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# Thermal Conductivity of Graphite Irradiated with Medium and High Neutron Fluences

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# Thermal Conductivity of Graphite Irradiated With Medium and High Neutron Fluences

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**Abstract -** *Calculations of the temperature distribution within a High Temperature Gas-cooled Reactor core need data on the thermal conductivity of graphite at high fast neutron fluences, and at various temperatures, e.g., in the range 400 to 1200 °C. In the past, however, most thermal conductivity measurements for simplicity were performed at room temperature. An existing model for the transformation of room temperature to high temperature thermal conductivity has never before been verified sufficiently, but is now tested using data on numerous graphite specimens that were irradiated at nearly constant temperature in the range 300 to 1400°C and measured at that same temperature. It turns out that the thermal conductivity remains constant at medium fluence on the same level for all graphites under investigation. At higher fluences, further decrease of thermal conductivity can be related to the volume changes beyond "turn-around" when structural changes induce volume expansion.*

## I. INTRODUCTION

Since a couple of years, the interest in High Temperature Gas-Cooled Reactors (HTGR) has markedly increased, not least because of the encouraging construction experiences and successful operation of the Japanese HTTR and the Chinese HTR-10 nuclear power plants. However, compared to these reactors as well as to the former HTGR designs such as the German Modular HTR, there is a trend to achieve higher power output, higher efficiency, and hence smaller production cost per kWh.

In order to maintain the safety features the HTR is known for, more and more interest has to be directed to the thermal conductivity of side reflector graphite after significant fast neutron irradiation. It is well known that thermal conductivity decreases very rapidly with increasing neutron fluence. However, a minimum thermal conductivity must be left over at all times to remove the afterheat from the core in case of a loss of pressure accident, thus to prevent the fuel temperature from exeeding the limit of about 1600°C, where release of some fission products has been proven to start and to increase with increasing temperature.

The reason for the decrease of thermal conductivity with increasing fast neutron fluence is the formation of small defects within the graphite crystallites acting as scattering centers and thus hindering the propagation of phonons. Depending upon the ambient temperature during irradiation which influences the defect mobility, an equilibrium between the formation and the annihilation of defects is reached, and the thermal conductivity remains constant for some period of time on a level which increases with increasing ambient temperature.

Calculations of the temperature distribution within the reactor core need to know the thermal conductivity of reflector graphite measured at the irradiation temperature, e.g., in the range 400 to 1200 °C. In the past, however, most thermal conductivity measurements for simplicity were performed at room temperature. An existing model[1] for the transformation of room temperature thermal conductivity to high temperature thermal conductivity has never before been verified sufficiently, but is now tested with respect to the temperature dependent saturation levels of the decrease of relative thermal conductivity mentioned above.

## II. EXPERIMENTAL METHODS

The paper is based upon data of numerous graphites that were irradiated in the Petten High Flux Reactor from 1970 till 1990. The irradiations were performed stepwise to increasing neutron fluences at nearly constant temperatures. Capsules equipped with thermocouples and containing about 100 specimens were kept at constant temperature by gas mixture control and by shifting the capsule vertically as fuel was burned and control rods were raised. The irradiated samples were measured before and after each irradiation step. Thermal conductivity was measured either in the temperature range from room temperature to irradiation temperature using cylindrical specimens 6 mm ∅ and 32 mm long[a], or at room temperature using cylindrical specimens 6 mm ∅ and 25 mm long.

Fig. 1 gives an example for thermal conductivity measurements at temperatures in the range 100 to 1000°C on a graphite specimen before and after four irradiations at 950 °C to fast neutron fluences of 0.26, 0.98, 1.75, and 3.58 * $10^{21}/cm^2$ (EDN)[b].

## III. EXPERIMENTAL RESULTS TO DETERMINE THE SATURATION LEVEL OF THERMAL CONDUCTIVITY IN THE TEMPERATURE RANGE 300 TO 600 °C

In the framework of the German nuclear reactor development programme, numerous graphites were irradiated in the High Flux Reactor (HFR) at Petten (The Netherlands) at temperatures from 300 to 600°C.

