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IMPROVED S(α, β) TABLES FOR TRIGA CRITICALITY AND REACTIVITY FEEDBACK CALCULATIONS

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ABSTRACT

The accurate evaluation of the reactivity feedback in TRIGA cores gained additional importance within the TRADE project. In this sub-critical system coupled with an external source the operational temperatures could reach 150^0 C and more. The suggested reactivity feedback curve provided by GA (GENERAL ATOMICS) which is sufficient for core shut down analysis appears to be inaccurate in comparison with new experimental data, in particular in the range of 80-140^0 C. The hydrogen bound in zirconium within the TRIGA fuel is the main cause for the negative reactivity safety mechanism. An immediate neutron up-scattering by hydrogen as a result of a fuel temperature increase shifts the neutron spectrum to lower fission cross-section energy range. The up-scattering of neutrons is governed by the scattering kernel which is a measure of the probability of a neutron to be scattered from its incident energy to another one and from its original angular direction to a new one after a collision. Those probabilities also known as the scattering law are presented within probability tables based on so call S (α, β) formalism which should conserve the energy and momentum laws. For bound isotopes like hydrogen in zirconium, the structure of the lattice and the intermolecular forces influence immensely the scattering probability.

The current study looks at various models and parameters concerning the generation of the molecule structure dependent S(α, β) tables and their impact on the reactivity feedback temperature coefficient of the TRIGA fuel. It is shown that different computational handling of the models for internal forces within the ZrH molecule in particular for the so called phonon spectrum description leads to variation of the reactivity feedback as well as the criticality value. The influence of adopting a new scattering kernel on the fuel pin criticality value is also presented for the first 8 S resonances of U238 within the TRIGA fuel pin.

KEYWORDS: S (α, β) scattering law, cross-section, hydrogen bound in zirconium hydride, reactivity feedback

1. INTRODUCTION

In the TRiga Accelerator Driven Experiment TRADE [1], it was foreseen to couple an external proton beam with a tantalum target in the central part of the core of the TRIGA reactor RC-1 Cassacia. An important objective of the project is to gain experience with the coupling of a powerful proton accelerator with a sub-critical system at power level which provides sufficient negative feedback effects. Therefore, it is important to evaluate accurately the temperature

dependent reactivity feedback at higher temperature (100-160^0 C). In particular reactivity measurements, which were performed in RC-1 [1], and the Slovenian TRIGA core[3], exhibited a large increase of the negative reactivity feedback at 80-140^0 C. This phenomenon of increased negative worth is not explicitly addressed in the TRIGA GA technical report [4] where only a smooth curve is presented, which fits broad temperature steps [5]. In the current study the sensitivity of the probability S(α, β) tables to the basic structure models of hydrogen bound in zirconium hydride is discussed for temperature step of 10 degree Celsius. Furthermore, the influence of the computational scheme, which process the basic structure data to MCNP compatible input file is analysed.

The scattering kernel treatment of heavy isotopes in the vicinity of its resolved resonance range was recently improved [6] and resonant dependent S(α, β) tables for U238 were generated, which replace the approximated solution in MCNP [7]. For the unique TRIGA fuel pin this effect is expected to be mitigated compared to LWR fuel pins (due to the reduced amount of U238 in the fuel). The criticality changes are presented in comparison with other parametric analysis.

2 Basic models for thermal neutron scattering files

2.1 Free-Gas Approximation

In the common expression for the temperature dependent double differential scattering kernel it is assumed that the nuclide behaves like an ideal gas as far as its thermal agitation is concerned. Using the Maxwell Boltzman distribution for the spectrum velocities of the specific nuclide one obtains the following expression:

$$\frac{d^2\sigma}{d\Omega dE'}(E \to E', \Omega \to \Omega') = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}}e^{-\frac{\beta}{2}}S(\alpha,\beta) \qquad (1)$$

where E and E$'$ are the incident and the secondary neutron energies in the laboratory system, Ω and Ω$'$ the corresponding scattering angle in the laboratory system, σ_b is the bound scattering cross section for the material, k_BT is the temperature in eV,

S(α, β) is the thermal scattering law and has the form:

$$S(\alpha,\beta) = \frac{\exp\left[-(\alpha^2+\beta^2)/(4\alpha)\right]}{2\sqrt{\pi\alpha}} \qquad (2)$$

