Thermal, Structural, and Radiological Properties of Irradiated Graphite from the ASTRA Research Reactor - Implications for Disposal[1]

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ABSTRACT

There is currently no consensus regarding the disposal of nuclear graphite. The two main problems include high activities of C-14 and H-3 as well as accumulation of Wigner energy (responsible for the Windscale Pile 1 fire in 1957). The release of Wigner energy from the graphite of the inner thermal column of the ASTRA research reactor has been studied by differential scanning calorimetry and simultaneous differential scanning calorimetry / synchrotron powder x-ray diffraction between 25 ^{o}C and 725 ^{o}C at a heating rate of 10 ^{o}C.min^{-1}. The graphite, having been subject to a fast-neutron fluence from ~10^{17} to ~10^{20} n.cm^{-2} over the life time of the reactor at temperatures not exceeding 100 ^{o}C, exhibits Wigner energies ranging from 25 to 572 J.g^{-1} and a Wigner energy accumulation rate of ~7 x 10^{-17} J.g^{-1}/n.cm^{-2}. The shape of the rate-of-heat-release curves, e.g., maximum at ca. 200 ^{o}C and a fine structure at higher temperatures, varies with sample position within the inner thermal column, i.e., the distance from the reactor core. Crystal structure of samples closest to the reactor core (fast-neutron fluence > 1.5 – 5.0 x 10^{19} n.cm^{-2}) is destroyed while that of samples farther from the reactor core (fast-neutron fluence < 1.5 – 5.0 x 10^{19} n.cm^{-2}) is intact. The dependence of the c lattice parameter on temperature between 25 ^{o}C and 200 ^{o}C as determined by Rietveld refinement for the nonamorphous samples leads to the expected microscopic thermal expansion coefficient along the c axis of ~26 x 10^{-6} ^{o}C^{-1}. However, at 200 ^{o}C, coinciding with the maximum in the rate-of-heat-release curves, the rate of thermal expansion abruptly decreases indicating a crystal lattice relaxation. The C-14 activity in the inner thermal column graphite ranges from 6 to 467 kBq.g^{-1}. Prior to interim storage or final disposal, thermal treatment of graphite irradiated under similar conditions to a fast-neutron fluence higher than ~5 x 10^{19} n.cm^{-2} should be considered. The graphite of the inner thermal column of the ASTRA research reactor has been treated by heating to 400 ^{o}C for 24 h prior to interim storage. This effectively removes 90 - 100% of the Wigner energy. Generally, incineration might still be the most reasonable disposal option for irradiated graphite considering the large waste volume reduction factor and the literal disappearance of the Wigner energy issue. However, questions concerning releases of C-14 need to be addressed.

[1] This is a condensed version of a more detailed article on the subject [1], with permission from Elsevier.

INTRODUCTION

The Wigner energy content (responsible for the Windscale Pile 1 fire in 1957) of any irradiated graphite slated for disposal should be assessed and taken into account in selecting and implementing an appropriate treatment and disposal option. The presence of significant quantities of ^{14}C in irradiated graphite presents additional difficulties in this respect.

The ASTRA reactor (Fig. 1.) – a 10 MW light water moderated and cooled pool type research reactor in operation in Seibersdorf, Austria, between 1960 and 1999 – is undergoing decommissioning. The bulk of the resulting material exhibits activities below applicable clearance levels, e.g., the concrete blocks cut from the biological shield, with the exception of activated core- and close-to-core structures as well as some radioactively contaminated components. The activated and/or contaminated material is being treated, conditioned, and stored on site. This applies, *inter alia*, to approximately 10 tons of reactor-grade graphite from the inner and outer thermal columns of the reactor in the form of blocks 10 cm x 10 cm x (60-100) cm. Over the 40 years of reactor operation, the graphite has been exposed to significant fast-neutron fluence, roughly estimated at up to 10^{20} n.cm^{-2} at temperatures not exceeding 100 ^{o}C. Preliminary measurements also suggested that the activity of the main slow-neutron activation product, C-14, in the graphite from the outer thermal column was on the order of 1 kBq.g^{-1}, with other radionuclides, e.g., Co-60 and Eu-152, present in trace amounts. Larger activities were expected in the inner thermal column graphite.

