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Nuclear data requirements for accelerator driven sub-critical systems – A roadmap in the Indian context

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Abstract. The development of accelerator driven sub-critical systems (ADSS) require significant amount of new nuclear data in extended energy regions as well as for a variety of new materials. This paper reviews these perspectives in the Indian context.

Keywords. Neutron data; spallation neutrons; (n, xn) reactions; thorium cycle; accelerator driven sub-critical systems; benchmark.

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1. Introduction

This paper briefly presents the perspective of the author on the nuclear data requirements of the accelerator driven sub-critical systems (ADSS) in the Indian context. The Indian activities [1–3] in nuclear data, as background information, both experimental and theoretical are available in detail and are not reproduced here to save space. The ADSS route [4,5] is especially attractive as it has the potential to accelerate the use of thorium for power production to meet the growing energy needs of the Indian population and to incinerate minor actinides and fission products in a sub-critical environment.

The on-going and preliminary research studies for the ADSS concepts use existing basic nuclear data developed for thermal, fast and fusion reactors, those generated towards fundamental physics understanding of the nucleus and origin of the Universe and applications such as in space and astrophysics.

India recognizes that basic nuclear data measurements involve cutting-edge technologies such as powerful neutron sources, advanced electronics for data analyses, and availability of pure isotopic and elemental targets. Encouragement is being given to these indigenous and creative efforts to develop all these capabilities and to sustain them over the longer term. Experimental basic neutron data measurements using accelerator- and reactor-based neutron sources and also a programme of critical facility [6] for integral validation of reactor physics data of advanced heavy water reactor [7] at BARC have been successfully initiated.

The leading particle physics team at CERN lead by Carlo Rubbia recognized [8–10] the importance of nuclear data needs right at the very beginning of the evolution of the concept of energy amplifier (EA). In this review paper, we make use of available data sources from the IAEA, published literature and CERN publications on energy amplifier and perspectives based upon India's work on nuclear data [1–3] in our attempt to present a bird's eye view of the nuclear data needs of the ADSS in the Indian context. The improvement of basic nuclear data is part of the overall strategy of improving and validating the simulation tools, which consist of computer codes, modeling approaches and computerized databases [11].

2. ADSS related nuclear data and availability of nuclear data

Conceptual studies of accelerator driven sub-critical systems (ADSS) have given a fresh look at the use of thorium fuel cycle in a lead–bismuth coolant environment. In parallel to the ADSS, Indian advanced reactor programmes [7,12–14] with respect to advanced reactors require improved nuclear data [15,16] that overlap with the on-going international efforts to develop innovative, inherently safe, proliferation-resistant and long-life cores, with features using thorium such as in INPRO and next generation nuclear energy systems known as 'Generation IV' [17].

The function of the spallation target is to convert 1 GeV protons generated by the proton accelerator to low energy (less than 20 MeV) neutrons and deliver them to the energy amplifier region. The actual conversion of fertile to fissile nuclei or the transmutation of radioactive nuclei to stable ones takes place in the ADSS blanket favourably in the resonance energy region (less than 50 keV) and thermal regions where the capture cross-section is the largest. In performing this design function, the spallation target must be cooled sufficiently. Nuclear data to predict nuclear collision, isotope production, formation of gases and heat generation are needed at this very first step. Nuclear data are also needed to reduce the risk related to radiological release or other detrimental consequences such as, for instance, that are resulting from off-normal situations (loss of coolant in the target, maladjustment of proton beam with respect to time and spatial distribution).

Maximizing neutron production is one of the tasks for a target design. The selection of high Z material is preferred, as the target should have a large number of nucleons per proton to cause intra-nuclear reactions. For such energies, the proton ejects bound neutrons directly. Consequently, the emitted neutrons tend to be forward peaked and have a very high energy up to the proton energy. The pre-equilibrium process related and 'boil off' neutrons are emitted almost isotropically with peak value at 1–2 MeV.

The expected performance of the target of EA depends upon the efficiency to slow down and transfer the resulting low-energy neutrons to the energy amplifying sub-critical medium.