Fig. 2 shows that at 300°C thermal conductivity very rapidly decreases with increasing fast fluence to a saturation level of about -90% of the pre-irradiation value. It is remarkable, that the relative changes are very similar for six different graphites, parallel as well as perpendicular to the preferred orientation.

This result is confirmed at 400°C irradiation and measuring temperature (Fig. 2) for even more (17) different graphite grades. However, the saturation level as expected is found at a higher level (smaller thermal conductivity decrease) of only about -80%.

At 500 and 600°C, the saturation levels are found at -68%, and -61% respectively. However, at higher fluences the thermal conductivity decreases again. This effect is attributed to structural changes within graphite at very high fast neutron fluences and hence is not expected to be material independent.

At irradiation temperatures above 600°C, for some nuclear graphites thermal conductivity sporadic measurements at irradiation temperature are existing. However, maximum fluences are too low to determine the saturation level. But a huge number of data received by room temperature measurements are existing. To be exact: The irradiation was performed at temperatures in the range 400 to 1400 °C, whereas the thermal conductivity before and after irradiation was measured at room temperature only.

## IV. CORRELATION OF HIGH TEMPERATURE AND ROOM TEMPERATURE THERMAL CONDUCTIVITY CHANGES UNDER FAST NEUTRON IRRDIATION

A model for converting changes of thermal conductivity by irradiation measured at room temperature to changes measured at irradiation temperature has been suggested long ago[1], but so far has never been thoroughly tested.

The model is based upon a separation of the dependence of thermal resistivity *1/k* on the measuring temperature from the dependence upon the irradiation damage at any temperature *T*:

$$\left(\frac{1}{k}\right)_T = \left(\frac{1}{k_0}\right)_T + \left(\frac{1}{k_{irr}}\right)_T \qquad (1)$$

This equation can be rearranged:

$$\left(\frac{1}{k_{irr}}\right)_T = \left(\frac{1}{k}\right)_T - \left(\frac{1}{k_0}\right)_T =$$

$$= \left(\frac{1}{k_0}\right)_T \cdot \left(\frac{k_0}{k} - 1\right)_T \qquad (1a)$$

and for 30°C:

$$\left(\frac{1}{k_{irr}}\right)_{30} = \left(\frac{1}{k_0}\right)_{30} \cdot \left(\frac{k_0}{k} - 1\right)_{30}$$

[a] Measurements were performed by L. Binkele, Kernforschungsanlage Jülich (KFA), Institute for Reactor Materials

[b] EDN ≡ Equivalent DIDO Nickel fluence

Equation (1$a$) can be transformed:

$$\left(\frac{1}{k_{irr}}\right)_T \equiv \frac{\left(\frac{1}{k_{irr}}\right)_T}{\left(\frac{1}{k_{irr}}\right)_{30}} \cdot \left(\frac{1}{k_{irr}}\right)_{30} =$$

$$= d_T \cdot \left(\frac{1}{k_0}\right)_{30} \cdot \left(\frac{k_0}{k} - 1\right)_{30}$$

From equation (1) follows:

$$\left(\frac{1}{k}\right)_T = \left(\frac{1}{k_0}\right)_T + d_T \cdot \left(\frac{1}{k_0}\right)_{30} \cdot \left(\frac{k_0}{k} - 1\right)_{30}$$

and finally:

$$\left(\frac{k_0}{k} - 1\right)_T = d_T \cdot \frac{(k_0)_T}{(k_0)_{30}} \cdot \left(\frac{k_0}{k} - 1\right)_{30} \qquad (2)$$

Equation (2) says that the relative resistivity change at high temperature $T$ can be calculated from the resistivity change at room temperature (30°C) by multiplying with the ratio of the unirradiated thermal conductivities at temperatures $T$ and 30°C. The additional factor $d_T$ defined as

$$d_T = \left(\frac{1}{k_{irr}}\right)_T \Big/ \left(\frac{1}{k_{irr}}\right)_{30}$$

can be assumed[1] to be nearly equal to 1, as it depends only upon the irradiation damage which in fairly good approximation is temperature independent.