The scattering law depends on only 2 variables:

- the momentum transfer κ

$$\alpha = \frac{E' + E - 2\sqrt{E'E}\cos\theta}{Ak_BT} = \frac{\hbar^2\kappa^2}{2Mk_BT}$$

where A is the ratio of the mass of the scattering atom M to the neutron mass, and,

- the energy transfer ε

$$\beta = \frac{E' - E}{k_BT} = \frac{\varepsilon}{k_BT}$$

2.2 Approximation for bound isotopes in molecules

For light isotopes which are bound in molecules the ideal gas assumption is not accurate and could lead to erroneous parametric evaluations. The reason lies in the fact that at thermal energies (mainly below ~1 eV) the scattering is strongly affected by the binding of the scattering nucleus in its solid, liquid or gas molecule. The interacting neutron can give up energy to excitations in the material, or it can gain energy so these effects change the reaction cross section and they modify the energy and the angle distribution of the scattered neutrons. Thereafter, the ideal gas thermal agitation of the target nucleus is replaced by a molecule structure dependent vibration distribution. For hydrogen bound in zirconium hydride it could be assumed that the hydrogen atom vibrates harmonically about its equilibrium state. If the vibration is isotropic it could be characterized by single frequency ω . This type of quantized harmonic lattice vibrations is commonly called in solid state physics phonons. Slaggie [8] showed however that the molecule structure of the hydrogen bound in zirconium is better presented by more frequencies as there are several different forces acting on the bound hydrogen. This led to his suggested phonon spectrum. A modified suggestion to the phonon spectrum distribution was recently published by IKE [Institut fur Kern Energy Stuttgart] [9] and shows a better agreement with regard to spectral experiments. The new generalized spectrum fits to several lattice structures dependending on the hydrogen content of ZrH_X where x varies from 1,5 to 2. The two mentioned phonon spectrums are plotted in Fig 1. The IKE curve has a well pronounced Gaussian peak about 0.137 eV. This is in accordance with the neutron total cross section characteristic of hydrogen bound in zirconium hydride based on Whittemore [10]. Such neutrons can gain or lose energy in amounts which are multiples of energies lying in a band centred roughly around 0.137 eV which in return fits the force constants of the ZrH lattice structure.

As the hydrogen - hydrogen interacting forces are neglected the splitting observed in the Slaggie curve in Fig. 1 do not exist for the IKE curve. The two spectrum curves denoted as $\rho(\beta)$ can be inserted in the scattering function which is being calculated within the LEAPR module [11]. The LEAPR module of NJOY [12] is used to prepare the scattering law S (α, β) and related quantities, which describe thermal scattering for bound moderators (in this case hydrogen and zirconium bound in zirconium hydride), in the ENDF-6 format used by the THERMR module of NJOY. For hydrogen bound in zirconium the phonon expansion model is used where its scattering function is being calculated using appropriate α, β values and a chosen frequency spectrum $\rho(\beta)$, in this study the two curves of Fig.1. All the above mentioned parameters are being supplied in an input file for NJOY module LEAPR.

The S(α, β) scattering function for solid-type frequency distribution is calculated using the phonon expansion method :

$$S_S(\alpha,\beta) = e^{-\alpha\lambda_S} \sum_{n=0}^{\infty} \frac{1}{n!}\alpha^n x \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\beta\hat{t}} [P_S(\beta') e^{-\beta'/2} e^{-i\beta'\hat{t}} d\beta]^n d\hat{t} \quad (3)$$

The frequency spectrum $\rho(\beta)$ is part of the P_s function in the above equation:

$$P_s(\beta) = \frac{\rho(\beta)}{2\beta \sinh(\beta/2)}$$

and the Debye-Waller coefficient is :

$$\lambda_s = \int_{-\infty}^{\infty} P_S(\beta) e^{-\beta/2} d\beta$$

For energy transfers above ~ 1.8 eV and up to the thermal cut-off energy (<5 eV) the neutron interaction may be treated within the short-collisions-time (SCT) approximation as a free gas of the scattering nuclei with an effective temperature T_{eff} [11]. This extension of the S(α, β) tables is done in THERMR module of NJOY in the calculation of the double differential neutron scattering cross sections.

