

ENDF/B-VI CHLORINE EVALUATION IS DEFICIENT

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ABSTRACT

The criticality safety evaluations for the Fuel Cycle Facility Electrorefiner [1] at Argonne-West were reviewed at Oak Ridge National Laboratory (ORNL) to help provide insight into problems that may be caused by inadequate cross-section data. The adequacy of ENDF/B chlorine was questioned because the evaluation was done in 1967 and is a nonresonance material even though chlorine has resonance structure. There are no validation experiments which are similar to the system being analyzed. Our analysis strongly suggests that the ENDF/B-VI data for natural chlorine, MAT 1700, are not adequate for all criticality safety applications and must be considered to be deficient for this reason. This conclusion was reached by comparing several different XSDRNPM calculations using the ENDF/B chlorine evaluation with the same calculation using the JENDL-3.2 chlorine evaluation. All the other cross sections in these calculations are taken from ENDF/B-VI; only the chlorine cross sections were changed.

XSDRNPM CALCULATIONS

The calculations are for Li/K/Cl salt with different concentrations of ^{235}U fuel, expressed in g/L of salt. The salt used in these calculations was 6 wt % Li, 22 wt % K, and 72 wt % Cl. The ^{235}U fuel concentrations vary from 20 to 5,000 g/L. Three series of calculations were done using

NAT-Li/K/Cl salt, Li-7/K/Cl salt, and Li-7/Cl salt as mixtures of lithium chloride and potassium chloride. The XSDRNPM calculations using the 199-group VITAMIN-B6 library [2] and the infinite homogeneous medium option were done at 773 K. As mentioned earlier, two sets of calculations were done, with ENDF/B-VI cross sections and with ENDF/B-VI except for the JENDL-3.2 chlorine. The k_{∞} using JENDL-3.2 chlorine varied from +15% to -7% relative to the XSDRNPM calculation using ENDF/B-VI chlorine. The behavior is due to difference in the chlorine cross sections because the other cross sections were from ENDF/B-VI. We will show that the differences are primarily due to the chlorine absorption cross sections.

Results using the JENDL-3.2 natural chlorine evaluation are compared with those using ENDF/B-VI in Figs. 1-3. Figure 1 demonstrates that the JENDL-3.2 calculations are several percent lower than the ENDF/B-VI calculations for fuel concentrations above 100 g/L. For fuel concentrations less than 100 g/L, the calculated k_{∞} values are less than 1. For the 20 g/L case the calculated k_{∞} is only about 0.21; the reason for this low value is a combination of the low fuel concentration and ^6Li absorption in the Nat-Li salt. Changing from Nat-Li/K/Cl to ^7Li /K/Cl has a pronounced effect on the calculated k_{∞} values for fuel concentrations less than 100 g/L as shown in Fig. 2. It is interesting to note that the JENDL-3.2 calculations are

