
Nuclear Data Development Related to the Th–U Fuel Cycle in India

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Abstract

This paper aims to share the excitement, challenges, and progress in the indigenous Indian efforts in nuclear data science in a generic context, in particular, towards the utilization of the thorium fuel cycle. To meet all the nuclear data needs in India for energy and non-energy applications, a Nuclear Data Physics Centre of India (NDPCI) was successfully formed in 2009. The NDPCI operates in a virtual mode at this time. Efforts are underway to create a sustainable physical center for the NDPCI. This paper also presents the interesting results of calculations that show a highly positive temperature coefficient for the BeO reflector in the KAMINI reactor, the only operating ^{233}U -fuelled reactor in the world.

Introduction

The nuclear data needs for the thorium fuel cycle are not only for the main fissile and fertile isotopes of thorium fuels, such as those that have already been discussed [1] (viz., ^{230}Th , ^{232}Th , ^{231}Pa , ^{233}Pa , ^{232}U , ^{233}U , and ^{234}U), but also inevitably include all the range of actinides for multiple fuels, those of coolants, structural materials, fission products, and minor actinides that are associated with the use of thorium-fueled reactors.

The Indian approach to nuclear energy, including committed efforts towards thorium utilization, involves the development of innovative reactor designs by using a multiphysics, multiscale modelling approach. India is energy-hungry, wanting to increase her human development index with her huge population, which is presently over 1.28 billion. Thorium fuel has a great potential to serve as a significant source of low-carbon electricity in India as part of a viable energy mix of options designed to last for several centuries. For the interested reader, a broad overview on current Indian perspectives on using thorium is nicely described in the contributions by Vijayan et al. [6], Wattal et al. [7], and Degweker et al. [8] in this ThEC13

proceedings. The official websites of the Department of Atomic Energy (DAE) [2] presents, to the interested reader, all the details of various on-going activities of the Bhabha Atomic Research Centre (BARC) three-stage program that, by design, inevitably involves closed fuel cycles with multiple fuels (U–Pu and Th–U) and with long burnup. India recognizes that nuclear data science is an essential base technology effort that forms part of the big data science in meeting all the Indian nuclear data needs for thermal, fast, fission–fusion, and accelerator-driven subcritical systems [3–5], which involve multiple fuel cycles with closed fuel cycles. It is recognized [3–5] that long-term nuclear data needs of closed fuel cycles with multiple fuels (U–Pu and Th–U), with high burnup, are demanding in the Indian context of the BARC three-stage nuclear power program.

Thorium Utilization Studies Need New and Improved Nuclear Data

The author believes that the best nuclear reactor design—one that would not be even remotely accident prone during its entire fuel cycle, that would create the minimum radioactive waste, would be fully proliferation resistant, would have the maximum tolerance of normal (and even remotely possible operator errors), man-made, and natural disasters—is yet to be made. Research associated with evolving new nuclear