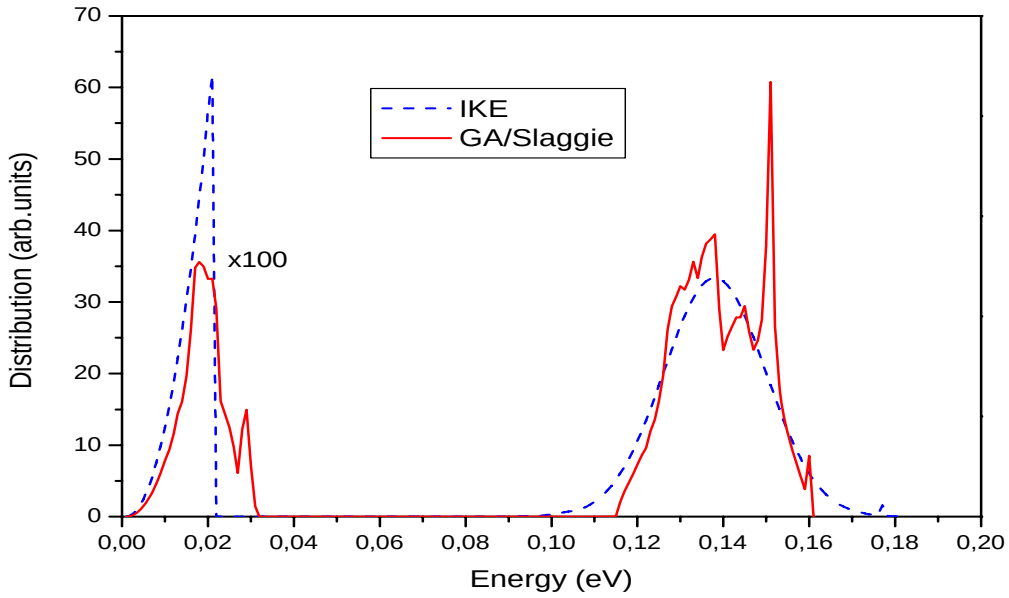

Figure 1: Frequency distribution of hydrogen bound in zirconium hydride

2.3 Improvement of resonance dependent scattering kernel for heavy isotopes

The Doppler Effect contributes about 20-25 % to the overall negative reactivity. The development of $S(\alpha,\beta)$ tables for isotopes with pronounced resonances in particular U238 [13] enables the replacement of the approximated solution of MCNP ("Sampling the target velocity"), This existing scattering kernel treatment in MCNP is actually based on the solution of Eqs 1 and 2. In this free gas model the scattering cross section is assumed to be constant which is a reasonable assumption for light isotopes but quite poor in the vicinity of the pronounced resonances of heavy isotopes in particular U238. The implementation of the new kernel in NJOY [14] in addition to the necessary modifications in the MCNP code allowed for the generation of appropriate U238 $S(\alpha,\beta)$ tables for TRIGA fuel pin criticality calculation.

3. Experimental results

The interest in accurate reactivity feedback coefficients within the TRADE project led to reactivity measurements in the Cassacia RC-1 core [2]. In addition the results of experimental temperature dependent criticality values were published for the Slovenian TRIGA core [3]. The investigated TRIGA fuel elements are cylinders of ternary alloy uranium-zirconium-hydride with H-to-Zr atom ratio 1.7 and the total U 8,5% of the mixture by weight for Cassacia reactor and with H-to-Zr atom ratio 1.65 and the total U 12% for Slovenian reactor and the same enrichment (20% enriched ^{235}U). The fuel cladding consists of stainless steel AISI 304 of 0.05 cm thickness and 7.8 g/cm^3 density. There are two graphite cylinders of 38.11 cm length at the top and the bottom of the fuel rod.

The reactivity measurements in RC-1 were carried out with the existing core configuration, i.e. with burned-up fuel. The measurements at the TRIGA Mark II at Slovenia were performed with completely fresh uniform core. In both experiments only one or two temperature measurement points were introduced from which an average temperature was evaluated. In Figure 2 the reactivity feedback per degree is plotted. The curves are subjected to uncertainties of about 3 degrees in temperature and about 2 pcm in the reactivity. All curves differ considerably from the GA curve between 110 -150^0 C. Two curves of the Slovenian core refer to the estimation of the total core averaged temperature based on measurements at point A located in the centre of the core (denoted in the figure with T_a). The radial form coefficient for T_a is C_a=1.26. The coefficient C_z is the axial form factor. The sensitivity to the form factor is realized from looking at the evaluations of changing the axial form factor by about 10% from 1.25 to 1.12. The other two curves of the Slovenian core are based on two point measurements at the above centre point (T_a) and in the first ring of the core (T_b) for which the radial form coefficient is C_b=1.22. The sensitivity to the axial form factor is again demonstrated. The differences between the curves emphasizes the necessity of a multi-point temperature measurement to assess more accurately the importance of the spatial flux distribution and the fuel content (fresh or burned up fuel) on the reactivity feedback.