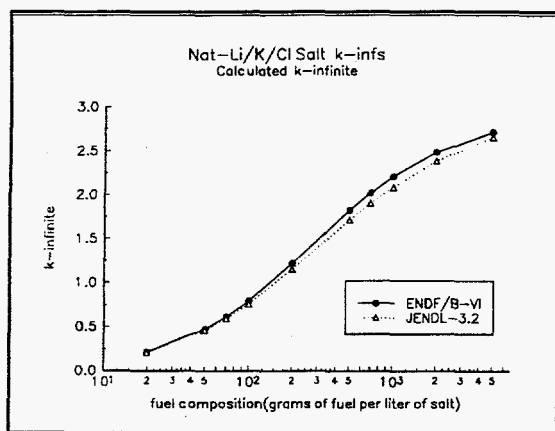


Figure 1. Nat-Li/K/Cl salt k_{∞}

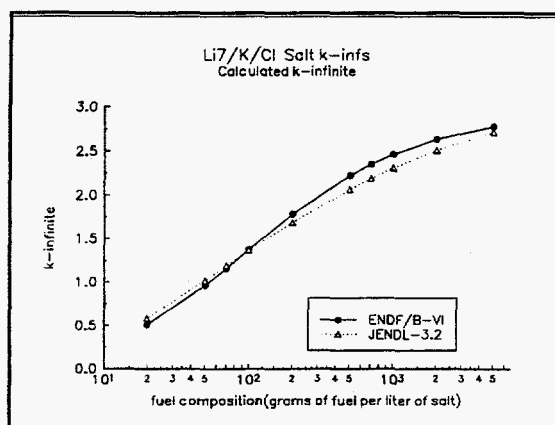


Figure 2. ${}^7\text{Li}/\text{K}/\text{Cl}$ salt k_{∞}

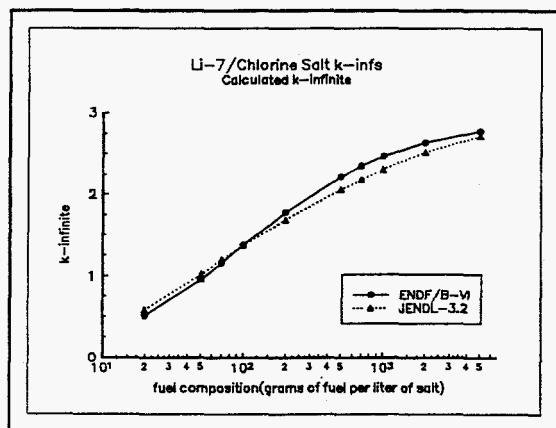


Figure 3. ${}^7\text{Li}/\text{Cl}$ salt k_{∞}

higher than ENDF/B-VI below 100 g/L but lower than ENDF/B-VI above 100 g/L. This behavior is a direct result of differences in the chlorine absorption cross sections. Figure 3 shows calculated k_{∞} values for the ${}^7\text{Li}/\text{Cl}$ cases; results show nearly the same trend as that seen for the ${}^7\text{Li}/\text{K}/\text{Cl}$ cases in Fig. 2. The percent differences in k_{∞} for each of the various cases are shown in Fig. 4; the differences vary from +15% to -7%.

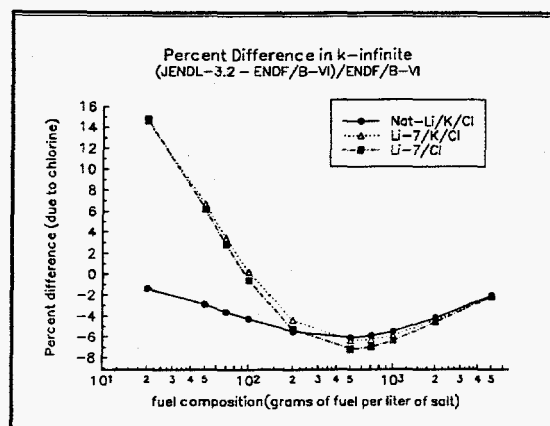


Figure 4. Percent difference in k_{∞}

In addition to the calculated k_{∞} values, we also compare the average energy of fission for the JENDL-3.2 and ENDF/B-VI calculations in Figs. 5 through 7. In Fig. 5, we see that the average energy of fission using JENDL-3.2 is higher for cases with fuel concentrations above 100 g/L. At 1,000 g/L the JENDL-3.2 value is 332 keV, compared to 299 keV with ENDF/B-VI, a difference of 11%. A similar difference is also seen in Fig. 6 with a difference of 11.3% at 1,000 g/L, and in Fig. 7, with a difference of 12.2% at 1,000 g/L. It is also interesting to note the rather large difference at 20 g/L in Figs. 6 and 7. This was not seen in Fig. 5 due to the effect of the ${}^6\text{Li}$ absorption. The spectra for the ${}^7\text{Li}$ cases using the JENDL-3.2 chlorine is much softer than for the ENDF/B-VI cases as demonstrated by the average energy of fission. Again, the explanation for this is

