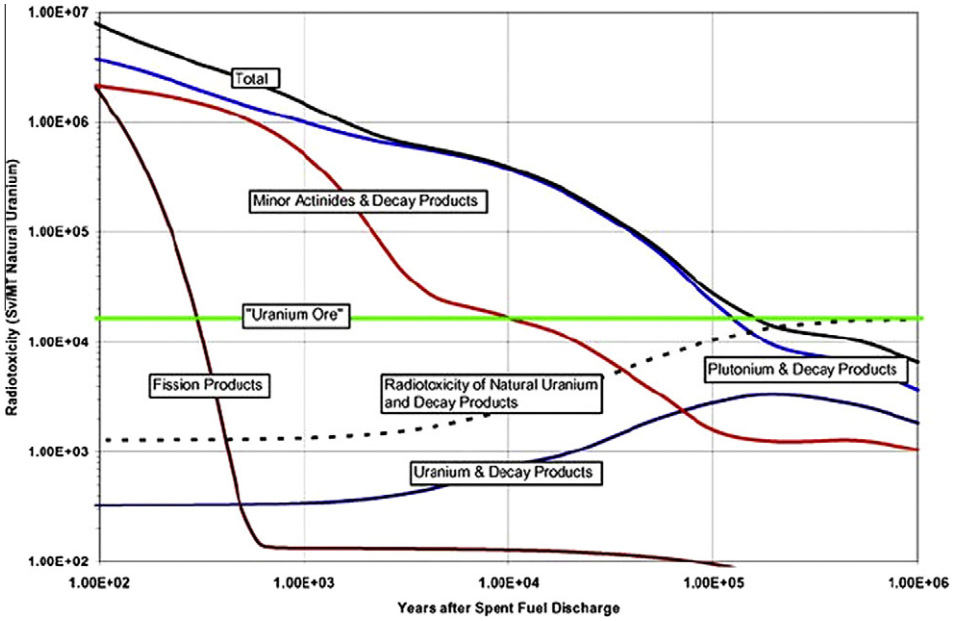


Fig. 11. Radiotoxicity of once-through spent nuclear fuel and its evolution in time (OECD, 2006).

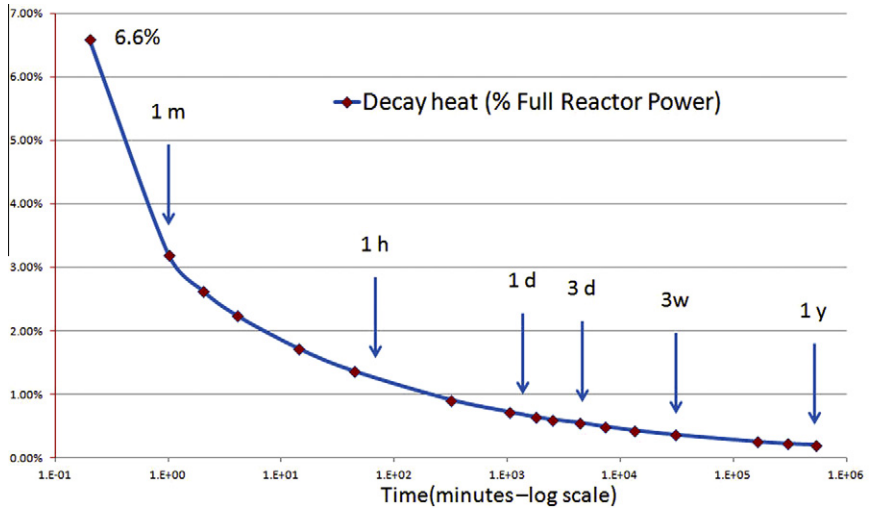


Fig. 12. Decay-heat after reactor shut-down in per-cent of full Power.

the parameters such as geometry of configuration, composition of materials and inventory of radioactive isotopes and nuclear data.

The physical quantities required for the different investigations for nuclear waste management are:

For criticality safety, waste management (HLW)

– Fission and capture cross-sections, fission yields, decay data, activation cross-sections.

Clearance

– Radio-isotope production cross sections, decay data (LLW).

Radiation shielding calculation (radiation transport, deep penetration)

– Elastic scattering cross-sections and angular distribution, neutron disappearance cross-section, nuclear structure and decay data.

Dose evaluation (linear energy transfer)

– Primary knock-on atom (PKA), displacement per atom (dpa), kinetic energy release (kerma) charged particle spectra.

The reactivity of spent fuel can change with cooling time. The key parameter for criticality safety k_{eff} is not a monotonously decreasing or constant function of cooling time, but the different contributions can increase or decrease depending on the overall isotopic composition and resulting neutron balance. The US NRC Staff Guidance 8 (ISG8) recommends that safety analyses be performed at a fixed cooling time of 5 years. Fig. 10 shows the trend for a 32-element, generic burnup-credit cask design (GBC-32). For burnup-credit criticality safety analyses performed at 5 years, increased cooling times result in an increasing conservative safety margin out to ~50 years. The additional benefit for cooling times between 50 and 100 years is insignificant (Parks et al., 2002).