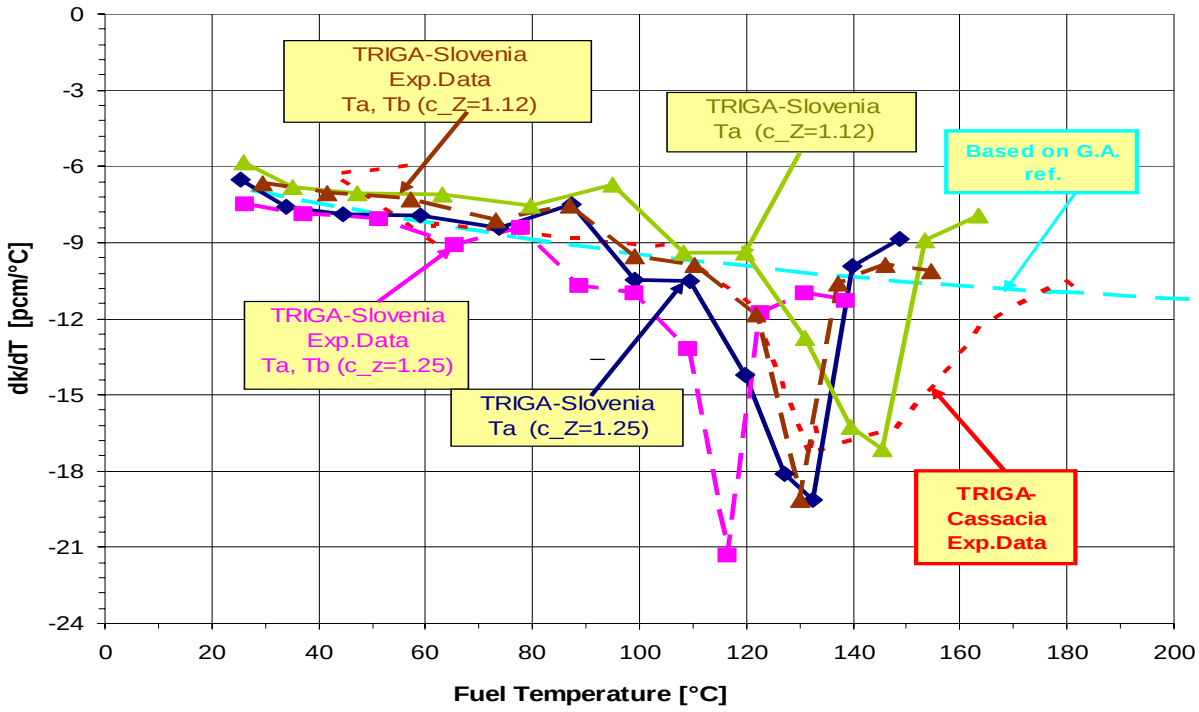

Figure 2: Experimental temperature dependent reactivity feedbacks of the Slovenian and the Cassacia TRIGA cores

4. The effect of different structure lattice models

In NJOY the THERMR module utilizes the LEAPR output. This output is based on the chosen phonon spectrum model and upon the number and range of α and β values which define also the energy limit from which the short collision approximation replaces the phonon expansion method.

The THERMR module generate then $S(\alpha,\beta)$ tables for a standard set of 59 standard energy points which range from almost 0 to about 4.6 eV. In the following table (1) the effects of the different models and different input parameters are presented. The tolerance value in the table refers to all relevant NJOY modules: BROADR, RECONR, THERMR.

Table 1. Parameters and lattice structure model for 4 analyzed probability tables.