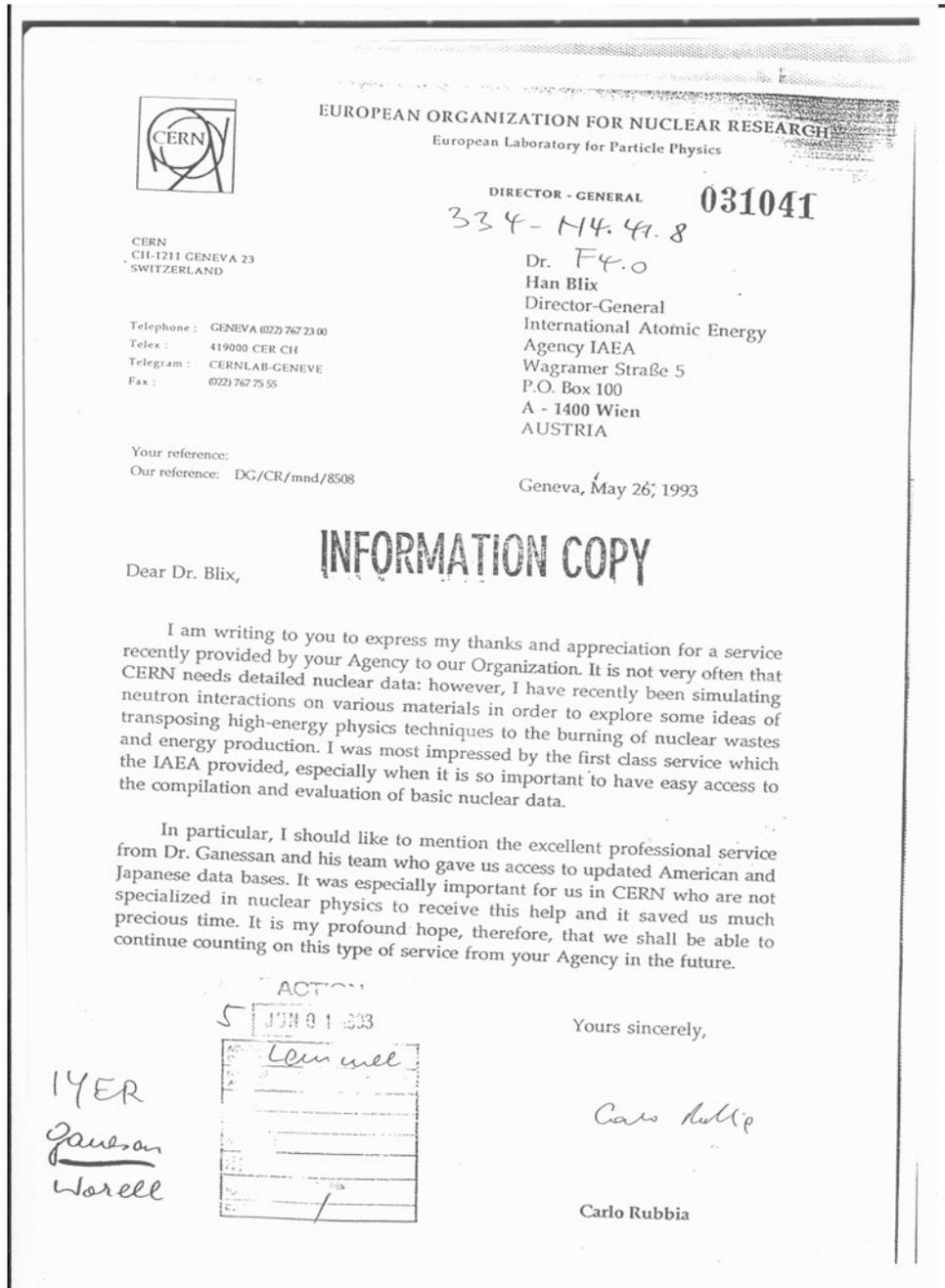
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energy systems using thorium within the context of the BARC three-stage program needs a significant amount of new and improved nuclear data, both at the differential and integral levels. The role of multiphysics multiscale modeling (MMM3) with “big” data science, including detailed nuclear data, in attempts to evolve the best reactor design cannot be over emphasized.

The letter from Prof. Carlo Rubbia, CERN, to Dr. Hans Blix, International Atomic Energy Agency (IAEA), in 1993 (Fig. 1) is an apt illustration of the fact that the strong need for basic nuclear data is automatically and naturally felt when serious studies of new physics designs (here, in the case of the Energy Amplifier by Carlo Rubbia and his team for utilizing thorium) are performed.