Equation (2) converting resistivity changes at high temperature $T$ to resistivity changes at room temperature 30°C and vice versa can be checked using measured data on IE1-24 graphite, a Gilsonite coke material. Its pre-irradiation thermal conductivity has been measured as a function of temperature, where the conversion factor $(k_0)_T/(k_0)_{30}$ can be deduced from.

Fig. 3 is showing thermal conductivity change data received from specimens irradiated at about 400°C and measured at room temperature. From the saturation level at -80% at 400°C (see Fig. 2) a saturation level for room temperature data is calculated to be -87% which is shown as a black bar in Fig. 3 in reasonable agreement with the data points.

A similar check is possible for 600°C irradiation temperature. Fig. 2 yielding a saturation level at -62% for high temperature thermal conductivity changes, and with the conversion factor $(k_0)_T/(k_0)_{30}$ for $T$ = 600°C, the saturation level for room temperature data is calculated to be at -76%, shown as the black bar in Fig. 4, which again is in good agreement with the data points.

Now, saturation levels for thermal conductivity data measured at irradiation temperature can also be calculated in case of irradiation temperatures above 600°C, if there are thermal conductivity data at room temperature available.

As the final result from all available data, material independent levels of the saturation of thermal conductivity changes at room temperature as well as at irradiation temperature can be calculated; they are plotted in Fig. 5 and Fig. 6.

## V. The Secondary Decrease of Thermal Conductivity

As mentioned in chapter II, at very high neutron fluences thermal conductivity decreases below the saturation level due to structural changes. This effect has been observed for the Young's modulus as well, and is explained by the progressive tightening of the structure as microcracks close and intercrystalline restraint increases[2].

Using the thermal conductivity data which are available now, it can be demonstrated, that the secondary breakdown is correlated to the structural changes as well. This is shown in Fig. 7 and Fig. 8 for a Gilsographite irradiated at about 1400°C.

This is the clue to thermal condcutivity behaviour of graphite at very high fast neutron fluences, if only dimensional or volume changes are determined wheras thermal conductivity has not been measured. From the saturation level (which has been proven to be material independent), the thermal conductivity starts decreasing again at the turn-around fluence aproaching zero thermal conductivity asymptotically according to a function like

$$\left(\frac{k_0}{k} - 1\right) = S + A_V \cdot (g - g_0) \qquad \text{for } g > g_0$$

where

$S$: saturation level of thermal resitivity change,
$A$: parameter to be determined,
$g$: fast neutron fluence ($10^{25}/m^2$ (EDN)),
$g_0$: fast neutron fluence ($10^{25}/m^2$ (EDN)) at turn-around,
$V_R$: function describing the volume increase after turn-around, definde as

$$V_R(g) = \frac{\frac{\Delta V}{V_0}(g)}{\left(\frac{\Delta V}{V_0}\right)_{\min}} - 1$$

As a first attempt to determine the parameter A, data of three different graphites irradiated at two different temperatures were used for a regression analysis. The preliminary result is A = -0.15 as an average. However, this value is regarded as a first approximation only, and should be improved as soon as more data showing secondary breakdown will become available

## VI. Conclusions

1. The relative changes of thermal conductivity at the saturation level are very similar even for very different graphites. This result is important for design calculations based upon graphite that has not been irradiated yet.
2. The formula for the transformation of irradiation induced thermal conductivity changes measured at room temperature to changes measured at irradiation temperature has been checked and found to be valid. This makes numerous thermal conductivity data measured at room temperature usable for high temperature applications.
3. The secondary breakdown of thermal conductivity at high fluences is related to the relative volume change after turn-around, thus yielding the irradiation behaviour of thermal conductivity even in cases where only dimensional changes have been measured.