Case	Phonon Spectrum Model	Energy grid	Tolerance	Cosine bins	Equi-probable energies interval
A	GA/Slaggie	59	0.005	8	20
B	GA/Slaggie	118	0.005	8	20
C	GA/Slaggie	118	0.001	8	64
D	IKE	79	0.001	16	64

In figure 3 the total cross section of the bound hydrogen in ZrH is plotted for the above four cases. The reference case A differs strongly already at lower energies. Above 0.55 eV only a refined energy point interpolation like cases C and D maintain the oscillating form. Yet one should keep in mind that experimental cross section evaluation [10] confirms cases C and D only up to about 0.55. Above this energy it is not clear if the intermolecular forces that act on the hydrogen are intact.

Figure 4 depicts the influence of the several methods on the criticality. The estimated criticality of case A is lower by ~300 pcm compared to the cases with higher energy grid number. This result emphasizes the need to increase the 59 standard energy point grid in the THERMR module. Figure 5 is basically the derivative of figure four and is particular interesting because it can be compared to some extent to the experimental values shown in Figure 2. All the models show an enhanced negative feedback in the 80 -140^0 C temperature range. In Cases C and D the enhanced negative values are moderate which might be attributed to the oscillating cross section above 0.55 eV. In the lower temperature range cases C and D are qualitatively and to some extent quantitatively closer to the experimental results. The difference of case C in comparison to case D is greater then the statistical error and could be attributed to the different phonon curves and to the enlarged equal-probable cosine and energy bins. Yet more accurate experiments accompanied by full fitted calculation should be done to evaluate those differences.

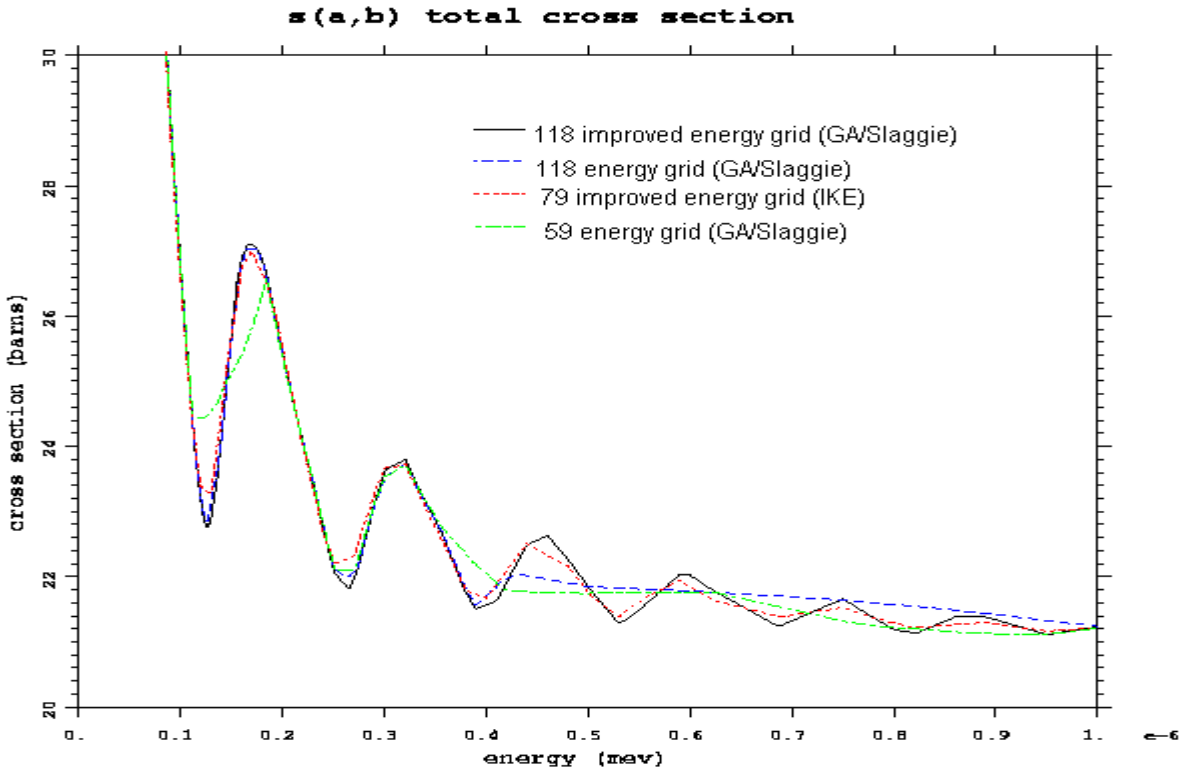

Figure 3: Total S(α, β) cross section of hydrogen bound in zirconium-hydride as obtained by MCNP.