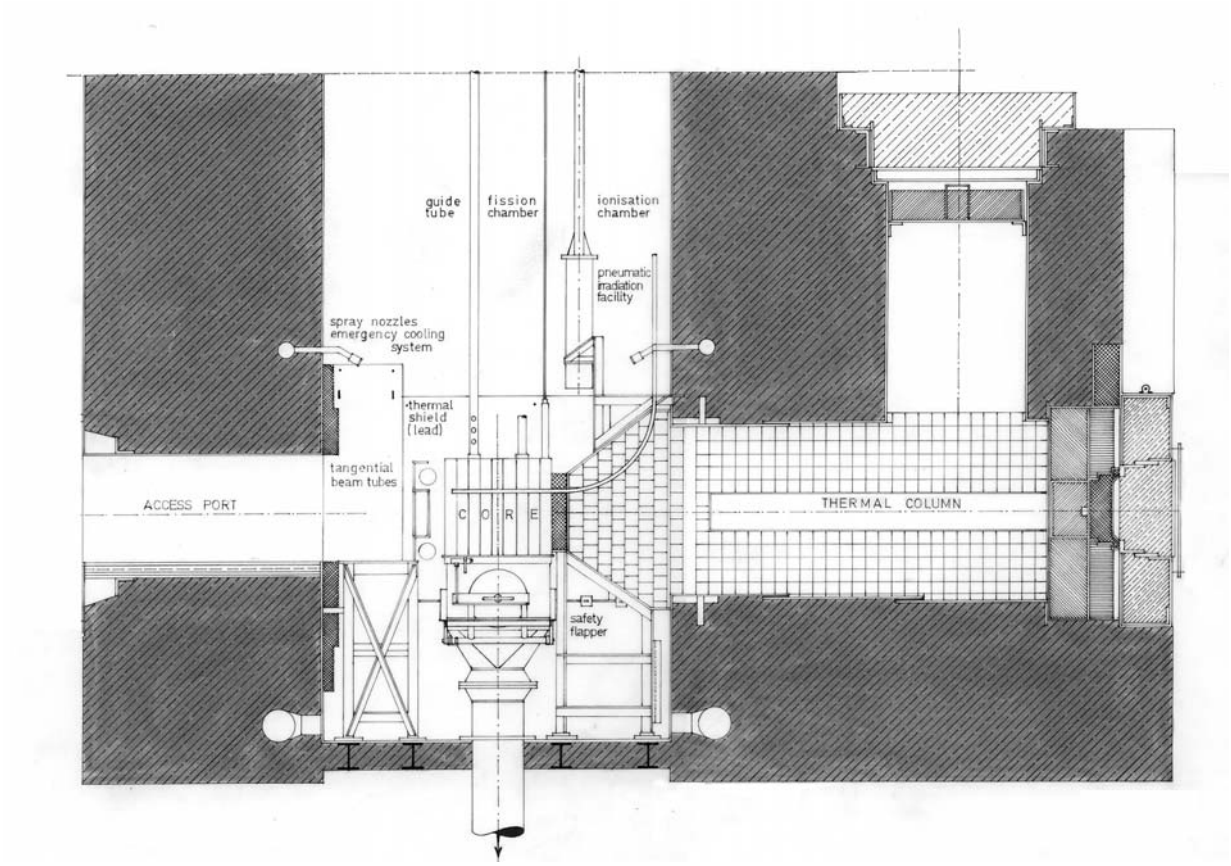

Fig. 1. Vertical section of the ASTRA research reactor block

To support the treatment, conditioning, interim storage, and final disposal of this material, it was necessary to quantify the Wigner energy content of the material and to understand the release of Wigner energy in the course of a relatively fast heat-up to a high temperature, emulating the thermal treatment of the material. (It became clear after the first heat-up experiments with individual blocks in a hot-cell facility that significant amounts of Wigner energy were indeed being released since the temperature indicated by a thermocouple embedded in the block was higher than the furnace control thermocouple during the first heat up but lagged behind it during the second one. Hence, storage of the material without thermal treatment was deemed unacceptable.) In addition to addressing these practical concerns, there was the possibility of gaining deeper insight into the Wigner energy phenomenon *per se*. This was due, in particular, to the availability of estimated fast-neutron fluence data for any position within the graphite thermal columns. Because of the gradual decay of the fluence with distance from the reactor core, samples of graphite spanning a wide range of fluences were available. Hence, the amount of Wigner energy stored and released and associated crystal structure changes in the graphite could be studied as a function of fluence.

EXPERIMENTAL

The pyramidal inner thermal column, with the maximum expected accumulation of Wigner energy, has been sampled by core drilling at 7 positions along the column's longitudinal axis labeled a through g, with distances from the surface near the reactor core ranging from 0.0 (g) to ~0.6 (a) m. Five samples from each position were machined on a lathe at reduced rpm with alcohol cooling so that the temperature is not expected to have exceeded 25 ^{o}C.

