Thermal Scattering Law ENDF Libraries for Liquid FLiBe

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ABSTRACT

Several advanced nuclear reactor concepts have been proposed in the past few years where FLiBe molten salt represents a major constituent of the core. In this case, neutrons produced in fission slow down and moderate in FLiBe (a eutectic with a mixture of 2:1 ratio of LiF and BeF_2) until they reach low energies (i.e, below 1 eV). At that stage, the thermalization process becomes dominant and the neutrons achieve a quasi-equilibrium energy state that is dependent on the temperature of the moderator. In neutronic simulations, the description of neutron thermalization is captured using the thermal scattering law (TSL), i.e., $S(\alpha,\beta)$, of the material in which low energy neutrons are interacting. $S(\alpha,\beta)$ defines the energy-momentum phase space that is available for an incoming low energy neutron. In addition, it is directly proportional to the double differential thermal neutron scattering cross section. In this work, the TSL of molten salt FLiBe is developed based on a generalized density of excitation states (GDOS) derived from atomic trajectories generated using classical molecular dynamics (MD) simulations that were performed with the LAMMPS code. The MD simulations utilized a Born-Mayer type atomic potential function that was verified to reproduce the properties of FLiBe including density and viscosity. The *FLASSH* code was used to evaluate the TSL's ENDF File 7 in a temperature range extending from 773 K to 1673 K. In addition, ACE type cross section libraries are produced and tested with the objective of contributing the data to the National Nuclear Data Center for inclusion in the ENDF/B-VIII database.

KEYWORDS: Molten salt, FLiBe, neutron, scattering law, nuclear reactor

1. INTRODUCTION

Neutron thermalization is the physical process that describes the evolution of the thermal neutron energy spectrum in the core of a nuclear reactor. For a "thermal" reactor it is a process that requires quantification to allow for accurate prediction of slow neutron (energy below a few eV) reaction rates, which drive the operational and safety characteristics of the reactor. In neutronic analysis, the physics of thermalization is compactly represented by the thermal neutron scattering law (TSL), which is quantified by the $S(\alpha,\beta)$ data libraries for a given material. Therefore, $S(\alpha,\beta)$ is material dependent and defines the exchange of momentum (α) and energy (β) between the atomic system and low energy neutrons until establishing the thermal spectrum. As a material property, $S(\alpha,\beta)$ is dependent on the conditions (temperature, pressure, etc.) and state of the atomic system (solid, liquid, and/or gas).

Furthermore, $S(\alpha,\beta)$ information is needed for all neutronically important materials (moderators, reflectors, fuel, etc.) in a reactor's core. Consequently, for reactors that utilize molten salt FLiBe, $S(\alpha,\beta)$ is needed for FLiBe and is currently missing from typical nuclear data sources such as the ENDF/B database. Over the past 15 years, modern methods emerged that utilize atomistic and quantum mechanical techniques for calculating the fundamental data needed in generating $S(\alpha,\beta)$ [1]. The availability of such methods has facilitated the process of TSL generation and in many cases provided first-of-a-kind libraries in support of

the needs of various communities engaged in nuclear reactor design and criticality safety analysis [2]. In addition, key technical improvements have been made in the process of calculating S(α,β) that surpassed the physics used in traditional tools such as the NJOY code [3]. This includes relaxing numerous approximations such as the incoherent approximation, the Gaussian approximation, the short collision time approximation, and the cubic approximation. In this work, classical molecular dynamics methods (MD) and the *FLASSH* platform are used to extract the fundamental information needed to evaluate the molten salt FLiBe S(α,β) along with the related inelastic thermal neutron scattering cross sections [4]. The S(α,β) data are generated as ENDF/B File 7 libraries and supplied to the US National Nuclear Data Center (NNDC).

