Thermal Conductivity and Electrical Resistivity of Simulated Fuel Elements*

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It is well-known that important changes may occur in the physical properties of bonded fuel rods during their lifetime within a reactor. These changes are caused by the high temperatures, environment, and lastly (and probably most importantly) by the high neutron fluxes. Neutrons have a strong effect in reducing the thermal conductivity, λ, by introducing defects such as vacancies and dislocations into the basal planes of graphitic material.[1] The reduction of λ raises the operating temperature which lowers the lifetime of the graphite. Although the quantitative effects of neutron irradiation on the λ of polycrystalline graphites have been extensively studied,[2-4] they have not been studied for carbonaceous material nor for carbonaceous material containing various quantities of spherical fuel particles which are envisioned for use in a high-temperature gas-cooled reactor (HTGR). The primary purpose of this experiment, therefore, was to measure the thermal conductivity of simulated fuel elements consisting of inert particles suspended in a carbonaceous matrix as a function of temperature, T, volume percent particle loading, and neutron fluence. The electrical resistivity, ρ, Seebeck coefficient, S, and coefficient of thermal expansion, CTE, were also measured as functions of the same parameters. The CTE and S, however, will not be discussed here.

Specimen Description and Measurement Technique

This experiment included extruded specimens with nominal simulated-fuel particle contents of 0, 13, 23, and 35 vol % with matrix densities of 1.75, 1.70, 1.65, and 1.60 Mg/m^3, respectively. These four were formed by extrusion with a matrix of 75.7 wt % graphite, 24.3 wt % thermax, and prepolymerized furfuryl alcohol. A series of slug-injected specimens, all with about 58 vol % inert particles, were fabricated with a matrix of 28.5 wt % graphite filler in pitch. The matrix densities of the latter specimens were about 0.60 Mg/m . Since measurements of λ after irradiation were done outside a hot cell, inert carbon particles were used as surrogates for fuel particles to minimize postirradiation radioactivity. After fabrication, all specimens were heat treated at 2070 K for 1.8 Ks in argon. The specimens were irradiated in the High Flux Isotope Reactor at Oak Ridge National Laboratory at nominal temperatures of 1220 K.

Steady-state longitudinal techniques[5] were used for most of the λ, ρ, and S measurements. The ρ was measured using a standard four-probe reversible-dc technique with the thermocouple elements serving as potential probes. Measurement uncertainties were about ±0.4%, ±5%, and ±0.1 μV/K for the ρ, λ, and S, respectively.

Results

Unirradiated Material — Thermal conductivity results obtained on the unirradiated specimens are shown in Fig. 1 as a function of T. The λ decreases smoothly with increasing particle content and λ decreases with increasing T. The two curves below 400 K were generated with a low-temperature technique on specimens from a previous experiment, and these are in relative agreement with the present results.

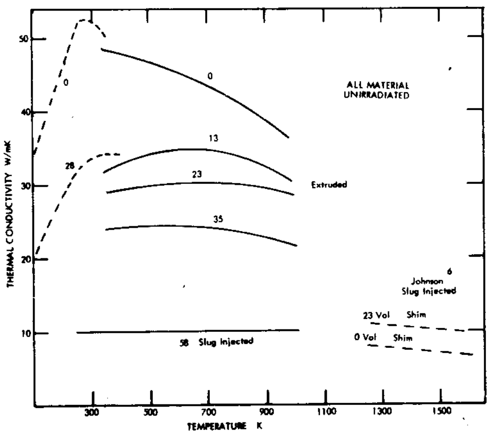

Fig. 1. The λ of Unirradiated Simulated-Fuel Elements versus T. The two low T curves were obtained on extruded elements with a different matrix.

The λ of unirradiated material at 400 K and the electrical conductivity, $\sigma = \rho^{-1}$, at 300 K are shown in Fig. 2 versus volume percent particle loading. The λ decreases smoothly with particle loading, and this curvature is mirrored by σ. This similitude

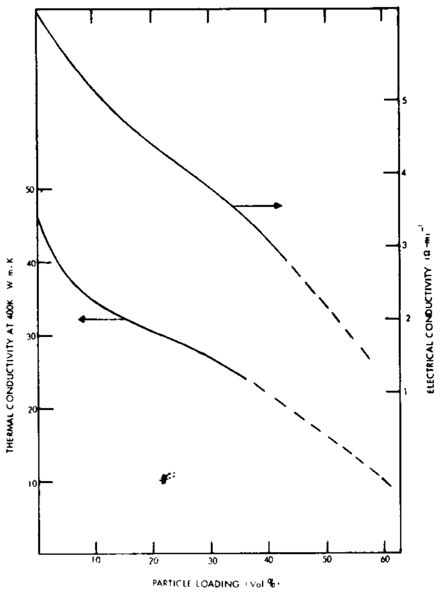

Fig. 2. The λ (400 K) and σ (300 K) of Unirradiated Simulated-Fuel Elements Versus Volume Percent Loading.

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indicates that, to a first approximation, λ and σ are dependent on the same variables within the composites.