Nuclear data on the spallation source descriptions are needed to achieve a target design with the overall goal of achieving the required neutron fluxes in the blanket volume with a minimum accelerator beam power. Another important aspect of target design is the beam window or windowless spallation source and if the spallation source should be solid or liquid. Nuclear collision data are common to both solid and liquid but liquid target like eutectic need additional data related to flow dynamics and problems arising from the gas formation. Thus, all nuclear data information are required in order to optimize neutron production, minimize neutron absorption in the target and maximize neutron leakage to the blanket.

The nuclear data requirements for ADSS span energies up to GeV energies, much beyond the energies involved in the design and operation of systems used in India (see papers in NWND [1]), the 10 MeV neutron energies being the upper limit in thermal reactor systems in BARC and 15 MeV for fast reactors at IGCAR, Kalpakkam. The CERN team, for instance [18], has addressed, with illustrative sensitivity studies, the importance of nuclear data for ADSS. In the IAEA Advisory Group Meeting [19], nuclear data needs have been stressed as important. The need for improved data of neutron cross-sections for transmutation has also been addressed, for instance, by [20].

In modeling ADSS, such as using the FLUKA code system (http://www.fluka.org/), accurate nuclear data are essential for the following tasks:

- The modeling of high-energy neutron and proton cascade from the initial energy of a GeV down to 20 MeV in the spallation target and the ADSS blanket. 20 MeV is referred to as the point of upper energy historically being used in the ENDF/B files covering energy ranges from 10^{-5} eV to 20 MeV for fission and fusion reactor applications.
- The neutron transport from 20 MeV downwards of neutrons (and protons) in the spallation plus the ADSS medium.
- The description of fuel evolution up to the required burn up (150 GigaWatt-Days per metric ton (GWD/Te) or larger) as a result of neutron interactions and nuclear decays (The evolution of all the major and minor actinides, fission products, isomeric states production and decays and sequential multi-step nuclear reactions need to be tracked. There are over 50 minor actinides and over 2000 fission products to be covered.)
- The nuclear activation of structural materials and coolant due to the presence of neutron fluence and proton beam-induced high-energy cascade.

Nuclear data are required [21] as a function of energy for all the materials present in the ADSS system to help perform the following simulation tasks of the ADSS. For ADSS, such data have to be generated from scratch and this has been recognized [22] to demand the pursuit of basic research.

ADSS studies require detailed nuclear data to perform neutron transport calculations, to calculate energy distributions of down-scattered neutrons and those required for shielding in ADSS. Nuclear data for $(p, xn), (p, xp), (n, xn)$ and (n, xp) reactions (see ref. [23] for the role of (n, xn) reactions) are needed as a function of incident energy and ejectiles emitted at various emission angle and energies for all materials directly exposed to the particle beam.

For shielding calculations, $(p, x\gamma)$, $(n, x\gamma)$ and (n, xn) yield data are needed. Additionally, individual nuclide production or spallation yields are required. Nuclear data for the calculation of displacement damage cross-sections along with hydrogen and helium production cross-sections up to high energies for structural materials are required. Transmutation and activation cross-sections are also required including cross-sections for the formation of isomeric states and those formed via two-step reactions for the research related to application of ADSS.

3. Perspectives on nuclear data for ADSS studies

Nuclear data activities for ADSS in the road map for ADSS, fall into four distinct categories:

(a) Measurements of basic nuclear data with specifications of covariance error matrix.
(b) Compilations coordinated by a classical data centre, computerized visualizations, large data files information management, evaluations, that include nuclear model based predictions, creating computerized ENDF/B files; use of Bayesian law.
(c) Physics laws based nuclear data processing of computerized files (ENDF/B) into different forms that are readily usable by codes to perform Monte Carlo or discrete ordinate calculations. The processed data should faithfully reproduce (see for instance, [24]) the quality of the corresponding basic nuclear data.
(d) Design and development of experimental ADS 'zero power' clean sub-crirtical facilities; integral measurements and integral validations and adjustments of nuclear data by using experimental ADSS facilities.