Fig. 1 Letter from Prof. Carlo Rubbia on nuclear data needs for the Energy Amplifier



Initial Indian Efforts at Kalpakkam on Nuclear Data for the Thorium Fuel Cycle

The author was involved in the nuclear data science efforts in India as early as the seventies and eighties, when a detailed look at some of the data statuses of the thorium fuel cycle was attempted. For instance, the evaluation of unresolved resonance parameters for ^{232}Th and ^{233}U was attempted [9, 10] in Kalpakkam in the late seventies. Attempts were also made [11] from the seventies onwards to create Indian processing code systems to use the freely available basic evaluated nuclear data libraries. Partial nuclear data evaluations were also attempted as the first exercise and India's first basic evaluated nuclear data file in ENDF/B format was on the ^{232}Th isotope, which provided evaluated nuclear data [12] above the resolved resonance region under the IAEA Nuclear Data Library (INDL) Project in the early eighties. The interesting fast reactor sensitivity study performed at Kalpakkam [13] in the late eighties by using different data sets for ^{232}Th and ^{233}U illustrated that the doubling time of a conceptual thorium-fueled fast reactor changed by a factor of nearly two owing to use of different nuclear data sets by changing only the data for ^{232}Th and ^{233}U and leaving the nuclear data of all other isotopes unaltered.

Nuclear Data Physics Experiments for the Thorium Fuel Cycle

The nuclear data available in the IAEA EXFOR database [14] clearly indicates increased experimental nuclear data activities in India in the area of nuclear data measurements in recent years. Due to space constraints, the detailed EXFOR entries and discussion of nuclear data, as well as graphs of nuclear data related to the thorium fuel cycle that have been generated in India will not be reproduced here, however, they can be found in [14].

Surrogate techniques are being employed [15, 17–19] by BARC to measure neutron-induced nuclear reaction data of unstable actinide nuclei. Consider the situation when we do not have an unstable target and also do not have a neutron beam: how can we get the neutron-induced cross sections in such cases? Indian nuclear physicists have been specializing in complex heavy-ion nuclear fusion reactions and associated multi-parameter measurements. This in-house, specific expertise helped BARC to enter the area of surrogate methodologies to generate neutron-induced nuclear reaction data. BARC has successfully obtained [15, 17–19], experimentally, by using surrogate nuclear techniques, the neutron-induced fission cross-section data of unstable minor actinides such as ^{233}Pa ($t_{1/2} = 26.975 \pm 0.013$ days), ^{234}Pa ($t_{1/2} = 6.70 \pm 0.05$ h), ^{239}Np ($t_{1/2} = 2.356 \pm 0.003$ days), ^{240}Np ($t_{1/2} = 61.9 \pm 0.2$ min), and ^{241}Pu ($t_{1/2} = 14.325 \pm 0.006$ years). In all these efforts [15, 17–

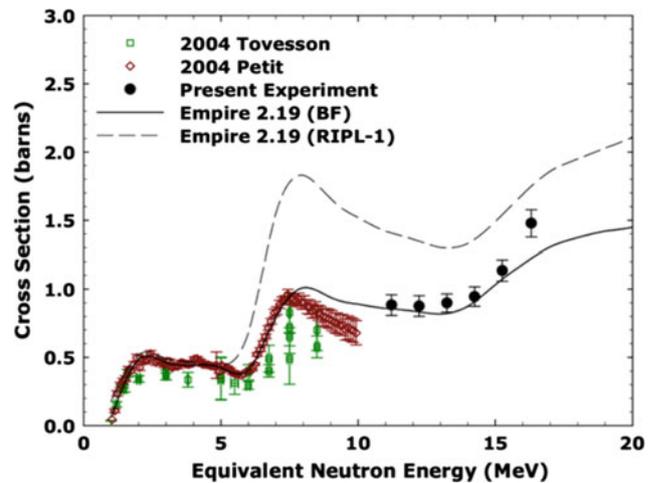
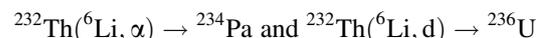


Fig. 2 Experimental $^{233}\text{Pa}(n, f)$ cross section along with EMPIRE-2.19 predictions [15]

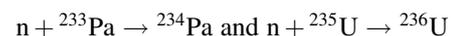
19], nuclear-model-based predictions using standard nuclear model codes, such as TALYS and EMPIRE, are also being performed.