## REFERENCES


1 B.T. Kelly, The thermal conductivity of graphite, Chemistry and Physics of Carbon **5**, 119 (1969).

2 R.J.Price, GA-A13524, 1975

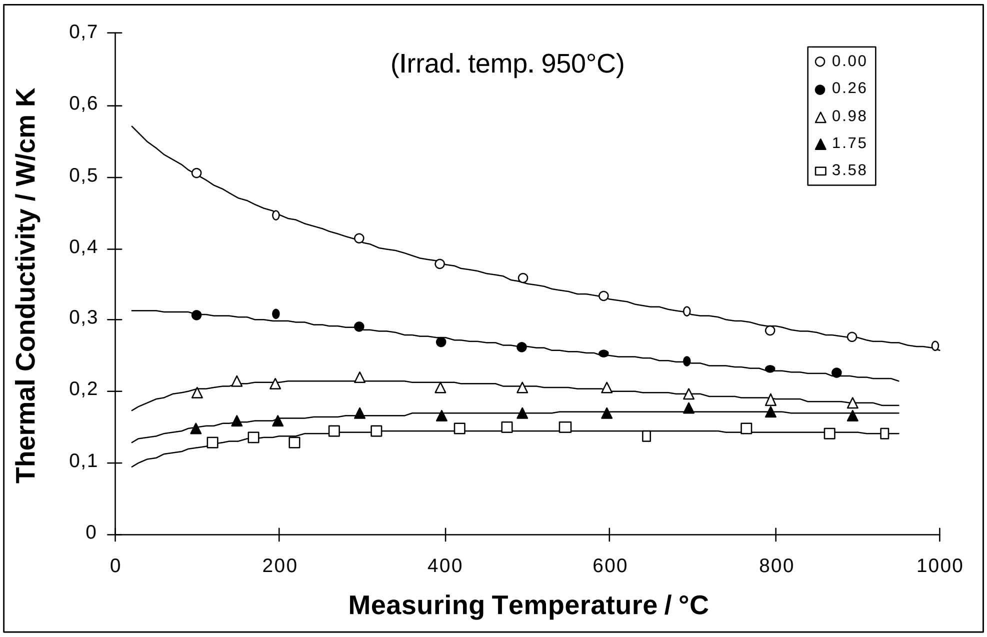


Fig. 1 – Thermal conductivity of a nuclear graphite as a function of temperature before irradiation (0.00) and after four irradiations at 950 °C to fast neutron fluences of 0.26, 0.98, 1.75, and $3.58*10^{25}/m^2$ (EDN).