The evolution of the inventory of the different isotopes (major, minor actinides, major fission products) must be estimated in a safety analysis with sufficient precision. Criticality depends both on the isotopic composition and the nuclear cross-sections and decay constants. Considerable effort is devoted internationally to improve the corresponding nuclear data and to reduce their uncertainties.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

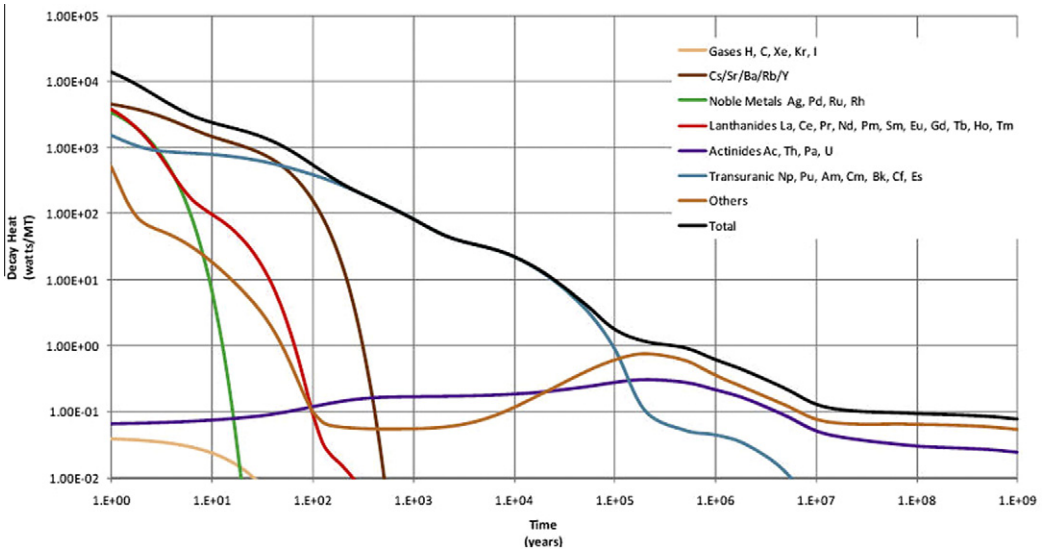


Fig. 13. PWR 60MWd/MT used fuel decay heat (Carter et al., 2011).

Table 6
Radionuclides requiring improved decay data.

Radionuclide	Half-life	Radionuclide	Half-life
35-Br-86	55.1 s	43-Tc-103	54.2 s
35-Br-87	55.65 s(β^-,n)	43-Tc-104	18.3 min
35-Br-88	16.36 s (β^-,n)	43-Tc-105	7.6 min
36-Kr-89	3.15 min	43-Tc-106	35.6 s
36-Kr-90	32.32 s	51-Sb-132	2.79 min
41-Nb-98	2.86 s	53-1-136	83.4 s
41-Nb-99	15.0 s	53-l-136 m	46.9 s
41-Nb-100	1.5 s	53-1-137	24.13 s (β^-,n)
41-Nb-101	7.1 s	54-Xe-137	3.82 min
42-Mo-103	67.5 s	54-Xe-139	39.68 s
42-Mo-105	35.6 s	54-Xe-140	13.6 s
43-Tc-102	5.28 s		

In burnup credit analyses for storage and transportation of spent nuclear fuels the following isotopes need to be taken into account

- Actinides ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu,^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am (11).
- Fission products: ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{133}Cs, ^{143}Nd, ^{145}Nd, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{153}Eu, ^{155}Gd (15).

The biologically significant long-lived radioisotopes in commercial spent fuel are shown in Table 5.

8. Radiotoxicity and decay-heat of spent fuel

One of the objectives of the nuclear fuel cycles is to eventually return to the geosphere in waste form materials with a total toxicity that is not higher than that of the ore extracted for fuel fabrication. During the power production, most of the radiotoxic materials are stored in a safe manner in the core of reactors, in spent fuel on-site storage facilities and in reprocessing plants. This objective can be met only if advanced fuel cycles with reprocessing and use of fast spectrum reactors are part of the overall scenario. A once-through fuel cycle (Fig. 11) cannot achieve this.

The radiotoxicity of the fission products dominates the total radiotoxicity during the first 100 years. Thereafter, their radiotoxicity decreases and reaches the reference level after about 300 years. The long-term radiotoxicity is solely dominated by the actinides, mainly by the plutonium and americium isotopes. For the design of more advanced fuel cycles achieving this goal, additional good quality nuclear data for actinides and fission products are required.

Quantification of decay heat induced by fission is important for the design of nuclear facilities and for the post-irradiation handling of nuclear fuels (fuel discharge, storage, transport and reprocessing, and waste handling). The decay heat decreases very quickly in percentage after shut-down of a power reactor, but the overall quantity is still high (Figs. 12 and 13).