The new hybrid surrogate ratio method (HSRM) approach [15] is being used to determine the $^{233}\text{Pa}(n, f)$ cross section in the excitation energy range of 11.5–16 MeV (Fig. 2).

In this BARC hybrid approach, a weakly bound projectile nucleus, ^7Li , was employed as a beam hitting a single target of ^{232}Th . We produce, thereby, in situ, the two nuclei, ^{234}Pa and ^{236}U , by two different direct reactions:



The ^{234}Pa and ^{236}U nuclei are treated as surrogates for the reactions:



The present BARC measurement [15] is unique in that the two residues are formed with an overlapping excitation energy spectrum. In the HSRM, the same target is used to populate the two element systems and, thus, by taking a ratio of the two reaction rates on the same target, systematic uncertainties due to target thickness, beam current, and dead time in the determination of the ratio of fission decay probabilities corresponding to ‘desired’ and ‘reference/monitor’ reactions, are eliminated. The EXFOR entry for this surrogate measurement of the $^{233}\text{Pa}(n, f)$ cross section [15] was completed. The EXFOR entry numbers 33,023 and D6075 for the above-mentioned data took into account two inadvertent errors in the analysis and publication. One error was taking the ratio of 233/234 as unity and the other was the usage of an outdated value of neutron separation energy for ^{234}Pa . The corrected plots from the EXFOR database [16] are comparable with the uncorrected versions, as shown in Fig. 3.

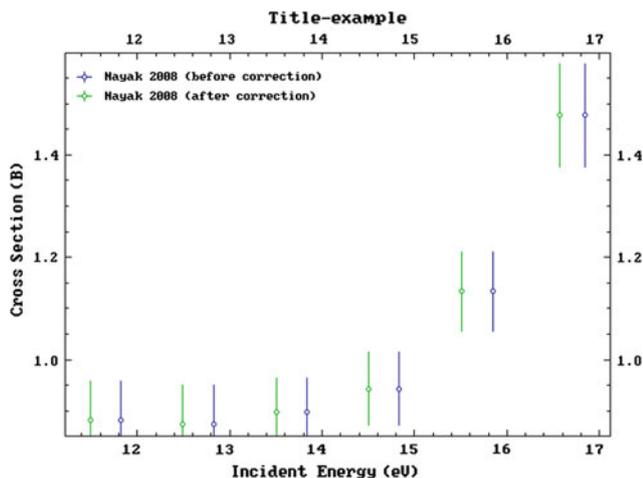


Fig. 3 The $^{233}\text{Pa}(n, f)$ cross section as a function of the equivalent neutron energy. The publication [15] had two inadvertent errors: 1 Skipping of center of mass to laboratory transformation and 2 adoption of 5.45 MeV as the $\text{Sn}(^{234}\text{Pa})$ value

The BARC team has also employed HSRM to determine the $^{239}\text{Np}(n, f)$ and $^{240}\text{Np}(n, f)$ cross sections in the equivalent neutron energy range 10.0–16.0 MeV [17] by using $^7\text{Li} + ^{232}\text{Th}$ and $^{6,7}\text{Li} + ^{238}\text{U}$ transfer–fission coincidence measurements. The $^{241}\text{Pu}(n, f)$ cross sections have been determined [18] by the surrogate ratio method in the equivalent neutron energy range 11.0–16.0 MeV by using $^{238}\text{U}(^6\text{Li}, d) \rightarrow ^{242}\text{Pu}$ and $^{232}\text{Th}(^6\text{Li}, d) \rightarrow ^{236}\text{U}$ transfer reactions at $E_{\text{lab}} = 39.6$ and 39.0 MeV, respectively. The BARC team has also carried out [19] measurements of the $^{234}\text{Pa}(n, f)$ cross section in the MeV energy region by employing the respective reactions, $^{232}\text{Th}(^7\text{Li}, \alpha) \rightarrow ^{235}\text{Pa}$ and $^{232}\text{Th}(^7\text{Li}, t) \rightarrow ^{236}\text{U}$. Generating the covariance error matrix for nuclear data obtained in such surrogate-based approaches is desirable.