2. Molecular Dynamics Analysis of Molten Salt FLiBe

FLiBe is a eutectic with a mixture of 2:1 ratio of LiF and BeF_2. Its melting temperature is 723 K. Therefore, it is a liquid under expected reactor operating temperatures (> 800 K). In this case, classical MD represents a highly suitable approach for extracting the generalized density of atomic states (GDOS) that is needed for S(α,β) calculations. Traditionally, the MD technique involves solving the Newtonian equations of motion for classical multi-particle systems. For FLiBe, the calculation was performed on a system that consists of 2000 Li^+, 1000 Be^{2+}, and 4000 F^- ions enclosed in a 43.67 Å edge length cubic box. The ions are randomly distributed within the box to faster reach equilibrium. The interaction potential (i.e., U) between ions is assumed to be of the Born-Mayer type as given by the sum of the following terms

$$U_1 = \frac{Z_i Z_j e^2}{r_{i,j}},\ U_2 = -\frac{c_i c_j}{r_{i,j}^{\ 6}},\ U_3 = \frac{D}{r_{i,j}^{\ 8}},\ U_4 = b_{i,j}\left(1+\frac{Z_i}{N_i}+\frac{Z_j}{N_j}\right)\exp\left(\frac{r_i + r_j - r_{i,j}}{\rho}\right), \qquad (1)$$

Where U_1 is the Coulomb potential, U_2 and U_3 are dipole-dipole potential terms, and U_4 is the Born-Mayer term. r_{ij} is the distance between ions, r_i is the radius of the ion, Z is the effective charge of an ion, N is the number of electrons in the outermost shell of the ion, and D, c, b and ρ represent potential parameters.

A system with 7000 ions was first equilibrated for 20 picosecond (ps) using conditions in which the number of atoms (N), pressure (P), and temperature (T) were held constant (i.e., NPT conditions) at multiple temperatures and zero pressure in different runs. The production phase consisted of two runs. A 200 ps equilibrium run was first conducted to determine the system properties for the corresponding temperature, i.e. density, viscosity and diffusion coefficient. A trajectory run, which persisted for 10 ps, was performed and the trajectories for each element in FLiBe was recorded to a text file for further analysis.

A snapshot of a sample in the system is shown in Figure 1. As it can be seen, the Be^{2+} and F^- ions form a tetrahedron binding structure BeF_4^{2-}, and the Li^+ ions interact with the surrounding BeF_4^{2-} clusters. By analyzing atomistic movement from the animation of atom trajectories within short time frames (< 100 femtosecond), the Be atoms are observed to be vibrating in the BeF_4^{2-} structure. Meanwhile, the F^- ions are vibrating and colliding with surrounding BeF_4^{2-} clusters. It is suggested in later analysis of the GDOS that the ionic liquid is weakly bonded and the bonds can be broken easily in comparison to the bonds in molecular liquids. Thus the exchange of F^- ions between BeF_4^{2-} clusters and the breaking and reforming of BeF_4^{2-} clusters during collisions are highly possible.

Figure 1 also shows the MD supercell that was constructed in the LAMMPS code to perform the simulations [5]. Fundamental properties of FLiBe were reproduced using the MD simulations and were found to be consistent with experimental observations [6]. The outcome of the simulations are atomic trajectories, i.e., time dependent positions and velocities of the atoms. To obtain the GDOS for each atomic species in FLiBe (F, Li, and Be), the velocity autocorrelation function for the species of interest was calculated and subjected

to a Fast Fourier Transform (FFT) operation. Figure 2 below shows the GDOS for F, Li, and Be in FLiBe. The GDOS was generated as a function of a selected temperature set, which demonstrates that the FLiBe TSL can be evaluated for the relevant temperatures of a given reactor design.