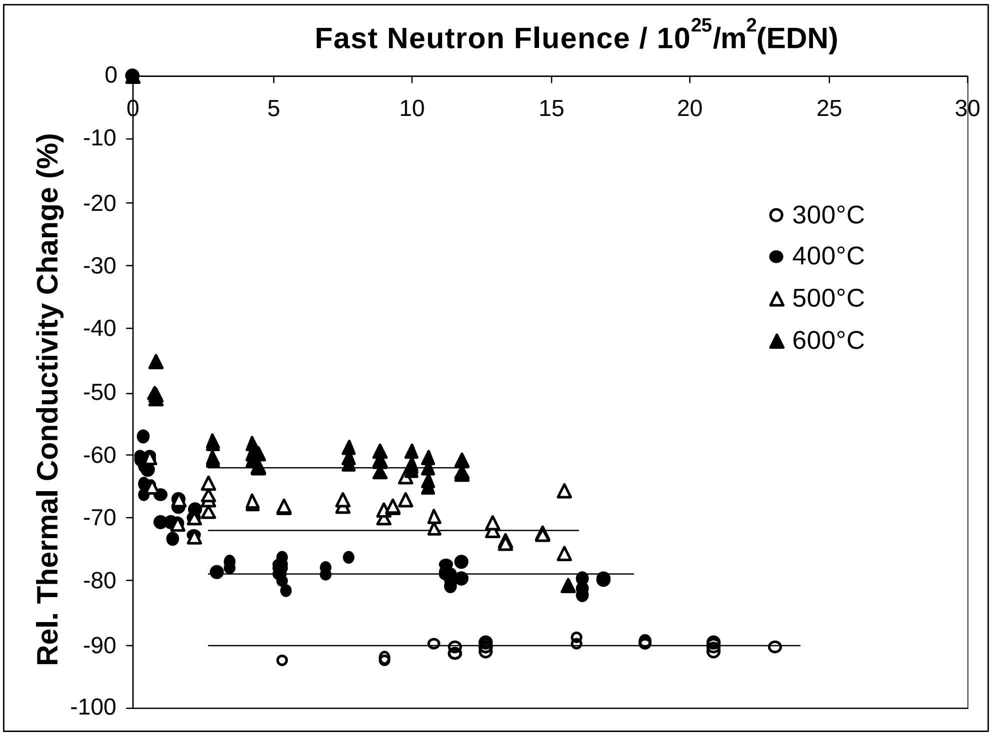


Fig. 2 – Relative thermal conductivity changes of different nuclear graphites, induced by irradiation at about 300, 400, 500, and 600 °C; measured at the irradiation temperature

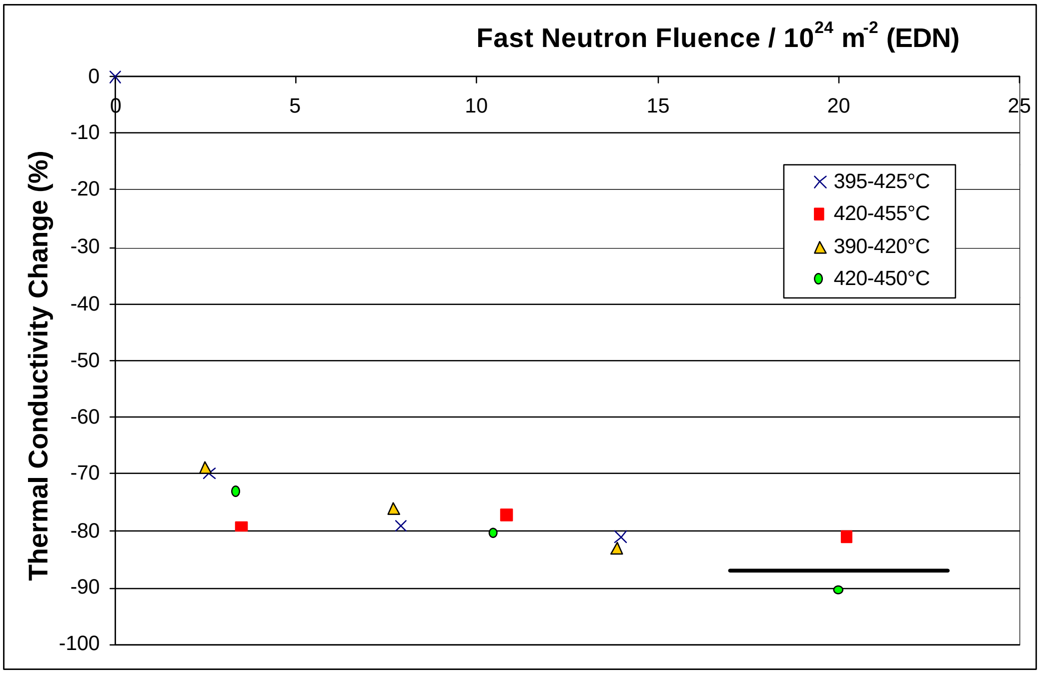


Fig. 3 – Relative thermal conductivity changes of IE 1-24 graphite, induced by irradiation at about 400 °C; measured at room temperature; saturation level (black bar) calculated from saturation level in Fig. 2.