In the thermal region, the $^{234}\text{Pa}(n, f)$ cross section was deduced experimentally [20] in the form of the $^{233}\text{Pa}(2\text{nth}, f)$ cross section by using a fission track technique by irradiation of ^{233}Pa in the APSARA thermal reactor. The cross section value was found to be 4834 ± 57 b. The $^{233}\text{Pa}(2\text{nth}, f)$ cross section was calculated theoretically by using the TALYS computer code and was found to be in good agreement with the experimental value after normalization with respect to $^{241}\text{Am}(2\text{nth}, f)$. It may be noted that ^{234}Pa has a half-life of 6.7 h. Of course, it remains to be determined under what neutron flux conditions reactions with large cross sections become important. Additionally, whether such reactions play a role in practical applications, for instance, in exotic thorium systems with unusually high neutron flux and transients, also needs to be investigated.

The cross sections and fission yield data of actinides were measured by BARC teams by using activation methods with neutrons from the spectrum of neutrons arising from $^7\text{Li}(p, n)$ reactions, in D-T (14 MeV) neutron facilities. Photon–nuclear reaction data using Bremsstrahlung gamma sources with electron accelerators have been experimentally generated for a range of targets and electron energies. An illustrative reference list of recently performed nuclear data physics experiments is presented in [15, 17–26].

Mirror Websites for Nuclear Data

The online nuclear data services (NDS) at BARC (<http://www-nds.indcentre.org.in/>) mirrors the nuclear data website of the Nuclear Data Section of the IAEA, Vienna (<http://www-nds.iaea.org>). Under this arrangement, online updating is performed every 12 h in the mirror with the IAEA website through a 2 MB direct link. The server is being maintained, with manpower and machinery, by the BARC Computer Division. It offers faster downloads. The India mirror site is getting increased usage.

Indian EXFOR Compilations of Nuclear Data

India became a member of the international nuclear reaction data compilation network (NRDC) by invitation in 2008. India has thus far contributed more than 264 EXFOR entries as accepted by the IAEA [27] at the time of writing. Introduction of the EXFOR culture [28] to people in India, including those in the area of basic nuclear physics, has become a relatively simple task with the new managerial initiatives of the NDPCI, which have included holding EXFOR workshops in India. Indian EXFOR compilation workshops in nuclear data compilations have become a role model [29] and have become a useful value addition to the nuclear physics community in India, providing greater visibility for the experimental measurements and research work done by India. The Indian EXFOR workshops have been phenomenally successful in a systemic sense, with the incentives towards sustainable viability arising from the demographic advantages in India. The importance of such highly focused training courses on EXFOR is well recognized by the BARC scientific community and is deliberately kept separate from the national nuclear physics conferences. The NDPCI is trying to form a permanent EXFOR compilation team at BARC. The NDPCI is responsible for all EXFOR compilations of experimental nuclear physics data generated in and published by nuclear physics facilities [28].

IAEA Coordinated Research Projects (CRPs)

India actively participated in and benefitted greatly from the IAEA CRP on “Evaluated Data for the Thorium–Uranium Fuel Cycle,” 2003–2006 [30]. Under the scope of this CRP, India contributed, for instance, the KAMINI benchmark [31] that was published in the International Criticality Safety Benchmark Evaluation Project (ICSBEP) Handbook. For more information on ICSBEP benchmarks, see: <http://icsbep.inl.gov/>.