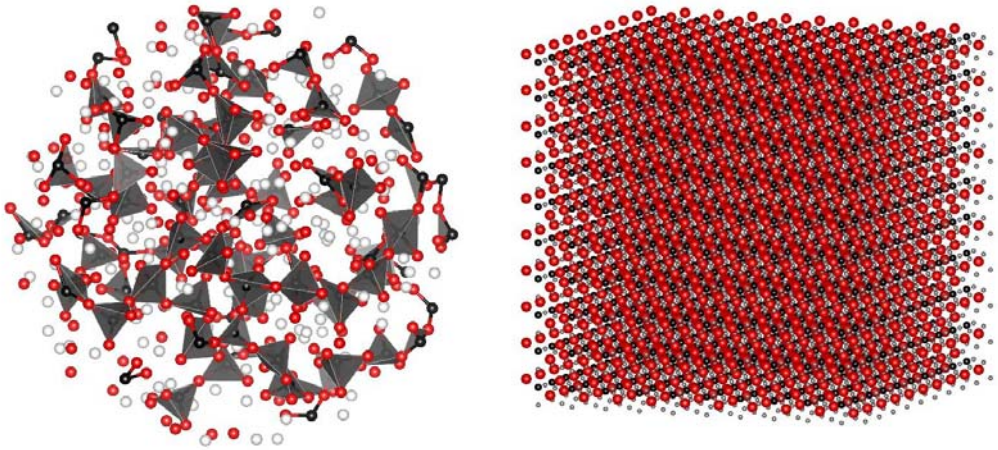

Figure 1. FLiBe MD molecular system, with black as Be^{2+} ions, red as F^- ions, and white as Li^+ ions. The BeF_4^{2-} clusters are shown as a tetrahedron structure (left). The FLiBe supercell used in the execution of the MD simulations (right).

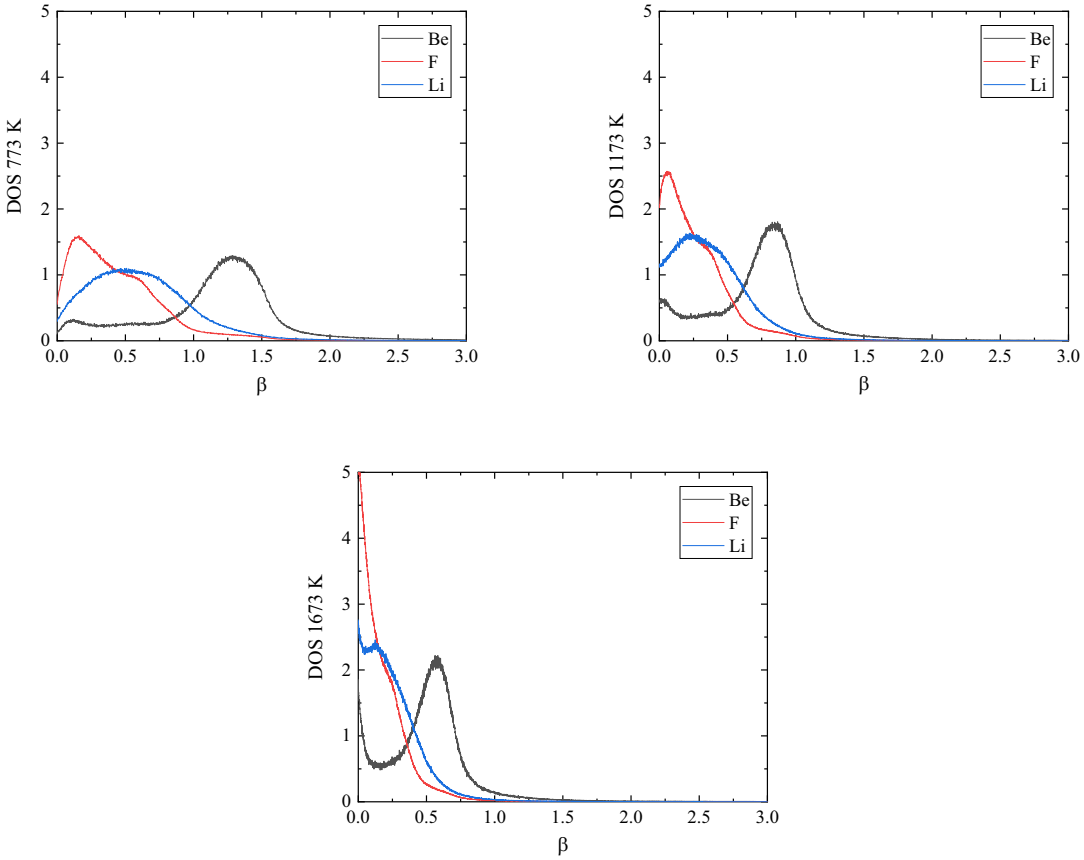

Figure 2. Total GDOS for Be, F, and Li in FLiBe at 773 K (left top), 1173 K (right top), and 1673 K (bottom).