The four steps mentioned above are iterative, implying that a long term effort in a sustainable manner should be evolved in India. Feedback from results in Step 4 for instance can lead to Step 1 by suggestions to perform new and improved measurements of basic data. The nuclear data needs in accelerator-based systems depend much on the neutron spectra and materials particular to the concept are to be studied. Ideally, we should plan to equip our programme in terms of manpower and resources with a comprehensive approach to cover as many known ADSS designs as possible to meet the nuclear data needs. Measurements of basic data are needed for a number of isotopes as outlined in the CERN reports [9,10] on their ongoing neutron time of flight (n_TOF) studies [1].

4. Nuclear data requirements for engineering design of the ADSS

The nuclear data requirements needed for a number of isotopes and elements [21] as illustrated in table 1, specifically influence each of the following engineering aspects in the overall design of ADSS:

- ADSS core design
- Spallation target design: For this, the variants are, beam falling on window or with no window, target to be solid or eutectic, heavy metals like W, Pb etc. or actinides like thorium, uranium etc. The corresponding thermal and physical properties of target materials for a reliable thermal hydraulic design of heavy elements are also required for the design.

- Quantification of core performance characteristics over its lifetime.
- Confirmation of system nuclear safety. This includes reliable prediction of feedback coefficients such as the coolant void reactivity and Doppler reactivity effects under all system conditions.
- Meeting licensing requirements from the licensing authorities (e.g., AERB, India).
- The degree of sub-criticality $(1 - k_{\text{eff}})$ has to be carefully optimized by balancing the objectives of minimizing accelerator power requirement (favours larger multiplication of source neutrons by smaller values of sub-criticality margin).
- The nuclear design of ADSS should be robust to preclude criticality or excessive power increases in response to operation or accidental additions of reactivity of uncertain magnitude (which favours larger sub-criticality margins) as well as bugs in the operation of accelerator which may mean sudden stop of beam current or reduction in beam current.

With ^{239}Pu–U or ^{232}Th–^{233}U fuel cycle which are of interest to India, quantitative prediction of burnup reactivity loss for the projected burnup extending to 150 GWD/Te and beyond is required. This very large burnup, as compared to the present day values (of about 7 GWD/Te in PHWRs, about 20 GWD/Te in Tarapur BWRs and about 40 GWD/Te in PWRs), demands, for development of optimized strategies, detailed and accurate nuclear data for higher isotopes of Pu, fission products, minor actinides and activation products. The change in energy-dependent neutron spectra and their influence on core parameters as a function of burnup should be correctly simulated. If the option is ^{232}Th–^{233}U fuel cycle, then ^{232}Th cycle needs detailed study in the light of high-energy neutrons which are capable of initiating (n, xn) reactions.

- Fuel management options and dealing with mixture of different fuel types require accurate nuclear data for all the elements involved.
- A detailed understanding of space–time dynamic behaviour of ADSS during operational transients and potential accident sequences need nuclear data of resonances to predict Doppler effect and delayed neutron fraction data for all fissions at the energies involved.
- The ability of the ADSS to accommodate various source transients should be demonstrated. This requires accurate knowledge of nuclear data even in the resonance region in order to predict with confidence all the feedback mechanisms including Doppler and coolant void coefficient of reactivity.
- The target or the spallation region contains materials that are exposed to direct beam protons and secondary neutrons ranging from primary beam energy down to very low energies. In the case of Carlo Rubbia's concept, this is a natural lead. In other versions it may be lead–bismuth eutectic and after its irradiation the products are different and highly radioactive. Similarly, there can be many variants of spallation target and all will need fresh look of nuclear data from the point of high energy of neutrons. Also, surrounding the spallation target region are fuel clusters, blanket and coolant/moderator materials that are not exposed to direct proton beam but which are irradiated by

a high flux of neutrons ranging from a hard spectrum with a significant number of medium energy neutrons, to an intermediate neutron energy spectrum, and finally to essentially a thermal/fast spectrum based upon the design of the ADSS.

