Zirconium Carbide TSL Development

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Overview

- Zirconium Carbide (ZrC) is a refractory material currently investigated for high temperature reactor applications
 - Fission product barrier in advanced TRISO
 - Corrosion barrier in space nuclear thermal propulsion (SNTP)
- ZrC studied as base of (U,Zr)C solid solution and UC₂ composite fuels for NERVA-type cores during Rover program
 - Testing of material performed at Nuclear Furnace 1 (NF-1) located at Los Alamos
- Thermal Scattering Law (TSL) evaluations generated with FLASSH as new contribution to ENDF/B-VIII.1
 - Standard incoherent approximation
 - New mixed elastic scattering format
 - Disordered alloy theory introduced to coherent elastic scattering to capture natural isotopic abundances
 - Work submitted to PHYSOR 2022

TSL Evaluation Methods



Zirconium Carbide Phonon Model Validation

- Crystal structure lattice parameter in agreement with experiment.
 - 0.471 nm vs. 0.470 nm
- Phonon dispersions relations in good agreement with experiment
 - Differential validation of phonon spectra
- Debye-Waller coefficient in reasonable agreement with experiment from 0 K – 1600 K
 - Integral validation of phonon spectrum
 - W is a measure of mean square displacement used directly in TSL
- C(ZrC) spectra localized around higher energy modes separated from lower energy modes
 - Similar behavior in metal-hydrides introduce regular structure S(α, β) (quantum oscillations) that strongly influence thermalization



Thermal Neutron Scattering Formalism

- TSL contains distinct (interference) and self (non-interference) components
 - Momentum transfer (α) and energy transfer (β) represented with unitless parameters

$$S(\alpha,\beta) = S_d(\alpha,\beta) + S_s(\alpha,\beta)$$
 $\alpha = \frac{E + E' - \mu\sqrt{EE'}}{Ak_BT}$ $\alpha = \frac{E' - E}{k_BT}$

- Phonon expansion used in incoherent approximation for inelastic scattering (p > 0) $S_s(\alpha, \beta) = \sum_p S_s^p(\alpha, \beta)$
- C(ZrC) and Zr(ZrC) both include: coherent elastic, incoherent elastic, and inelastic
 ⁹¹Zr and ¹³C have small incoherent nuclear potential

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E'} = \frac{1}{4\pi k_B T} \sqrt{\frac{E'}{E}} \left[\sigma_{coh} S^0(\alpha, \beta) + \sigma_{inc} S^0_s(\alpha, \beta) + (\sigma_{coh} + \sigma_{inc}) \sum_{p>0} S^p_s(\alpha, \beta) \right]$$

Disordered alloy theory invoked to treat coherent elastic

$$\sigma_{coh,el}(E) = \frac{1}{E} \sum_{E_I < E} \frac{1}{G_I} |F(\mathbf{G}_I)|^2 - \begin{bmatrix} Past Method: & F(\mathbf{G}_I) = \sum_{\mu} \sqrt{\langle \sigma_{coh,\mu} \rangle} \exp(i\mathbf{G}_I \cdot \mathbf{R}_{\mu}) \exp(-\langle W_{\mu} \rangle G_I^2) \\ Disordered Alloy: & F(\mathbf{G}_I) = \sum_{\mu,j} f_j \bar{b}_{\mu,j} \exp(i\mathbf{G}_I \cdot \mathbf{R}_{\mu}) \exp(-W_{\mu,j} G_I^2) \end{bmatrix}$$

ZrC TSL Evaluation Parameters

- C and Zr potential scattering cross sections extracted from ENDF/B-VIII.0 nuclide evaluations at incident neutron energy of 0.0253 eV
 - Incoherent cross section from V. F. Sears, Neutron News, 29, (1992) 26-37
 - Isotopic average cross section for incoherent elastic and inelastic scattering
 - Isotopic average scattering length for coherent elastic scattering
- C(ZrC) and Zr(ZrC) evaluated at 13 temperature:
 - 77 K, 293.6 K, 400 K, 500 K, 600 K, 700 K, 800 K, 1000 K, 1200 K, 1400 K, 1600 K, 1800 K, 2000 K
- Phonon expansion order of 300 used for convergence of $S(\alpha, \beta)$
 - Eliminate the need for short collision-time-approximation in FLASSH
- (α,β) grid valid to 5 eV incident neutron energy
 - Default automatic grid in FLASSH
 - α -grid of 200 points $\left[\frac{0.415322}{4}, \frac{790.5139}{4}\right]$

 - β-grid of 283 points [0,197.63]

ZrC TSL Evaluation

- Zr(ZrC) TSLs characteristic of heavy elements in crystalline compounds
 - Structure up to 1-phonon energy transfer and relatively featureless beyond
 - Approximating free-gas (FG) for high momentum transfer
- C(ZrC) has strong quantum oscillator effect to multiple phonon orders
 - Oscillations in TSL correspond to phonon order
 - Energy transfer involves quantized exchange of higher energy phonons with substantial downscattering and upscattering
 - Differs from $H(ZrH_x)$ where upscattering is unlikely at room temperature
 - Shallow oscillations persist as FG behavior present for large momentum and energy



C(ZrC) Secondary Neutron Distributions

- C(ZrC) is compared to reactor-grade graphite (RxGr)
 - RxGr represents conventional TSL for chemical binding of C
- Epithermal range: ZrC down-scattering is quantized and greater than RxGr
 - Behaviors more similar at higher temperature but some quantization remains
- Neutrons in the phonon energy range have unusually probable high energy losses
 - Near complete energy loss
 - Atypical of non-hydrogenous solids
 - Improbable for FG or typical carbon compounds
- Thermal range: Quantized energy loss is forbidden in thermal energy range and below
 - Up-scattering is dominant
 - Spectral hardening is expected
 - Increase probability of quantized upscattering with temperature
 - Consistent with metal hydrides



*(a)-(c) secondary spectra at labeled energy; (d) integrated cross section at labeled temperatures

Conclusions

- New TSL evaluations for ZrC have been generated for contribution to ENDF/B-VIII.1
 - Support criticality safety analyses for high temperature reactor systems
- New evaluations extend physics of elastic scattering used in ENDF evaluations
 - Use of mixed elastic scattering
 - Introduction of disordered alloy theory to improve coherent elastic treatment
- C(ZrC) exhibits quantum oscillator behavior that is atypical of nonhydrogenous compounds
 - May be important for evaluation of systems with significant ZrC content
 - TRISO coatings
 - SNTP applications
 - Initial critical mass calculations demonstrate a C(ZrC) impact compared to FG
 - Further effects testing is in progress