Irradiated Material — After irradiation, the electrical resistivities of all surviving specimens were measured near 300 K. The ρ versus fluence is shown in Fig. 3. The ρ of the slug-injected materials is much greater than that of the extruded materials and increases steadily with increasing fluence. The ρ of the extruded specimens increases with fluence initially, saturates above about 4 to 6 × 10^{25} n/m^2, and then begins to increase again above 6 to 9 × 10^{25} n/m^2.

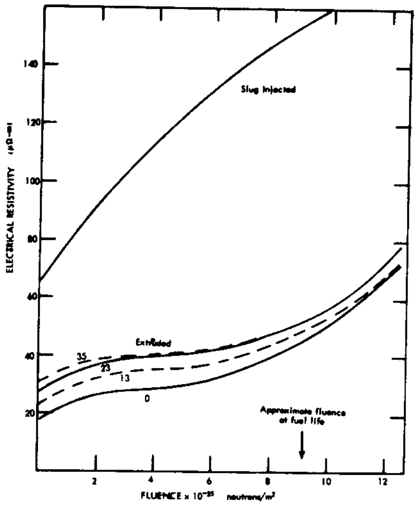

Fig. 3. The Room Temperature ρ of Extruded and Slug-Injected Fuel Elements Versus Neutron Fluence.

The smoothed λ results are given in Table I. Over the entire temperature range, the matrix material for the extruded specimens (*i.e.*, the specimen with 0 vol % particles) has a steadily reducing λ with increasing neutron fluence. The λ of the 13 and 23 vol % specimens remains essentially constant with increasing fluence until the fluence goes above 5 × 10^{25} n/m^2. The λ of the 35 vol % specimen actually increases with fluence initially and remains quite high beyond 5 or 6 × 10^{25} n/m^2. The λ of the extruded specimens remains high over the fluence range where ρ of each material approaches a constant value. The unirradiated slug-injected specimen and the one irradiated to 2.34 × 10^{25} n/m^2 contained 58 vol % fuel particles, whereas actual fuel normally contains a lower percentage fuel with the difference being composed of graphite particles. For example, the last specimen listed in Table I consisted of 36.5 vol % simulated-fuel particles and 20.2 vol % shim. The λ of this specimen near 400 K was only 5 W/mK. Thus the λ of the 35 vol % extruded specimen after 9.0 × 10^{25} n/m^2 was a factor of 2.6 greater than a slug-injected specimen with similar simulated-fuel particle loading and fluence.

Discussion of Results

The λ and ρ results are primarily controlled by the carbonaceous matrix material and the effect of neutrons on different components of this matrix. The matrix density of all extruded specimens is a factor

Table I. Smoothed Values for the Thermal Conductivity of Simulated Fuel Elements Versus Temperature, Neutron Fluence, and Particle Loading

V^a	n^b	Thermal Conductivity, W/mK						
		400 K	500	600	700	800	900	1000
0	0	47.5	47.2	46.2	44.6	42.1	39.0	35.3
0	2.0	40.4	42.3	43.3	43.3	42.4	40.6	38.0
0	8.1	20.0	21.2	21.9	22.0	21.7	21.0	--
0	12.1	9.6	10.1	10.5	10.6	10.6	10.6	10.5
13	0	33.0	34.6	35.2	34.9	34.0	32.2	30.6
13	2.0	32.8	34.1	34.5	34.2	33.6	32.3	30.4
13	4.0	29.0	29.6	29.9	29.7	29.4	28.8	28.0
13	8.1	18.8	19.8	20.5	21.0	21.0	20.6	19.7
13	12.1	11.2	12.0	12.5	12.7	12.5	12.0	11.0
23	0	29.5	30.0	30.2	30.4	30.2	29.6	29.2
23	2.3	28.6	30.0	30.8	30.5	29.6	28.1	26.0
23	4.5	28.1	28.7	28.8	28.6	28.0	27.1	26.0
23	9.0	18.0	18.8	19.2	19.2	18.9	18.3	17.6
35	0	24.2	24.6	24.7	24.2	23.2	22.0	--
35	2.3	27.6	29.0	30.0	30.2	29.6	28.0	26.0
35	4.5	22.2	24.0	25.6	26.7	27.8	28.5	28.5
35	9.0	12.5	13.4	14.2	14.4	14.0	13.0	11.5
58^c	0	10.2	10.8	11.2	11.3	11.2	10.8	10.4
58^c	2.3	11.8	12.0					
58^d	8.6	~5.0						

[a]Volume percent simulated-fuel particle loading.

[b]Neutron fluence × 10^{-25} n/m^2

[c]All particles were simulated fuel.

[d]36.5% simulated-fuel particles and 20.2% shim.

of 2.6 greater than the matrix density of the slug-injected specimens. At all irradiation stages this density difference has a dominant effect on the properties.

The most interesting aspect of the data is the initial increase of λ in the 35 vol % extruded specimen with neutron irradiation. The λ of this material, as well as λ of the 13 and 23 vol % specimens, remains high beyond a fluence of 5 or 6 × 10^{25} n/m^2. This initial increase of λ in the 35 vol % specimen is caused by the partial ordering of the carbonaceous material surrounding the graphite filler particles under the influence of the neutron flux and strain. This effect has been observed previously in similar structures where the heat treatment was at such a low temperature (2070 K) and the neutron irradiation occurred at such a high temperature (1220 K).[7]

References

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