3. Thermal Scattering Law and Inelastic Cross Sections for Molten Salt FLiBe

The thermal scattering law (TSL) for Be, F, and Li in liquid FLiBe were generated using the *FLASSH* nuclear data code [4] that is developed by the Low Energy Interaction Physics (LEIP) group at North Carolina State University. The *FLASSH* code delivers significant improvements in the theoretical treatment and user experience aspects of thermal neutron data generation. Many common approximations (e.g., incoherent, cubic, atom site, short collision time (SCT), etc.) have either been relaxed or removed completely, and a graphical user interface (GUI) is used to guide the user, with syntax and error checks, through what is traditionally a tedious setup process. The coherent elastic routine has also been generalized, allowing for the cubic or exact Debye-Waller matrix treatment of any crystal structure. Other useful features include automatic α/β gridding, a built-in conversion tool for translating *FLASSH* output to ENDF-6 format, and parallel computing capabilities using OpenMP, among others.

In addition to its ability to treat virtually any crystalline solid material, the *FLASSH* Liquid Physics module enables the processing of liquid TSLs [7]. This is made possible by a collection of liquid models (used to treat different fluid types) coupled to several processing routines. The first of these constructs the β convolution grid using physics-based bounding techniques along with an iterative normalization scheme so that the diffusive part of the TSL can be computed over this grid. Subsequently, the diffusive part is convolved with the solid TSL to give

$$S_s(\alpha,\beta) = \left(S_{\text{diffusive}}(\alpha,\beta) * S_{\text{solid}}(\alpha,\beta)\right)(\beta), \tag{2}$$

where $S_{\text{diffusive}}$, S_{solid}, and S_s represent the diffusive, solid, and total TSLs, respectively, interpolated back to the user β grid. The resulting total TSL (under the incoherent approximation) is a fundamental input in the double differential cross-section as given by

$$\left.\frac{d^2\sigma}{d\Omega dE'}\right|_{\text{inelastic}} = \frac{\sigma}{2k_B T}\sqrt{\frac{E'}{E}}S_s(\alpha,\beta), \tag{3}$$

where σ, E', E, T represent the total bound cross-section, final and initial neutron energies, and temperature, respectively. In the case of liquid FLiBe, the Schofield model was employed to represent the diffusive behavior. Temperature dependent density of states (DOS) distributions and diffusive parameters (i.e., c and d [3], which were used to extract the diffusive DOS from the total GDOS, yielding the bound DOS) were used in the input, and high phonon orders (i.e., >1000) were included to properly account for high-α contributions (due to large Debye-Waller λ values).

The TSLs, secondary distributions, and total scattering cross-sections (Figures 3 – 8) for Be, F, and Li show numerous similarities in their overall behavior and temperature dependence. With the exception of Be at higher β (Fig. 3 (right)), each of the TSLs displays a lack of structure in β space, which is indicative of FLiBe's atomic structure (or relative lack there-of). The distribution's shift from lower to higher β with increasing temperature is also observed in β space for each FLiBe constituent. In α space (e.g., Fig. 3 (left)), the upward distribution shift at high β values with increasing temperature reverses at very low β values (e.g., $\beta = 0$ (overlapped by $\beta = 1 \times 10^{-9}$) and $\beta = 1 \times 10^{-9}$). This is consistent with the higher probability of quasi-elastic (very low β) interactions at lower temperatures. Each of the secondary distributions (e.g., Fig. 4 (left)) display sharp quasi-elastic peaks, but are otherwise relatively featureless (again, with the exception of Be). As the neutron's incident energy increases, the distributions convey an increase in the down-scattering probability and a decrease in the up-scattering probability. Furthermore, as temperature increases, the probability of quasi-elastic interactions decreases in favor of those characterized by larger energy exchanges. The total scattering cross-section plots (e.g., Fig. 4 (right)) display a strong temperature dependence at lower energies, and the spread of values taken at these energies across the entire temperature range is ordered greatest to least as Be, F, and Li, respectively.