DSC experiments were performed with power-compensated instruments PerkinElmer DSC-7 and PerkinElmer Pyris Diamond. The DSC-7 settings were: ice-water cooling (0 ^{o}C), 99.9999 % Ar purge gas at 20 $cm^3.min^{-1}$, temperature range 25 – 625 ^{o}C, heating rate 10 $^{o}C.min^{-1}$, and sample weight ~65 mg. The Pyris Diamond settings were: circulating-liquid chiller cooling (-5 ^{o}C), 99.9999 % Ar purge gas at 20 $cm^3.min^{-1}$, temperature range 25 – 725 ^{o}C, heating rate 10 $^{o}C.min^{-1}$, and sample weight ~80 mg. DSC/XRD experiments were performed on the Sector 10 insertion device beam line (Materials Research Collaborative Access Team) at the Advanced Photon Source, Argonne National Laboratory, with an instrument essentially consisting of a PerkinElmer DSC sample holder modified to allow for simultaneous XRD, attached to a diffractometer, and a remotely connected PerkinElmer Pyris 1 DSC instrument. The Pyris 1 DSC settings were: circulating-liquid chiller cooling (-10 ^{o}C), 99.9999 % Ar purge gas at 20 $cm^3.min^{-1}$, temperature range 25 – 525 ^{o}C, heating rate 10 $^{o}C.min^{-1}$, and sample weight ~65 mg. Each DSC scan was accompanied by a full 2θ XRD scan every 10 ^{o}C. A cryogenically-cooled Si(111) double-crystal monochromator was used to select the incident beam wavelength of 0.082650 nm (15.000 keV). Higher energy harmonics were rejected by a polished, uncoated, ULE glass mirror. The resolution of the instrument was set by the convolution of the incident beam size (0.35mm x 0.50mm V x H) with the acceptance of the double slit collimator in front of the detector (0.65mm x 2.5mm, 650mm from the sample). The resulting 2θ resolution was about 0.06° (FWHM). X-rays were detected by an Oxford-Danfysik NaI scintillation detector. The detector was scanned from 4 deg to 45 deg at a rate of 1 $deg.s^{-1}$, with data points acquired in 0.015 s bins, for a data point spacing of 0.015 deg. XRD patterns were evaluated by Rietveld refinement. In both DSC and DSC/XRD experiments, two identical scans were performed with each sample. Subtraction of the second-run from the first-run data, division by sample mass, and integration over a specified temperature range, yields the specific Wigner energy released in the first run.

The C-14 analyses were performed by combustion of ~300 mg graphite powder that remained after machining the DSC and DSC/XRD sample disks at 800°C in an O_2 atmosphere within a Carbolite® oven. The off-gases were bubbled through two impingers each filled with 40 ml 4 N NaOH solution in order to absorb the CO_2. 1-ml aliquots of these solutions were mixed with 15 ml Ultima Gold XR liquid scintillation cocktail and counted in a Wallac Quantulus liquid scintillation counter.

THEORY AND CALCULATIONS

Displacement of atoms from their normal lattice positions in solids by fast particles was first predicted by E. P. Wigner in 1942. He showed that fast neutrons produced in the fission of uranium would possess enough energy to displace ~2 x 10^4 carbon atoms per neutron. Subsequent calculations by Seitz put the number closer to ~2 x 10^3. The resulting entrapment of the displaced atoms at non-lattice points and the creation of vacancies are accompanied by a number of changes in physical characteristics of the solid, such as a decrease in thermal conductivity and *a* lattice parameter, and an increase in elastic modulus, electrical resistance, breaking strength, *c* lattice parameter and internal (Wigner) energy.

Upon heating to sufficiently high temperatures, the displaced atoms presumably diffuse back to the vacancies. The energy, released as heat, can be measured by standard calorimetry techniques. The results of numerous previous measurements of Wigner energy have been recounted elsewhere [1]. The effective heat capacity of irradiated graphite, c_p', is defined as:

$$c_p' = c_p - \frac{dH}{dT} \qquad \text{(Eq. 1)}$$

where c_p is the normal heat capacity of graphite and dH/dT is the heat release per unit temperature rise. If c_p' is negative over a temperature range, the material is self-heating and the possibility of a large temperature rise exists. For simplicity, the worst-case scenario of an adiabatic energy release at a constant heat capacity is considered here. The associated temperature rise is then given by:

$$\Delta T_{\max} = \frac{\Delta H_{Wigner}}{c_p} \qquad \text{(Eq. 2)}$$

where the value of c_p be taken as 1 $J.^oC^{-1}.g^{-1}$. (c_p actually varies between 1 $J.^oC^{-1}.g^{-1}$ at 100 ^oC and ~2 $J.^oC^{-1}.g^{-1}$ at 1000 ^oC.) Reaching temperatures above ~400 ^oC is considered sufficient for thermal oxidation.