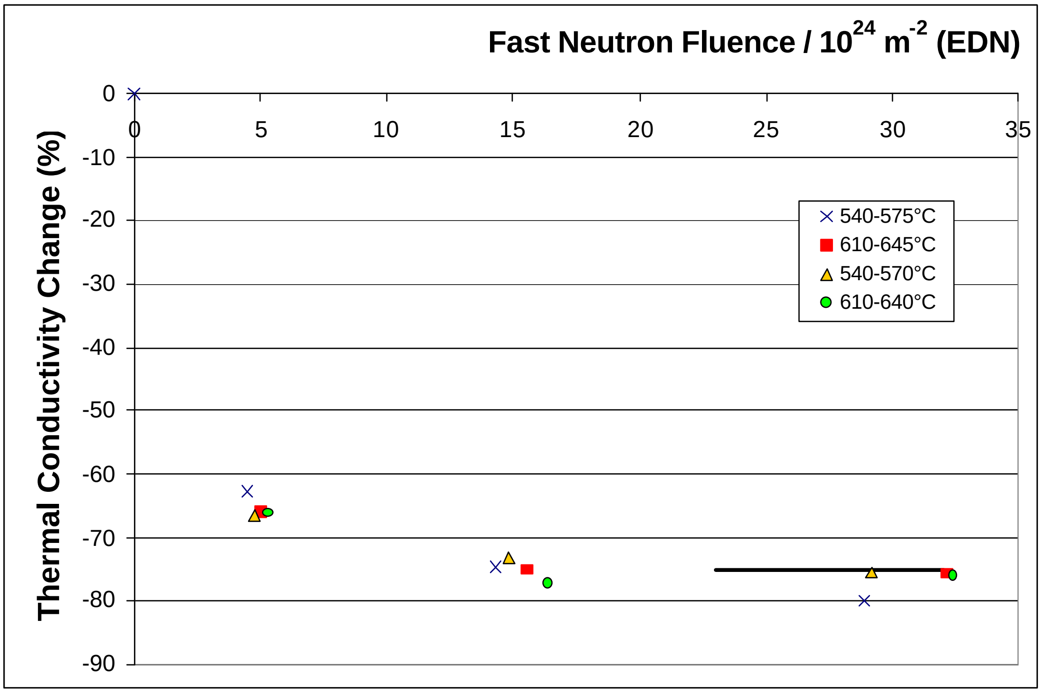


Fig. 4 – Relative thermal conductivity changes of IE 1-24 graphite, induced by irradiation at about 600 °C; measured at room temperature; saturation level (black bar) calculated from saturation level in Fig. 2.