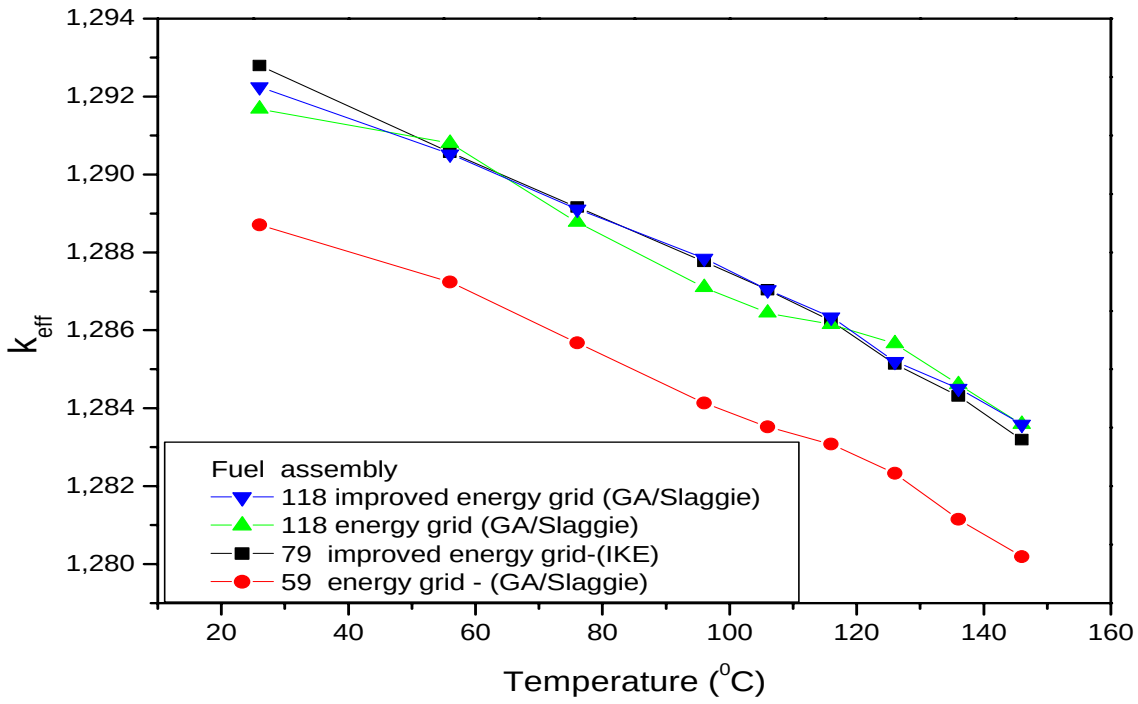

Figure 4: MCNP calculated criticality values using four different $S(\alpha, \beta)$ input files for hydrogen bound in ZrH.