As of now, the link to the ADS databases in the mirror website in India [25] provides updated nuclear data, both basic and processed for a number of nuclides (such as ^{238}U up to 30 MeV; ^{208}Pb up to 200 MeV, ^{27}Al up to 150 MeV etc.), as a starter, for use in studies towards energy amplification, ultimate disposal of nuclear waste by incineration and thorium utilization extending to energies up to GeV.

5. Why the nuclear data are still not well-defined after many decades of nuclear power?

In the opinion of the author, nuclear energy has been successfully introduced to human civilization without adequate and detailed knowledge of the basic nuclear data of nuclei. This has been achieved by the nuclear community by carefully performing a number of one-to-one integral experiments. This is exactly the challenge facing nuclear scientific community when faced with new designs such as ADSS. The nuclear data input to reactor design strictly involves the microscopic cross-sections at energy $E_0, \sigma(E_0)$, which is obtained as the response quantity:

$$\sigma(E_0) = \frac{1}{N} \int \sum (E)\delta(E - E_0)\mathrm{d}E, \tag{1}$$

Table 1. The isotopes or elements requiring complete nuclear data for neutron and gamma transport for ADSS [21].

Object	Nuclides/elements
Target materials	^{209}Bi, ^{208}Pb, ^{207}Pb, ^{206}Pb, ^{204}Pb, Pb, ^{186}W, ^{184}W, ^{183}W, ^{182}W, W, ^{181}Ta, Ta Zr, Sn, Hg, U, Pu, F, Cl, Na, Fe, Al
Po production	$^{209}\mathrm{Bi}(p, xn)^{207,208,209}\mathrm{Po}$, $^{209}\mathrm{Bi}(n, \gamma)^{210}\mathrm{Bi} \rightarrow {}^{210}\mathrm{Po}$
Minor actinides	^{237}Np, ^{238}Np, ^{241}Am, ^{242m}Am, ^{242}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{248}Cm
Long-lived FP	^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, ^{135}Cs
Fuel compositions	^{238}U, ^{235}U, ^{239}Pu, ^{238}Pu, ^{14,15}N, O, F, Cl
Th cycle	^{232}Th, ^{231}Pa, ^{232}Pa, ^{233}Pa, ^{233}U, ^{234}U
Structural materials	Zn, Cu, Ni, Co, Fe, Mn, Cr, Ti, Ca, Ar, Al, Mg, Na, O, N, C, B, Be, He, ^{3}T
Shielding	O, Si, P, Ca, Ti, Fe

where N is the number of target nuclei interacting with the neutrons, $\sum(E)$ is the macroscopic cross-section at energy E and $\delta(E - E_0)$ is the Kronecker delta function. Equation (1) already tells us that we need to be in deep cutting-edge technology to be able to measure cross-sections precisely and accurately. In order to experimentally determine the cross-section at a given energy, we ideally need to obtain the response function using a Kronecker delta function like mono-energetic neutron source. Obviously, in practice, we then need highly mono-energetic neutron source of high intensity that is tunable in neutron energy, pure samples of sufficient number of atoms and efficient detector systems. The analyses of experimental data to obtain 'cross-sections' is also very challenging due to the corrections to be applied due to the thickness of the sample, neutron beam profile, energy and detector resolutions, spatial non-homogeneities in the target sample and several corrections such as due to Doppler broadening and impurities.

Even in the case of eta of ^{235}U, it has not been possible to obtain convergence among the recent data sets. The values of eta of ^{235}U among recent data files (2002) ENDF/B-VI.8 (USA), JENDL-3.2 (Japan) and JEF-2.2 are still very discrepant. For more details refer to [26] wherein we illustrated that nuclear energy has been introduced to mankind without adequate knowledge of nuclear data, by illustration with the case of eta of ^{235}U, as example.