Interestingly, under the scope of the IAEA WIMS library update project (WLUP) [32], improved nuclear data for thorium nuclide chains became available for the users of the WIMSD lattice cell code. The Indian reactor physics community use the updated WIMSD nuclear data libraries [32] provided by the IAEA in all design calculations. Using the XnWLUP software [33], we generated over 100,000 inter-comparison plots of multigroup data sets (including, number of nuclides, partial reactions, evaluated files, ratio of discrepancies, zoomed portions in energies) in WIMSD format to have a look at the discrepancies in nuclear data for various isotopes. The KAPS-1 over-power transient could be explained [34] only with the use of the new WLUP libraries. The safety and operational requirements of power plants are engineered with a number of one-to-one mock-up experiments providing adequate and conservative safety margins. However, the KAPS-1 over-power transient incident sent out a strong signal to the Indian nuclear community that in general, whether the U–Pu or Th–U fuel cycle is used, the design manuals of operating nuclear systems should be updated every few years as and when improved nuclear data becomes available. Indian scientists are aware of the limitations of the use of WIMSD conventions in advanced reactor designs. For instance, the fission spectra effect in WIMSD conventions owing to fissions in isotopes of the thorium fuel cycle was studied in a recent research paper [35].

Analyses of Irradiation of Thorium Bundles in PHWRs

The Indian post-irradiation experiment (PIE) of thorium [3, 31, 36] spanned two decades and involved hundreds of experts, as is the case with such PIE experiments associated with operating reactors. Identical loading of thorium bundles [36] was used in the 220 MW_e power stations, KAPP-1&2, KAIGA-1&2, and RAPS-3&4, to attain flux flattening in the initial core. The thorium oxide in all the 35 bundles put together in a reactor is about 400 kg. The bundles loaded in KAPP-1&2, KAIGA-1&2, and RAPS-3&4 went through over 500 days of full power operation, amounting to about 12,000 MWd/tonne burnup and these bundles have already been discharged from the core. After a cooling period of five

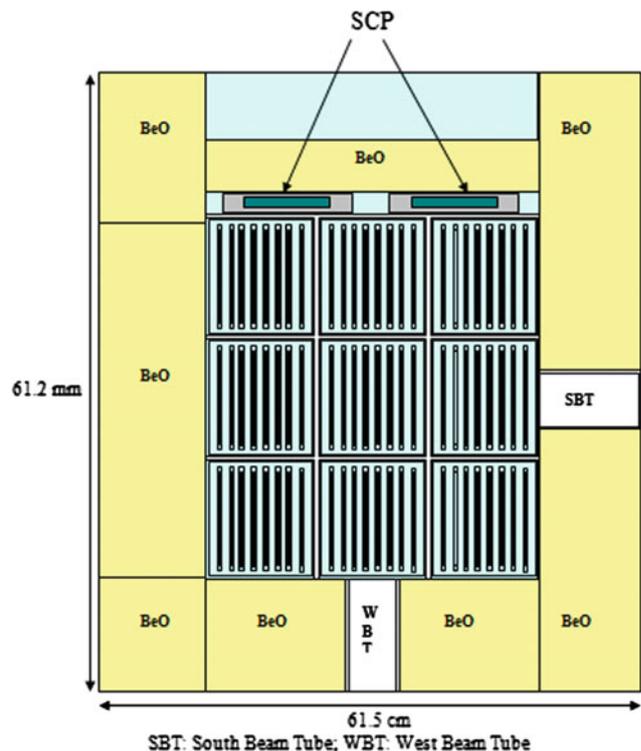


Fig. 4 Horizontal cross-section view of KAMINI core-reflector assembly. (After <http://www.igcar.ernet.in/benchmark/science/26-sci.pdf>). See [39] for full specifications as used in the calculations