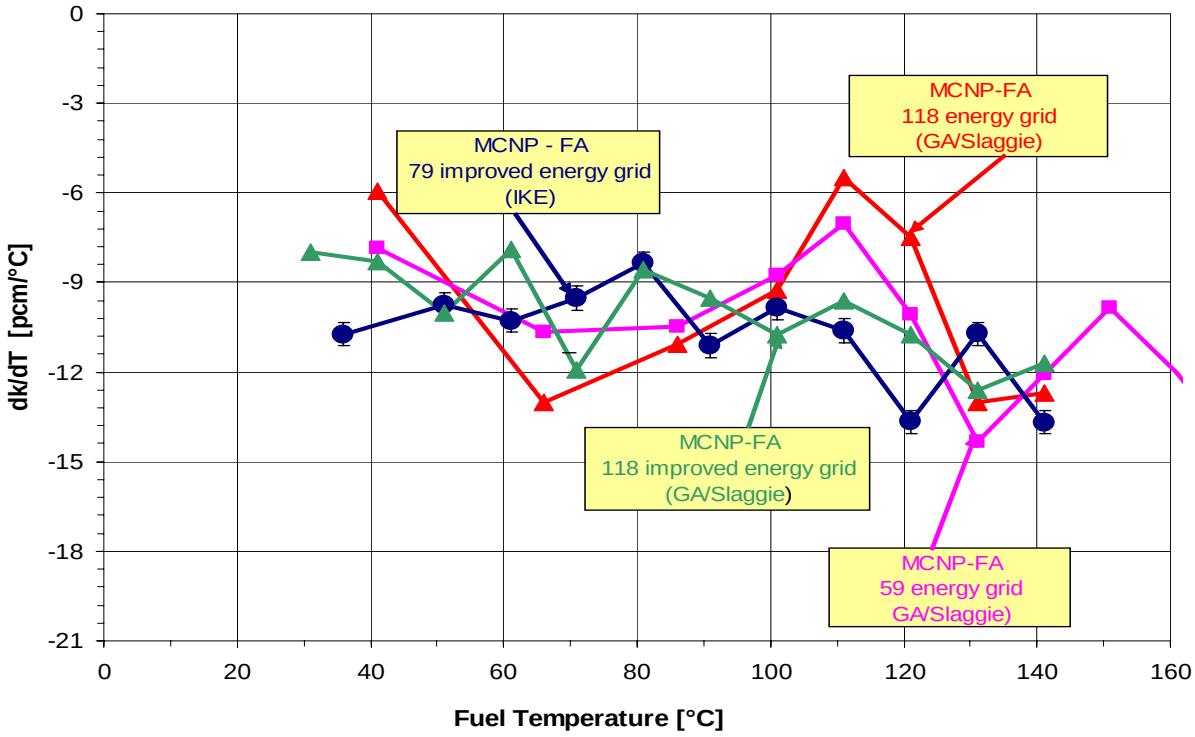

Figure 5: Fresh TRIGA fuel pin MCNP calculations for four different cases.

4.1 Additional parameters concerning the probability tables

The size of the tables are dependent on the number of cosine bins for the angular distribution and the number of equal probable out-scattered energy points for each incident energy. The size of the tables showed minor criticalities changes for the values analysed in this study.

For the second scatterer in a molecule it is usually recommended to use the ideal gas treatment [9]. Nevertheless as data exists for zirconium in zirconium hydride a comparison between the free gas model and the bound zirconium based probability $S(\alpha,\beta)$ tables was performed which showed a systematic growth of about 20 pcm for all temperatures in criticality when the free gas model is used. For the reactivity feedback evaluation it had therefore no influence.

5. Improved $S(\alpha,\beta)$ tables for resonant dependent cross sections

The scattering-law treatment was extended for U238 to deal with the influence of the modified energy dependent scattering kernel proposed in ref [13, 14] on the TRIGA criticality. The new scattering kernel takes into account the influence of the resonances on the scattered neutron from its initial energy and spatial angle to its final energy and angular direction. It should be mentioned that the thermal agitation of the target isotope is handled in the modified kernel as an ideal gas complying with the Doppler broadening of the scattering cross section itself which is actually the integrated value of the scattering kernel over all angles and energies.

938 probability tables were generated for each of the four temperatures, based on the new double differential scattering kernel [13]. The calculated tables at 26, 126, 326 Celsius (300, 400, 600 K) and (for academic reason) at 926(1200 K) Celsius cover the 6.0 – 210 eV energy range, in particular the 8 most significant S-resonances of U238.

Introducing the new kernel for U238 scattering within the MCNP code for the TRIGA fuel pin leads to the criticality values shown in table 2. In the low temperature range which was foreseen for the TRADE project the Doppler broadening of the scattering kernel has no significant impact compared to the effects obtained by the scattering law for the hydrogen in zirconium. At 326 Celsius which is an operational temperature for several TRIGA cores the impact of the resonant dependent scattering kernel is enhanced to some extent. For the reactivity feedback this effect is still negligible. At 926 Celsius the criticality change due to the modified kernel is noticeable which confirms the physical aspect of the kernel, (yet has no practical use for common TRIGA cores).

Table 2: Criticality values for TRIGA fuel pins calculated with MCNP code. The standard deviation for all calculations is $6*10^{-5}$.