The total decay heat (as well as quantification of the light-particle and electro-magnetic components) as a function of cooling time impacts significantly on the safety and economics of nuclear power generation. This requires comprehensive sets of nuclear data: neutron cross-sections, fission yields and decay data (primarily for fission products and actinides – half-lives, and mean light-particle and electromagnetic energies), and estimates of the uncertainties in these data. Further improvements of decay data are required for proper decay heat estimation. Calculations adopting mean decay data derived from total absorption gamma-ray spectroscopy (TAGS) measurements are able to describe the total decay heat reasonably well. Focus will be on particular fission-product radionuclides that merit such measurement in order to improve decay heat calculations without the need to resort to theory. The radionuclides that contribute the greatest uncertainty of the calculated decay heat for $^{235}U_{th}$ are short cooling times uncertainties in fission yields; half-lives of $^{98}Y^{m}$ and ^{100,102}Nb; energy releases for ^{87}Br, ^{89,97}Sr, ^{101,102}Nb, and ^{143}La.

Ill-defined radionuclides contributing significantly to the decay heat of irradiated nuclear fuel, and for which TAGS would be beneficial are shown in Table 6.

9. Sources of uncertainty in predicting the radioactive source term

There are a number of uncertainties that propagate through the procedures used to predict the radioactive source term; these are:

- Nuclear data uncertainties: nuclear cross-sections, distributions of secondary neutrons.
- Radiation source description: space-, energy-distribution.
- Geometry modelling: configurations, dosimeter locations.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003

– Material compositions.
– Mathematical methods and simplifications used in computations.

An increased effort is devoted today to providing nuclear data with their uncertainties. The assumption is made that the values follow a normal statistical distribution, thus the average or expected value is used for calculations and the corresponding uncertainties are the second moment of the distribution (standard deviation and correlation matrix = covariance matrix). Many evaluated nuclear data come today with the corresponding covariance data; tools for processing such data for practical use have been developed (Ivanov et al., 2012), so that the source term can be derived with associated confidence bounds.

10. Conclusions

Good quality nuclear data are essential to be able to characterise the different radioactive waste forms, and to provide quality prediction of the source term for waste management. Of major importance are data for the build-up of several radionuclides such as actinides and fission products; also decay-heat issues are important. These data are required to study the different scenarios for power production and waste minimisation; efforts are devoted to improving the quality of the data through validation against experiments, re-evaluation, adding covariance data for uncertainty quantification. A wealth of validated data is available from several Nuclear Data Centres.

References

Carter, J.T., Jones, R.H., Luptak, A.J., 2011. Used Fuel Disposition U.S. Radioactive Waste Inventory and Characteristics Related to Potential Future Nuclear Energy Systems. May 2011, FCRD-USED-2011-000068.

Hwang, R.N., 2010. In: Azmy, Y., Sartori, E. (Eds.), Resonance Theory in Reactor Applications. Nuclear Computational Science, Springer, 2010, pp. 217–290. ISBN: 978-90-481-3410-6.

IAEA, 2004. Application of the Concepts of Exclusion, Exemption and Clearance, Safety Guide No. RS-G-1.7.

Ivanov, K., Avramova, M., Kamerow, S., Kodeli, S., Sartori, E., Ivanov, E., Cabellos, O., 2012. Benchmark for Uncertainty Analysis In Modeling for Design, Operation and Safety Analysis of LWRs; volume I: Specification and Support Data for the Neutronics Cases, April 2012, NEA/NSC/DOC(2012) 10.

McFarlane, R.E., 1998a. An Introduction to the ENDF Formats, UC-LANL LA-UR-98-1779.

McFarlane, R.E., 1998b. Radioactivity Data, LANL <http://t2.lanl.gov/tour/tb004.html>.

McFarlane, R.E., Muir, D.W., 1994. The NJOY Nuclear Data Processing System, Version 91. LA-12740-M (1994). <http://t2.lanl.gov/codes/njoy99>.

OECD, 2002. Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles – A Comparative Study. ISBN: 92-64-18482-1.

OECD, 2003. Plutonium Management in the Medium Term – A Review by the OECD/NEA Working Party on the Physics of Plutonium Fuels and Innovative Fuel Cycles (WPPR), NEA4451, ©. ISBN: 92-64-02151-5.

OECD, 2006. Physics and Safety of Transmutation Systems – A Status Report, NEA No. 6090. ISBN: 92-64-01082-3.

OECD, 2013. Minor Actinide Burning in Thermal Reactors – State of the Art Report.

Parks, C.V., Wagner, J.C., Gauld, I.C., 2002. Research to support expansion of U.S. regulatory position on burnup credit for transport and storage casks; practices and developments in spent fuel burnup credit applications. In: Proceedings of an Technical Committee meeting, Madrid, 22–26 April 2002, IAEA-TECDOC-1378.

Please cite this article in press as: Sartori, E. Nuclear data for radioactive waste management. Ann. Nucl. Energy (2013), http://dx.doi.org/10.1016/j.anucene.2013.02.003