The dimensional changes and the thermal expansion coefficient of irradiated graphite have also been studied previously [1]. Generally, expansion in the direction perpendicular to the basal planes (*c*) and contraction in the direction parallel to them (*a*) has been observed, although the relative magnitudes of the changes depend upon the exposure conditions. X-ray diffraction patterns of highly irradiated graphite exhibit broad and asymmetric peaks suggesting a decrease in the degree of crystallinity.

There is still mostly speculation about the nature of the Wigner energy buildup and release at the atomic level. The recombination of interstitials and vacancies, also known as Frenkel pairs, is considered to be the key step in Wigner energy release. In quantum-mechanical studies, the energy of a widely spaced Frenkel pair has been calculated as ~13 – 15 eV and that of a close-bound or intimate Frenkel pair as ~10 – 11 eV. The recombination of the intimate Frenkel pair is suggested as the cause of the Wigner energy release peak observed around 200 ^{o}C [1].

The connection between the fast-neutron fluence, Φ_f, and the number of atoms displaced, *i.e.*, the number of Frenkel defects formed, is provided by the displacement cross section, σ_d:

$$n_F = \sigma_d \cdot \Phi_f \qquad \text{(Eq. 3)}$$

with n_F equivalent to the displacements per atom (dpa) if dpa < 1. The displacement cross section of graphite (and other materials) has been the subject of intense study. The value for graphite is 11.5 x 10^{-22} cm^2. Hence, using Eq. 3., the dpa can be calculated for any sample a through g provided the value of Φ_f at sample location is known. At the same time, a simple relationship between Wigner energy, ΔH_{Wigner} [$J.g^{-1}$], Frenkel defect energy, h_F [eV], and n_F can be established:

$$n_F = \frac{M_C}{N_A} \frac{\Delta H_{Wigner}}{1.602 \cdot 10^{-19} h_F} \qquad \text{(Eq. 4)}$$

where N_A is Avogadro's number, and M_C is the molar mass of graphite. This equation enables the dpa calculation from the experimentally determined ΔH_{Wigner} and the theoretical value of h_F. Ideally, the dpa values obtained from Eq. 3. and Eq. 4. should be identical. Any discrepancies would point to inaccuracies in one or more of the values of ΔH_{Wigner} (directly measured), Φ_f (indirectly measured), σ_d (indirectly measured), and h_F (theoretical).

In order to be able to correlate the thermal and structural data obtained in the course of this work with fast-neutron fluence, its profile within the inner thermal column was needed. The original idea was to reconstruct the fluence profile from the available reactor operation records. However, having the calculations performed turned out to be a major effort associated with very high cost (~$300,000). As a result, the fluence was estimated from all available information, including previous measurements at various irradiation positions, reactor core and thermal column geometry, and the known attenuation characteristics of water (pool), lead (shield), aluminum (liner), and graphite for fast neutrons (see Fig. 1). The fast-neutron fluence (E > 0.1 MeV) within the inner thermal column spans the range from 1.7 x 10^{20} $n.cm^{-2}$ vis-à-vis the reactor core down to 6.4 x 10^{16} $n.cm^{-2}$ at the rear face of the thermal column.

Even though scattering of fast neutrons is responsible for the Wigner energy buildup in graphite, the activity of ^{14}C – the main thermal neutron activation product in graphite – has also been measured. The presence of significant quantities of ^{14}C in irradiated graphite is the second major factor in selecting and implementing any disposal option.

RESULTS AND DISCUSSION

A set of DSC plots obtained with one set of samples a through g in the course of a DSC/XRD experiment is shown in Figure 2. Samples a through e exhibit a maximum rate of heat release around 200 ^{o}C and a negligible heat release at 525 ^{o}C. The character of the DSC plot obtained with samples f and g is entirely different in that no sharp peak at 200 ^{o}C is present and significant heat release continues at 525 ^{o}C. The graphite c_p curve indicates that for samples c, d, e, and g, the effective heat capacity, c_p', as defined by Eq. 1 is negative over an increasing temperature range beginning at ~175 ^{o}C with the associated potential for self-heating. Interestingly, even though sample f exhibits the second largest Wigner energy release, its rate-of-heat-release curve is such that c_p' is positive throughout the temperature range. In addition, samples a through c show two additional peaks at higher temperatures initially described by Iwata [8]. This so called fine structure is apparently a signature of processes with higher activation energy than the Frenkel pair recombination responsible for the 200 ^{o}C peak. The elucidation of these processes would, however, require a separate study.