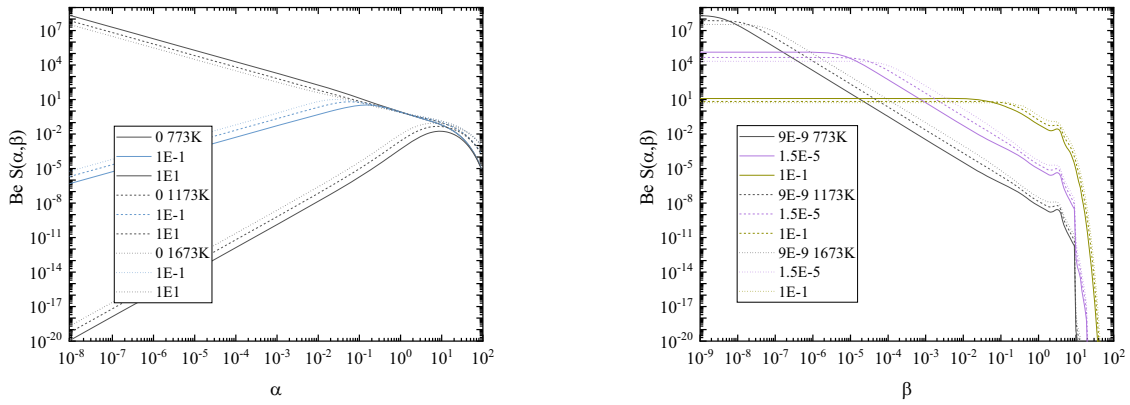

Figure 3. TSL vs. α (left, for various β values) and TSL v. β (right, for various α values) for Be in FLiBe at 773 K, 1173 K, and 1673 K.

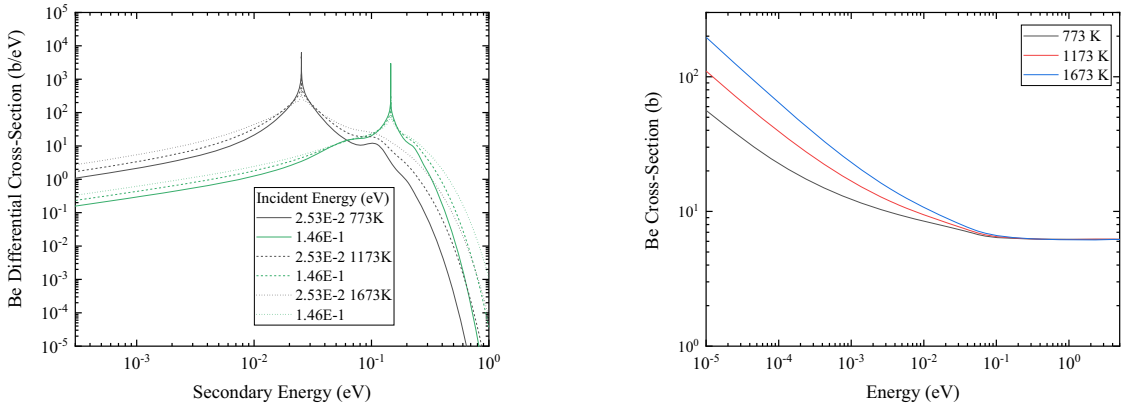

Figure 4. Secondary distribution (left, for various incident energies) and total scattering cross-section (right) for Be in FLiBe at 773 K, 1173 K, and 1673 K.