years, samples were obtained from one of the irradiated ThO₂ bundles and have been analyzed experimentally by alpha spectrometry for ²³²U and by thermal ionization mass spectrometry for ²³³U, ²³⁴U, ²³⁵U, and ²³⁶U by two different groups at BARC. The previous analyses by the two teams at BARC gave an under-prediction factor of 6–8 for the production of ²³²U. The discrepancy was resolved and traced to the fact that the nuclear data of effective one-group values for cross sections of isotopes of the thorium fuel cycle and the use of assumptions (e.g., self-shielding) in the ORIGEN code package are applicable to traces of thorium in natural uranium rods, but not for the irradiation of bulk thorium rods used for power flattening in our pressurized heavy water reactors (PHWRs). Such discrepancies in the estimation of ²³²U are not acceptable as it leads to large errors [37] in shielding back-end facilities for the thorium fuel cycle. As a general remark, it may be noted that the ²³²Th(n, 2n) reactions that occur above 6.5 MeV are the main contributor to the production of ²³²U, in this case of thorium loading in PHWRs. We should note that it is a challenge to characterize the uncertainty in such threshold activation rates in thermal reactors as the fraction of neutron flux above 6 MeV calculated by lattice cell codes in the super cell model, is in the range of 0.05 % and the flux above 6.5 MeV is further very uncertain as the fission neutron spectrum, that is, the amount of virgin fission neutrons above 6.5 MeV, have large

Table 1 Calculated reflector BeO temperature dependence of k_{eff} of KAMINI

Temperature (K)	Density (g/cc)	k_{eff}	Std. deviation	Δk
293	2.9299	0.99007	0.00025	0
600	2.9058	1.00131	0.00025	0.01124 (11.2 mk)
800	2.8903	1.00648	0.00025	0.01641 (16.4 mk)
1200	2.8598	1.01385	0.00025	0.02378 (23.8 mk)

uncertainties to begin with. The on-going IAEA CRP on improving nuclear data of prompt fission neutron spectra [38] is important also in this perspective.

Positive Temperature Coefficient of the Reflector in KAMINI

The Kalpakkam MINI (KAMINI) reactor is a ^{233}U -fueled, light water moderated, natural convection cooled, beryllium oxide reflected, zero power research reactor (Fig. 4). The KAMINI reactor is located at the Indira Gandhi Centre for Atomic Research at Kalpakkam, India, and is the only operating ^{233}U -fueled reactor in the world. Because of the highly efficient reflector material, BeO, it has a very low fuel inventory (~ 612 g). The reactor is designed to operate at a nominal power of 30 kW. The built-in maximum excess reactivity of the reactor is restricted to 300 pcm (3 millik since 1 mk = 0.001 = 100 pcm), and this reactivity is apportioned to maintain various effects, such as coolant temperature load, irradiation sample load, operating margin, xenon load, and long-term burnup losses.

India contributed KAMINI [39] to the ICSBEP handbook in 2005. We used the ICSBEP specifications [39] for the KAMINI calculations. In the calculations, we set the reflector at various higher temperatures, keeping the temperature of the core part at room temperature. We briefly present the results of these calculations in Table 1. The reactivity jumps by several dollars when only the reflector is heated, while keeping the temperature of the core unchanged. Our calculations show a sizable positive temperature coefficient of the BeO reflector in KAMINI reactor at Kalpakkam.

We understand why the reflectivity worth goes up with temperature in the case of KAMINI. KAMINI is a highly reflected system. The use of massive BeO as a reflector in KAMINI contributes as much as 0.45 to the k_{eff} value. An increase in temperature of the BeO reflector results in increase in the scattering cross sections as illustrated in the WLUP multigroup comparison graphs (Fig. 5). Below 4 eV, neutron scattering cross sections in BeO are affected by chemical binding (Be in BeO). The density effect due to increase in temperature reduces reactivity, as we would expect, but this is not the dominant factor in these results.