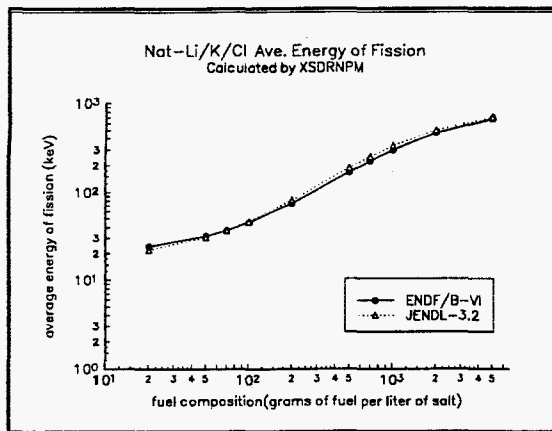


Figure 5. Nat-Li/K/Cl average energy of fission

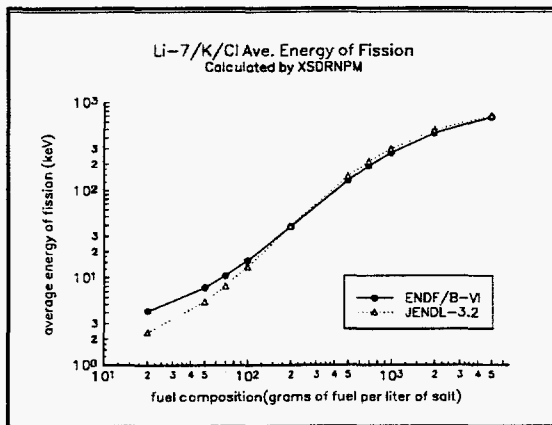


Figure 6. $^7\text{Li}/\text{K}/\text{Cl}$ average energy of fission

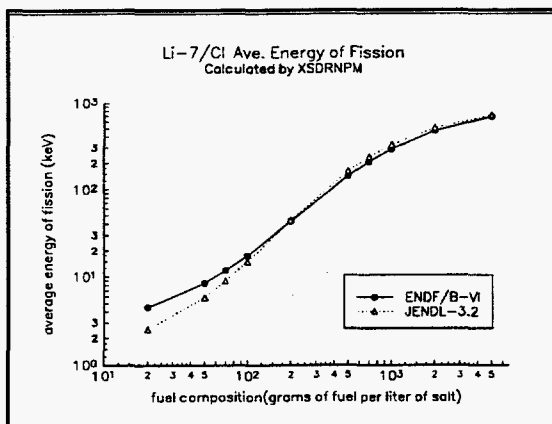


Figure 7. $^7\text{Li}/\text{Cl}$ average energy of fission

due to differences in the chlorine absorption cross sections.

DISCUSSION OF RESULTS

The JENDL-3.2 total and capture cross sections are compared with ENDF/B-VI in Figs. 8 and 9. The total cross sections are rather similar except that the ENDF/B-VI evaluation has structure not seen in JENDL-3.2 between 1 and 10 keV and between 230 keV and 1 MeV. The capture cross sections are nearly the same up to 10 eV and differ greatly above 10 eV. Figure 10 compares the capture cross sections above 100 eV. The JENDL-3.2 evaluation has MLBW resonance parameters for the resolved resonance range up to 230 keV. ENDF/B-VI has only pointwise cross sections and does not have resonance parameters. The capture and total cross sections in ENDF/B-VI are not consistent and the capture evaluation only represents the average cross section. The difference between the JENDL-3.2 and ENDF/B-VI capture cross sections is quite large as shown in Fig. 10. The ENDF/B-VI average capture is higher between 100 eV and 100 keV and much lower above 100 keV. It should be noted that resonance self-shielding cannot be done for ENDF/B-VI since the evaluation for the capture cross section does not represent the resonance structure. This is a serious limitation in the ENDF/B-VI evaluation.

We expected that the difference in chlorine capture cross sections would explain the differences in the calculated k_{∞} values shown in Figs. 1 through 3. However, we discovered that only changing the chlorine capture did not remove the differences in the calculated k_{∞} values. Additional checking revealed that the (n,p) cross sections were also different by a significant amount, as shown in Fig. 11. The (n,p) is larger than the capture cross section above about 300 keV and is thus more important than capture for the cases with higher fuel