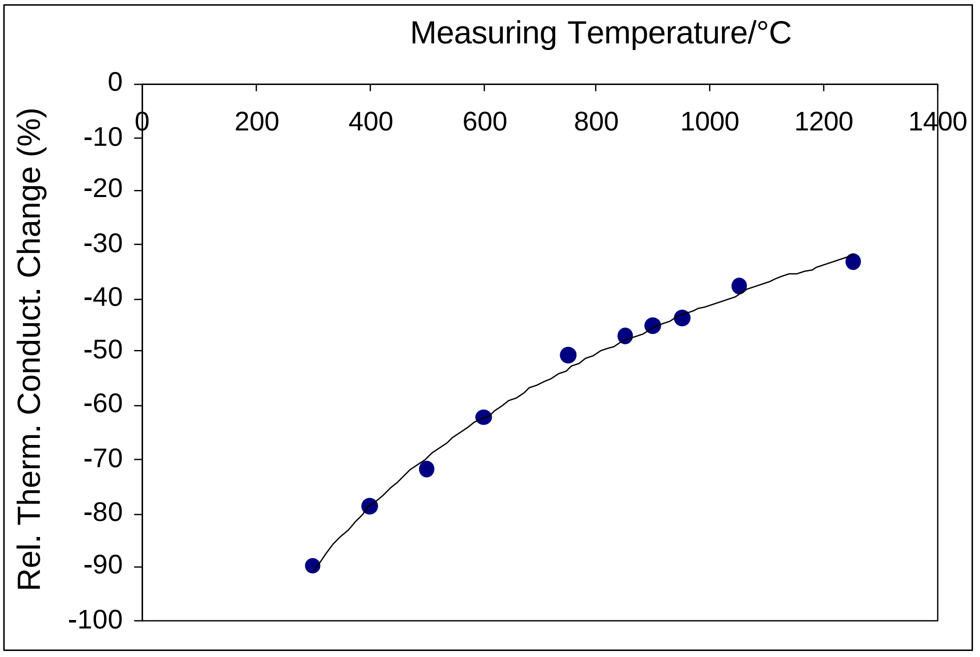


Fig. 5 – Relative thermal conductivity changes at saturation by fast neutron irradiation as a function of irradiation temperature; thermal conductivity is measured at irradiation temperature

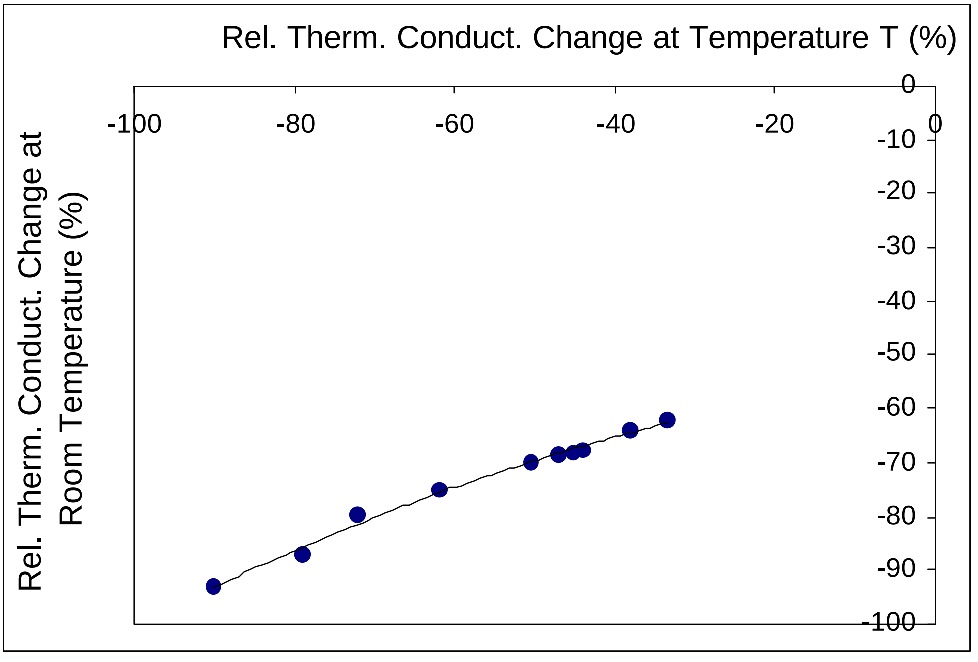


Fig. 6 – Saturation levels of relative thermal conductivity changes by fast neutron irradiation at different temperatures; correlation between the saturation levels determined at room temperature vs saturation levels determined at irradiation temperature.