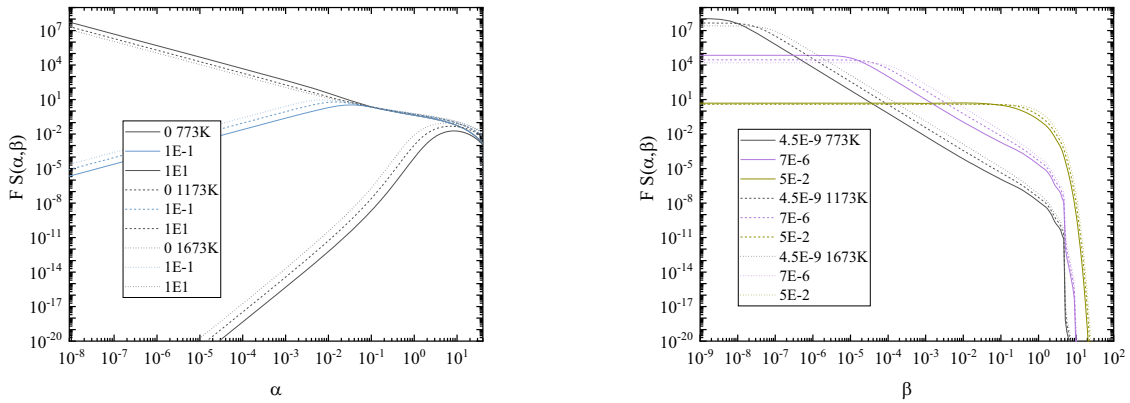

Figure 5. TSL v. α (left, for various β values) and TSL v. β (right, for various α values) for F in FLiBe at 773 K, 1173 K, and 1673 K.

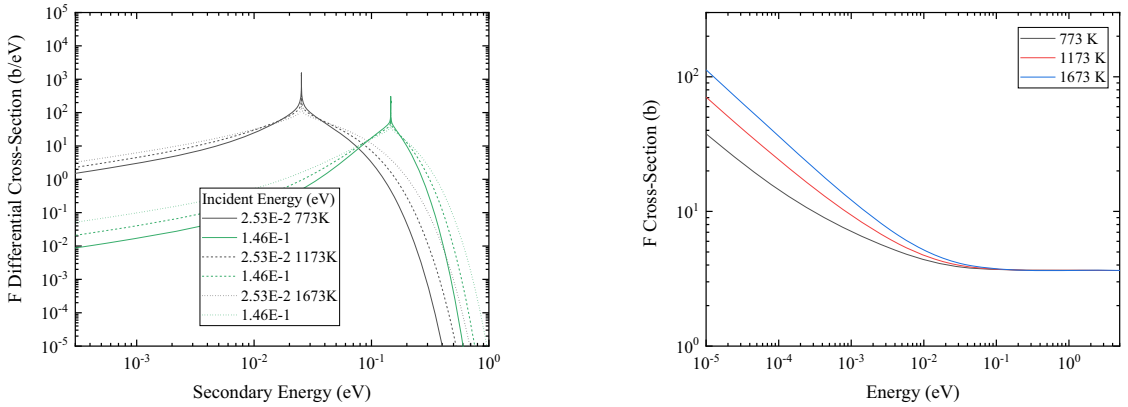

Figure 6. Secondary distribution (left, for various incident energies) and total scattering cross-section (right) for F in FLiBe at 773 K, 1173 K, and 1673 K.