Temperature	Criticality with normal MCNP treatment	Criticality with 938 $S(\alpha,\beta)$ tables for U238
26^0 C	1.28891	1.28871
126^0 C	1.27965	1.27940
326^0 C	1.25461	1.25424
926^0 C	1.19826	1.19757

6. Conclusions

The changes in criticality and reactivity feedbacks due to different parameters for the generating the $S(\alpha,\beta)$ tables were discussed. For light isotopes there are three levels of differences. The first group concerns the basic structure lattice model which is expressed by the

phonon spectrum within the LEAPR module. The presented results could not conclusively demonstrate whether one of the two phonon spectrum models is more reliable. Additional experiments should be performed to improve the data base of temperature dependent reactivity feedbacks in TRIGA fuel.

The second level of differences concerns the input for the relevant modules in NJOY. The tolerance value for the point-wise cross section description and the size and the values of the energy grid in the THERMR module affect considerably the criticality and the reactivity feedback coefficient. The differences between the standard 59 energies grid set to the other models emphasizes the need to increase in THERMR the energies points set to the suggested generalized (for all relevant bound isotopes) 79 values energy grid set by IKE [9]. Higher number of energy grid points could be used for specific analysis of the cross section value itself and for refinement of the generalized 79 point set results.

The third level of investigation concerns the size of each table. Within the error bars of the experimental results, there was not a clear trend of the exact influence of the number of the cosine bins or the equi-probable energy interval on the criticality. Improvement of the experimental data could illuminate this part of investigation.

The $S(\alpha, \beta)$ treatment for heavy isotopes in particular for U238 seems to have limited impact concerning the parameters in this study.

The dense temperature dependent $S(\alpha, \beta)$ scattering kernel tables for bound hydrogen were prepared for Monte Carlo calculations with the continuous energy option of the MCNPX code, beta version 2.5.d**.** The consequences of newer versions which treat differently the $S(\alpha, \beta)$ tables within MCNPX should be further analyzed in the future.

7. References

[1] The Working Group on TRADE (TRIGA Accelerator Driven Experiment) – TRADE Final Feasibility Report, March 2002.

[2] R. Rosa , RC-1 Casaccia. Activities performed on the 6-7 weeks of 2004. Reported on the TRADE official Web Site – "In pile Experiments" section. February 2004. Data evaluated by M. Schikorr FZK (private communication).

[3] I. Mele , M. Ravnik, A. Trkov. TRIGA MARK II Benchmark Experiment, Part I: Steady State Operation, Nuclear Technologies p. 37 Vol 105 , Jan 1994. . Data evaluated by M. Schikorr FZK (private communication)

[4] General Atomics, TECNICAL FOUNDATIONS OF TRIGA , GA-471 (1958).

[5] Badea M. Broeders C.H.M, .Dagan R., Travleev A,. Neutron Physics Investigations for TRIGA Fuel, Jahrestagung Kerntehnik Düsseldorf, 2004.

[6] Dagan R. "On the use of $S(\alpha, \beta)$ tables for nuclides with well pronounced resonances, ANE pp. 367-377 vol. 32, 2005.

[7] J.F. Briesmeister, Editor, "MCNP – A General Monte Carlo N–Particle Transport Code, Version 4C", LA-13709-M, Manual ,2000.

[8] E.L.Slaggie, "Central Force Lattice Dynamical Model for Zirconium Hydride", General Atomic report GA-8132 (July 1967).

[9] Mattes M., Keinert J. "Thermal Neutron Scattering Data for the Moderator Materials H_2O, D_2O and ZrH_x in ENDF-6 Format and ACE Library for MCNP(X) Codes" , INDC (NDS)-0470 , April 2005.

[10] Whittemore W. L. McReynolds A. W. Phs. ReV 113 , 1959

[11] MacFarlane, R.E., New Thermal Neutron Scattering Files for ENDF/B-VI Release 2 , LA-12639-MS, March 1994,

[12] MacFarlane, R.E., Muir, D.W., The NJOY Nulcear Data Processing System, Version 91 (LA-12740-M), 1994.

[13] Rothenstein, W., Dagan, R., "Ideal Gas Scattering Kernel for Energy Dependent Cross Sections", Ann. of Nuclear Energy 25, 209-222, 1998.

[14] Rothenstein, W., "Proof of the formula for the ideal gas scattering kernel for nuclides with strongly energy dependent scattering cross sections", Ann. of Nuclear Energy 31, 9-23, 2004.

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