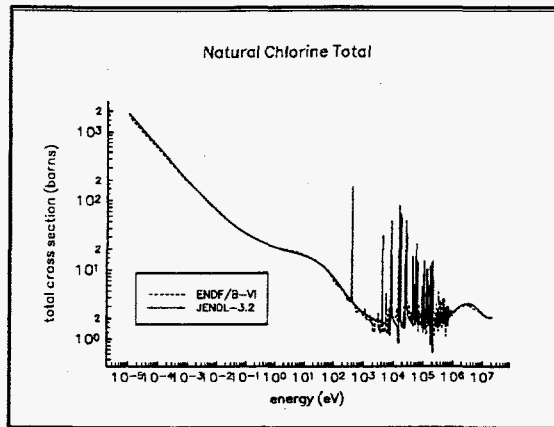


Figure 8. Natural chlorine total

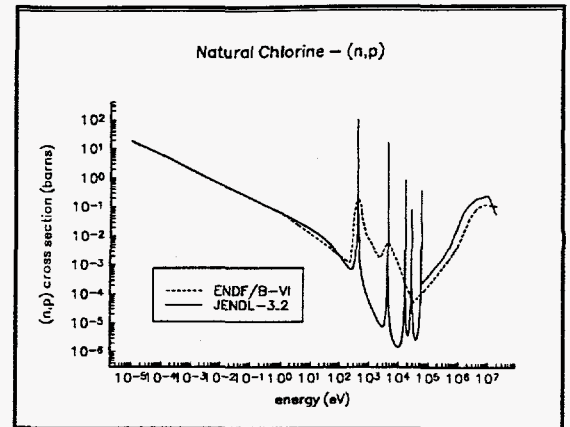


Figure 11. Natural chlorine (n,p)

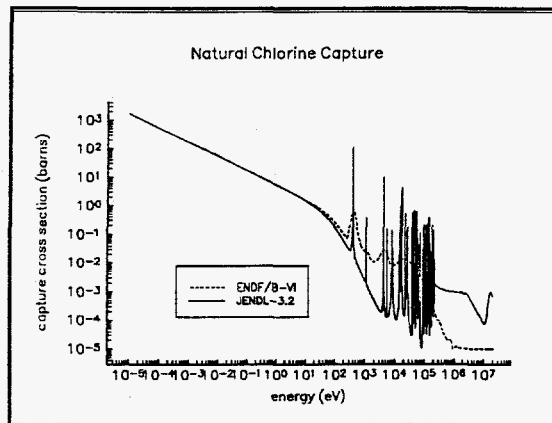


Figure 9. Natural chlorine capture

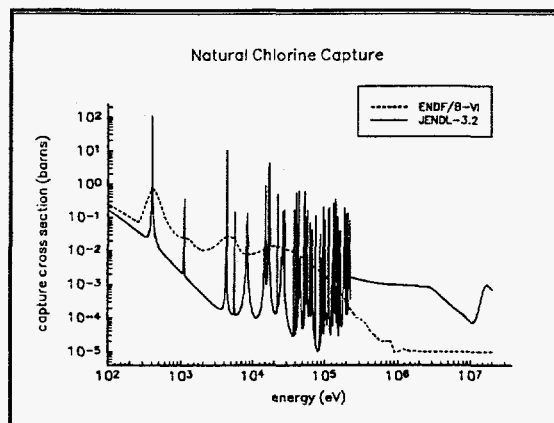


Figure 10. Natural chlorine capture

concentrations. In order to account for most of the difference in the calculated k_{∞} values, both the chlorine capture and (n,p) cross sections must be changed.

So far we have shown that there are large differences in k_{∞} for the calculations using the JENDL-3.2 evaluation, relative to the ENDF/B-VI evaluation. These differences are largely a result of differences in the two absorption cross sections. Thus we need to address the question of which of the two evaluations agree best with the measured data. As discussed in the previous paragraph, we need to look at both the capture (n, γ) and (n,p) reactions. In the energy range below 230 keV the JENDL-3.2 capture cross section is calculated from MLBW resonance parameters. The resonance parameters are based on the work of R. L. Macklin [3] and Mughabghab [4]. In the energy range below 230 keV, the JENDL-3.2 capture is definitely an improvement over ENDF/B-VI. Above 230 keV, there is much less to go on. We have measured data for ^{37}Cl capture but no measured data for either ^{35}Cl or natural chlorine. The JENDL-3.2 capture was calculated with the optical and statistical model code CASTY. The ^{37}Cl capture cross section was adjusted to agree with measured data of A. G. Dovbenko [5]. The magnitude of this adjustment is not given in the