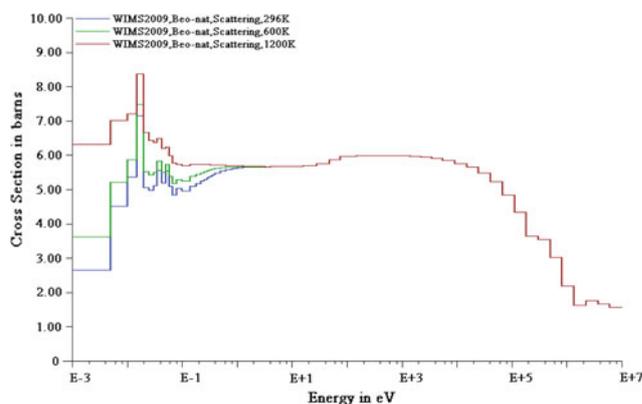


Fig. 5 Typical scattering cross sections of Be in BeO, generated by using the XnWLUP package [33] at 296, 600, and 1200 K. Note the very high temperature dependence below 4.6 eV

The author has informally suggested to the team at the Indira Gandhi Centre for Atomic Research (IGCAR) to experimentally confirm this new effect.

Formation of the Nuclear Data Physics Centre of India (NDPCI)

The great importance of nuclear data for the BARC three-stage program was institutionally recognized by the Department of Atomic Energy in 2004 as a result of the KAPS over-power transient incident [3, 5, 34]. A change of mindset took place in a systemic context, moving towards recognizing the importance of nuclear data physics for energy and non-energy applications [3, 5]. The DAE has declared nuclear data physics as a thrust area.

Activities of the Nuclear Data Physics Centre of India include the following actions towards satisfying the needs of the U–Pu and Th–U fuel cycles:

- Measurement of neutron- and charged-particle-induced cross sections as related to the Th–U fuel cycle, ADSS, AHWR, CHTR, shielding, fast reactors, and other programs of the DAE [2, 6–8], also medical isotopes production;
- Generation and use of co-variances as part of nuclear data evaluation and usage;

- Compilation and evaluation of nuclear reactions and nuclear structures; EXFOR compilations, ENSDF-related compilations;
- DAE-BRNS sponsored NDPCI theme meetings and national conferences on topics in nuclear data physics;
- Advanced reactor applications to enable use of updated nuclear data libraries in plug-in formats, such as for discrete ordinates and Monte Carlo codes;
- Coordination on nuclear data physics involving IAEA NDS and to be a single window from India to IAEA NDS;
- International collaborations with: the CERN neutron time-of-flight facility (n_TOF) under the BARC memorandum of understanding, Korea, IAEA CRPs;
- Identification of faculty and support for formation of useful local neutron data centers in Indian universities and institutes.

The NDPCI, despite its phenomenal progress since 2004, in the opinion of the author, is still in the lower part of its learning curve. NDPCI recognizes that a large amount of work will be essential to create ENDF/B formatted evaluated nuclear data files using new experimental data. These tasks include follow up with “raw” nuclear data compilations, critical evaluation using inference and data evaluation/assimilation methodologies, production of new ENDF/B formatted libraries, including co-variances extending to higher energies, and quality assured nuclear data processing activities to provide the designers/users of innovative systems with “ready to plug-in” processed data that has been integrally validated for use in applications. Nuclear data sensitivity studies such as in [13, 40] are part of activities promoted by the NDPCI.

Concluding Remarks

The indigenous Indian efforts in nuclear data science, especially those towards utilization of the thorium fuel cycle, are expected to continue with their associated excitement and challenges. Many topics mentioned during the talk are not covered here due to space constraints. The formation of the NDPCI and India’s successful activities [41] in nuclear data science, since 2004, has helped India in her efforts to graduate from the previous status of India as a user of freely available plug-in multigroup nuclear data and processed evaluated nuclear data files to the status of a contributor of improved/new basic nuclear data. The NDPCI is currently operating in virtual mode. Efforts by BARC are underway to create a sustainable physical center for the NDPCI.

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