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Nuclear data for radioactive waste management

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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-

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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/.

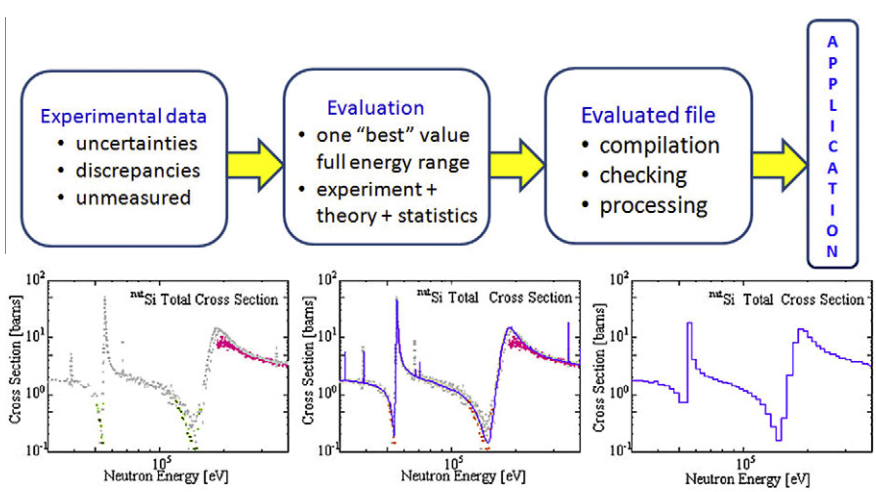

Fig. 1. Different steps in nuclear data evaluation and processing.

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