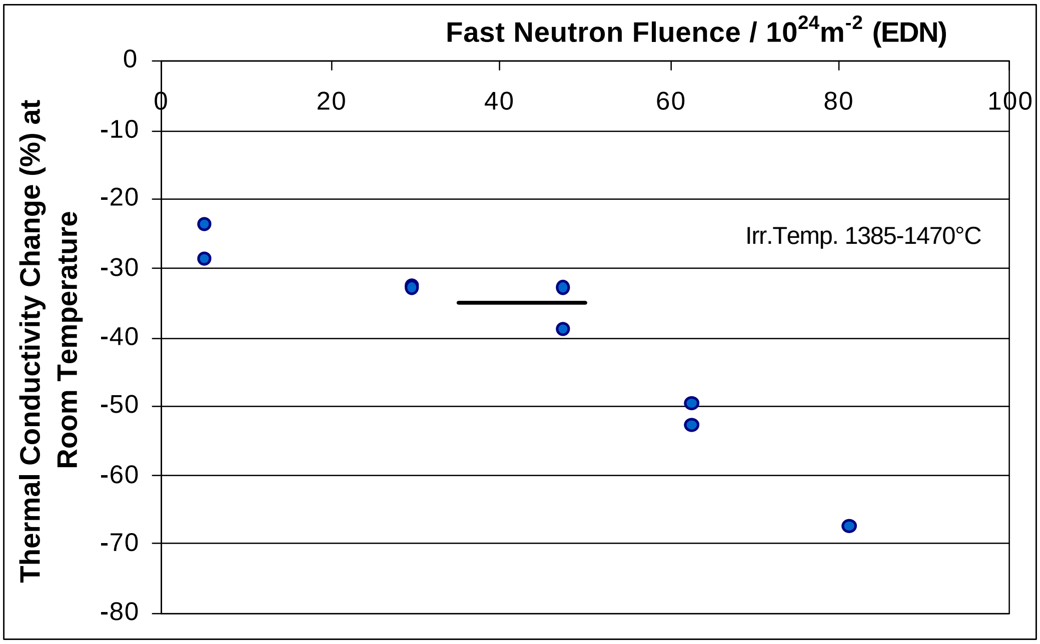


Fig. 7 – Irradiation induced thermal conductivity change of a Gilsographite, irradiated at about 1400°C, measured at room temperature; the black bar indicates the assumed saturation level

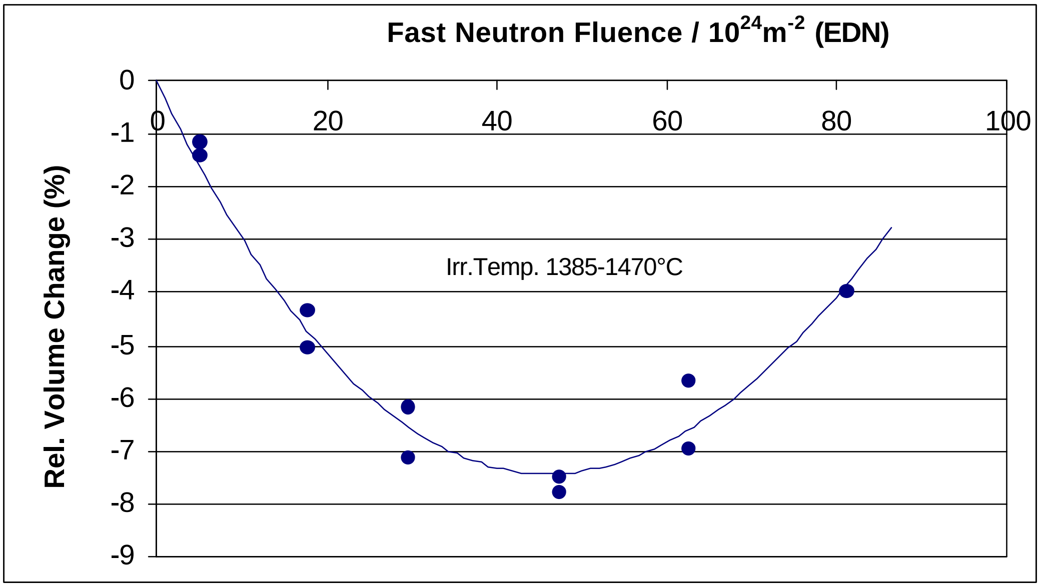


Fig. 8 – Irradiation induced volume change of a Gilsographite at about 1400°C irradiation temperature