JENDL-3.2 FILE 1 comments section and is thus unknown. The Dovbenko measurements span the energy range 150 keV to 2.1 MeV. For the energy range 300 keV to 2 MeV, the ^{35}Cl capture is from about 1.5 to 2.4 times higher than the ^{37}Cl capture. The natural chlorine capture cross section is obtained by adding the two contributions after multiplying by the isotopic abundances. For the energy range from 300 keV to 2.1 MeV, the chlorine capture is about 0.9 times the ^{35}Cl capture. In the absence of measured data, it is difficult to estimate the uncertainty of the JENDL-3.2 capture, but the calculations may be good to within about 50%. The difference between the JENDL-3.2 evaluation and ENDF/B-VI is on the order of a factor of 100 for the energy range 300 keV to 2.1 MeV; thus it would appear that the JENDL-3.2 evaluation is definitely better. Based on these arguments, it would be expected that the JENDL-3.2 chlorine capture cross section is better and should give better results for the k_{∞} calculations considered in this study. As noted previously, the chlorine (n,p) reaction is very important in the determination of the calculated k_{∞} for those cases with higher fuel concentrations. The chlorine (n,p) cross sections are compared in Fig. 11. The JENDL-3.2 (n,p) cross section is 2 to 4 times larger than the corresponding ENDF/B-VI value between 0.1 and 3 MeV. There is a rather limited amount of experimental data for the (n,p) reaction. The ^{37}Cl (n,p) threshold is 4.182 MeV, so this reaction is not important for the work considered in this paper. The natural chlorine (n,p) cross section below 4.182 MeV is entirely due to ^{35}Cl . There are very few measurements of the ^{35}Cl (n,p) cross section in the energy range of interest for this work (0.1 to 3 MeV). Additional measurements in this energy range would be useful for criticality safety applications.

MODIFIED CHLORINE EVALUATION

A modified ENDF/B-VI evaluation was generated in the course of this work to investigate the impact of changing both the capture and the (n,p) cross sections in order to demonstrate that these reactions are the primary cause of the differences in k_{∞} between the ENDF/B-VI and JENDL-3.2 libraries. The capture and (n,p) cross sections were changed to be nearly the same as those in the JENDL-3.2 library. Calculations with this modified evaluation are compared with the original ENDF/B-VI and JENDL-3.2 in Fig. 12.

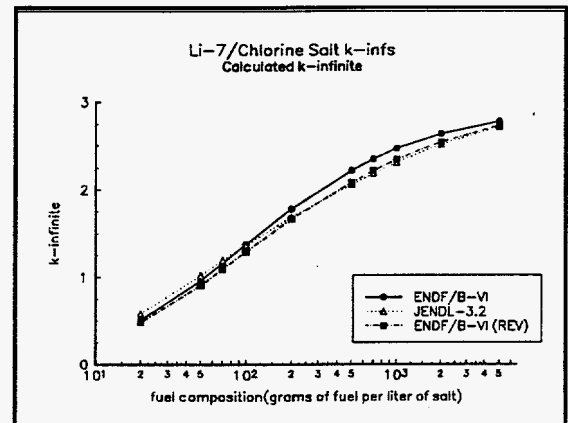


Figure 12. $^7\text{Li}/\text{Cl}$ salt k_{∞}

CONCLUSIONS

The lack of resolved resonance parameters is a serious limitation in the ENDF/B-VI evaluation. Also the ENDF/B-VI capture above 100 keV is not in agreement with measured data. The JENDL-3.2 (n,p) cross section is 2 to 4 times higher than ENDF/B-VI between 0.1 and 3 MeV. Based on the calculations and analysis in this study, we conclude that the ENDF/B-VI chlorine evaluation is deficient for intermediate and fast systems. The ENDF/B-VI chlorine evaluation is inadequate and a revised evaluation is definitely needed to meet

criticality safety needs. Evaluations for ^{35}Cl and ^{37}Cl , rather than natural chlorine, would be preferred.

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