Nuclear data requirements influence the design of ADSS and can be met in different ways. In modeling the ADSS and comparing with actual reality based upon ADSS experiments, we have unavoidably the effects of errors in nuclear data, errors and approximations in modeling and tolerances in fabrication. A direct consequence of these effects put together is that implementation of ADSS would certainly need sub-critical experimental facilities and integral ADSS experiments along with improved nuclear data to speed up the production of an accurate and reliable physics design. With improvements in data, the need for a number of costly integral experiments can be drastically reduced. Note that the experimental validation efforts in critical facilities can never exactly verify the simulated states of higher burnup. Therefore, the methods and data at operating conditions and at higher burnups get integrally visible for purposes of validation only by actual experience. In this sense, the successful development of nuclear energy is yet to be verified for multiple recycling at large burnups. Intense neutron sources, purer elemental/isotopic target samples, more efficient detectors and better electronics evolve as a result of improvements and innovations in cutting-edge technologies. With such an evolution, as a general rule, the generation of new nuclear data at the international facilities such as CERN n_TOF, GELINA etc., should continue to be performed in an iterative manner based upon the evolving and perceived needs.

6. CERN n_TOF: New measurements of nuclear data at CERN

The high-resolution resonance nuclear data are generated using neutron time-of-flight techniques using advanced facilities such as the 150 MeV electron LINAC (800 MHz, 10–400 m flight paths, 10 meV to 20 MeV neutron energy region) at Geel, Belgium, the 800 MeV proton LINAC at Los Alamos National Laboratory (20 Hz, spallation, <500 keV neutrons, 20 m path length) and the CERN n_TOF

[27] which is briefly mentioned below. These are essential as lack of accuracy in resolved and unresolved resonance data poses a number of problems in safety related concerns of nuclear systems (see, for instance, [28]). Pulsed Van de Graff with few microamperes current or the D-T accelerators producing 14 MeV neutrons are also used for measurements of cross-sections using ^{7}Li(p,n) reactions. In the fifties, the Fermi neutron choppers were successfully used to pulse the reactor neutrons in order to measure low-energy neutron resonances by time-of-flight techniques.

As an exciting development in the last eight years, new measurements (see [27] for instance) of cross-sections using the neutron time-of-flight facility for applications to ADSS have provided an accelerated effort towards generating new nuclear data and in resolving discrepancies. The n_TOF facility at CERN uses a proton beam momentum of 20 GeV/c with an intensity (in dedicated mode) of 7×10^{12} protons/pulse. It has a repetition frequency of 1 pulse/2.4 s and a pulse width of 6 ns (rms). The number of neutrons per proton is 300. The lead target dimensions are $80 \times 80 \times 60$ cm^3. The coolant for which water is used moderates the neutrons. The moderator thickness in the exit face is 5 cm and the neutron beam dimension is 2 cm (FWHM). The CERN n_TOF facility is very unique. It has the highest instantaneous intensity neutron source worldwide at a 185 m flight path used in TOF cross-section measurements, excellent energy resolution, innovative data acquisition system based on flash ADCs (2 Tbytes/day via CERN CASTOR system), latest generation of detectors and beam monitors. The detector systems are at the cutting edge technology and include for (n,γ) measurements the use of ultra-low neutron sensitivity C6D6 detectors and high performance total absorption calorimeter. For fission cross-section measurements the CERN team uses low background PPAC set-up, the FIC detector. The beam monitors are characterized redundantly by ^{197}Au$(n,\,\gamma)$, ^{6}Li$(n,\,\alpha)$T, ^{235}U(n,f) monitors and a fully characterized facility for neutron cross-section measurements. The CERN n_TOF is the best facility for radioactive, rare, low cross-section samples and high-energy measurements. In the Indian context, it can be mentioned that for design and development of indigenous ADSS, India needs a fresh look on its own nuclear data and to generate its own resonance cross-section measurements.