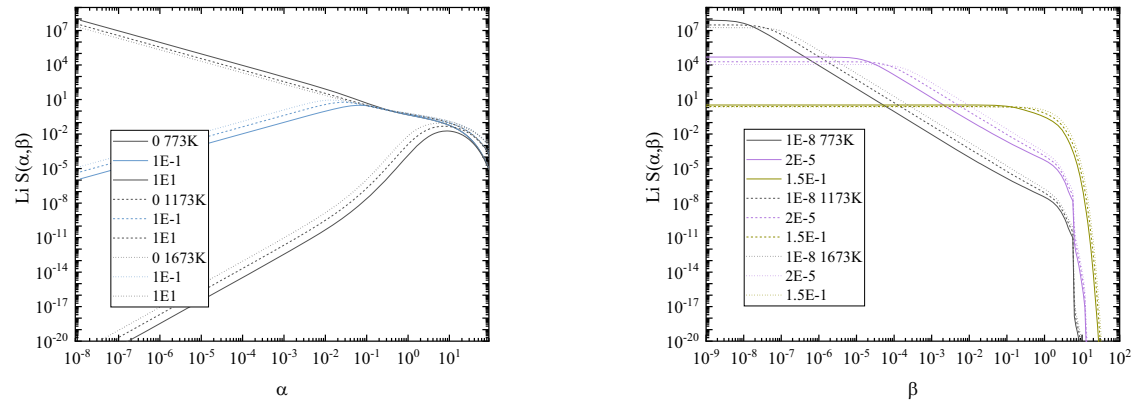

Figure 7. TSL v. α (left, for various β values) and TSL v. β (right, for various α values) for Li in FLiBe at 773 K, 1173 K, and 1673 K.

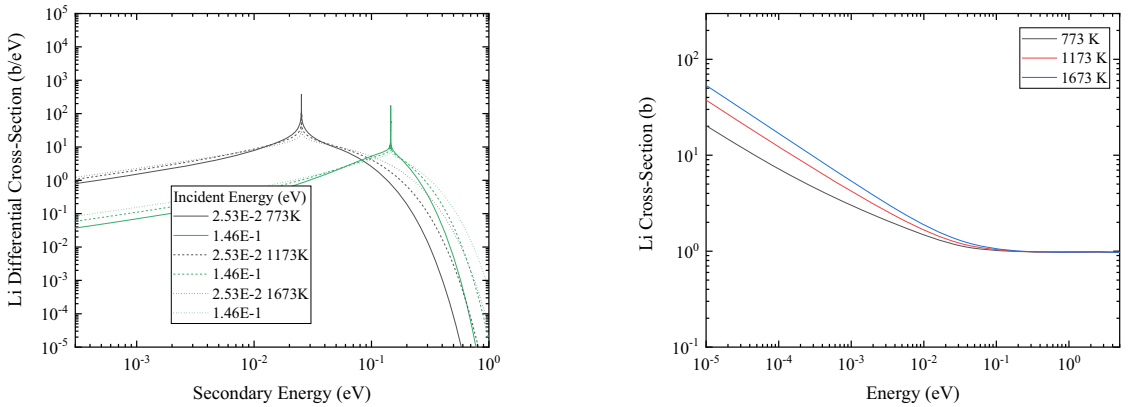

Figure 8. Secondary distribution (left, for various incident energies) and total scattering cross-section (right) for Li in FLiBe at 773 K, 1173 K, and 1673 K.

The above TSL data, e.g., appearing in Figures 3, 5 and 7 has been generated in the ENDF/B File 7 format and is currently being processed for contribution to the US National Nuclear Data Center (NNDC). Furthermore, an effort to validate this data is underway by examining available experiments such as presented in the molten salt reactor experiment that was performed at Oak Ridge National Laboratory (ORNL) [8], which is currently under consideration as a benchmark by the International Reactor Physics Experiment Evaluation (IRPhE) Project as the MSRE-MSR-EXP-001 benchmark [9,10].

4. CONCLUSIONS

The thermal scattering law (TSL), i.e., $S_s(\alpha,\beta)$, was evaluated for F, Li and Be in molten salt FLiBe in a temperature range extending from 773 K to 1673 K. Classical molecular dynamics techniques, as implemented in the LAMMPS code, were found suitable for generating the GDOS of each species in FLiBe. The GDOS was used in the *FLASSH* code to evaluate the TSL and generate the thermal neutron inelastic scattering cross sections. FLiBe File 7 data is undergoing quality assurance checks and is in the process of being contributed to NNDC.

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