7. The nuclear data of isotopes of thorium fuel cycle

The status of nuclear data of the major and minor isotopes ^{230}Th, ^{232}Th, ^{231}Pa, ^{233}Pa, ^{232}U, ^{233}U and ^{234}U in the thorium fuel cycle have received significant attention in the last 10 years compared to the seventies and eighties. The nuclear data of isotopes of thorium fuel cycle, for which enough attention was not paid until 1999 (see [29], for instance) has been significantly improved in the last 6 years as a result of a recently concluded IAEA-CRP entitled, 'Evaluated nuclear data for thorium–uranium fuel cycle' [30]. The basic and processed nuclear data libraries for the isotopes mentioned above are available to the public at the IAEA website [30] wherein more details are available and are not reproduced here to save space. Note that the data of isotopes in the U–Pu cycle themselves also need further improvement for advanced reactor systems particularly for long burnup cores [15,16].

8. Experience and lessons towards understanding the needs of data activities for thermal reactors in India and inferences for ADSS

The IAEA-CRP on 'Final stage of the WIMSD library update project' has been highly successful [31] in its impact [1] on the Indian nuclear programme. For instance, the KAPS-1 overpower transient could be explained [32] only with the use of new IAEA WLUP libraries. It should be stressed that the author dismisses the use of WIMSD conventions and the WIMSD library to study ADSS systems. However, based upon our experience in using the updated WIMSD libraries [31] for all types of thermal reactors in India, we can state the following with respect to perspectives on ADSS: Reliable design and operator's manual for each stage of the nuclear fuel cycle of the ADSS based upon accurate knowledge of nuclear data will help in safe use of nuclear energy by providing proper guidance on safety precautions and behaviour of ADSS under all system conditions. There is a need to continuously update nuclear data in reactor analyses over several decades to come even when the ADSS reactor systems would be already successfully operating.

9. Criticality benchmark of international quality

BARC has produced benchmark numbers for criticality of several pure minor actinides such as ^{231}Pa, ^{241}Am, ^{243}Am, ^{232}U and ^{233}Pa [33–36]. These studies are practically useful and used already by studies creating European manuals on radioactive transport regulations [37]. The efforts on such creation of benchmark numbers involve considerable amount of sensitivity studies with quality assurance in understanding and using the ENDF/B formatted files.

India has completed the creation of Indian KAMINI experimental criticality benchmark of international quality (see refs [30,38,39]). The Kalpakkam MINI (KAMINI) reactor is a ^{233}U fueled light water moderated and beryllium oxide reflected research reactor. Our report on KAMINI has been accepted as a benchmark of international quality. The uncertainty in the KAMINI benchmark in characterizing the predicted k_{eff} is ± 0.0059. As a result, in 2005, India has formally joined [38] the select band of countries contributing to the handbook of experimental criticality benchmarking of international quality organized by the US-DOE and the NEA (Paris). Preparation of an Indian experimental benchmark on thorium irradiation experiments and burnup measurements in PHWRs is in progress [39].

10. Concluding remarks

Accurate knowledge of nuclear physics data helps to improve and sustain the system intelligence for energy security of India. Measurement and evaluation of recommended values of accurate nuclear data belong to cutting-edge science and form an important component of evolution of basic nuclear physics and nuclear programme.

The quality assurance in design and safety studies in ADSS systems in the next few decades require new and improved nuclear data with high accuracy and energy resolution for thousands of nuclear reactions. This inevitably requires also the development and use of cutting-edge facilities, recognizing this perspective on par with the pioneers and advancing fundamental science. The experimental programmes in India and participation in international collaborations described [1–3] are being strengthened.

The roadmap in nuclear data for ADSS encompasses a wide range of activities in the field of measurements, compilations, computerized visualizations and large data file information management, evaluations which include nuclear model-based predictions, creating computerized ENDF/B files, physics laws-based nuclear data processing for multi-group and Monte Carlo applications, integral measurements and validations by us in experimental critical facilities. These voluminous numerical databases include not only interactions with neutrons but also with gammas and charged particles as incident beams. Indian researchers recognize the need for collaborating with advanced centers and international laboratories (see for instance [16,39–41]). In India, we are including all the national laboratories and university teams using the DAE–BRNS mechanisms (see [3]) in order to evolve a streamlined and coherent activity of nuclear data for ADSS and other applications that will be sustainable.

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