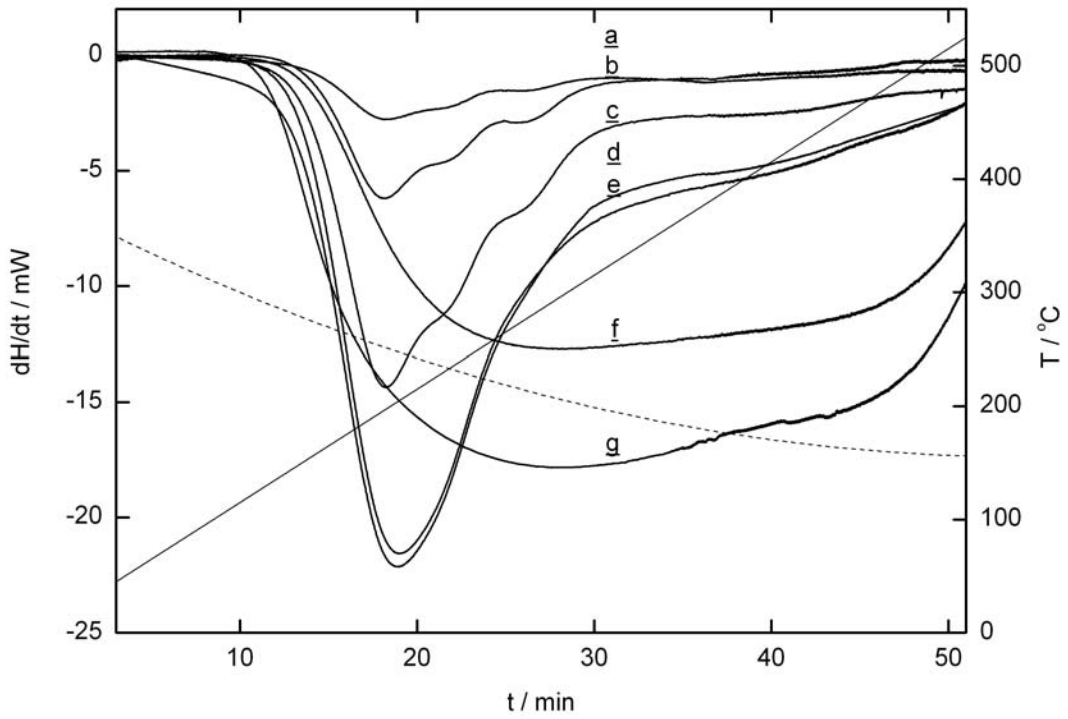

Fig. 2. DSC curves obtained between 25 ^{o}C and 525 ^{o}C at 10 ^{o}C.min^{-1}. Heavy line – heat flow, thin line – temperature, dashed line – heat flow due to c_p of graphite

Integration of the DSC plots between 25 ^{o}C and 525 ^{o}C yields directly the Wigner energy released within this temperature range. Even though some of the data extend to 625 or 725 ^{o}C, the upper integration limit was kept at 525 ^{o}C for consistency. Furthermore, the Wigner energy release above 525 ^{o}C at 10 ^{o}C.min^{-1}

proved to be insignificant. The results are given in Table I as well as in Figure 3. Each data point is an average of measurements on five samples. The Wigner energy ranges from 25 $J.g^{-1}$ for sample a to 572 $J.g^{-1}$ for sample g. The slope of the ΔH_{Wigner} *vs.* Φ_f curve at zero fluence (Wigner energy accumulation per unit of fluence in the linear region) is ~7 x 10^{-17} $J.g^{-1}/n.cm^{-2}$. The adiabatic temperature rise according to Eq. 2 is also given in Table I. Samples g and possibly f could conceivably undergo thermal oxidation. However, only sample g has been shown to be self-heating over a wide temperature range. Thus it is the only sample presenting a real hazard in this respect. These considerations suggest that, prior to interim storage or final disposal, thermal treatment of the ASTRA graphite as well as graphite irradiated under similar conditions to a fast-neutron fluence higher than ~5 x 10^{19} $n.cm^{-2}$ should be considered. It is interesting to note that the Wigner energy release exhibited by sample d is significantly higher than a simple interpolation between c and e would suggest. This would imply a fast-neutron fluence peak in the middle of the inner thermal column – a rather unusual proposition.

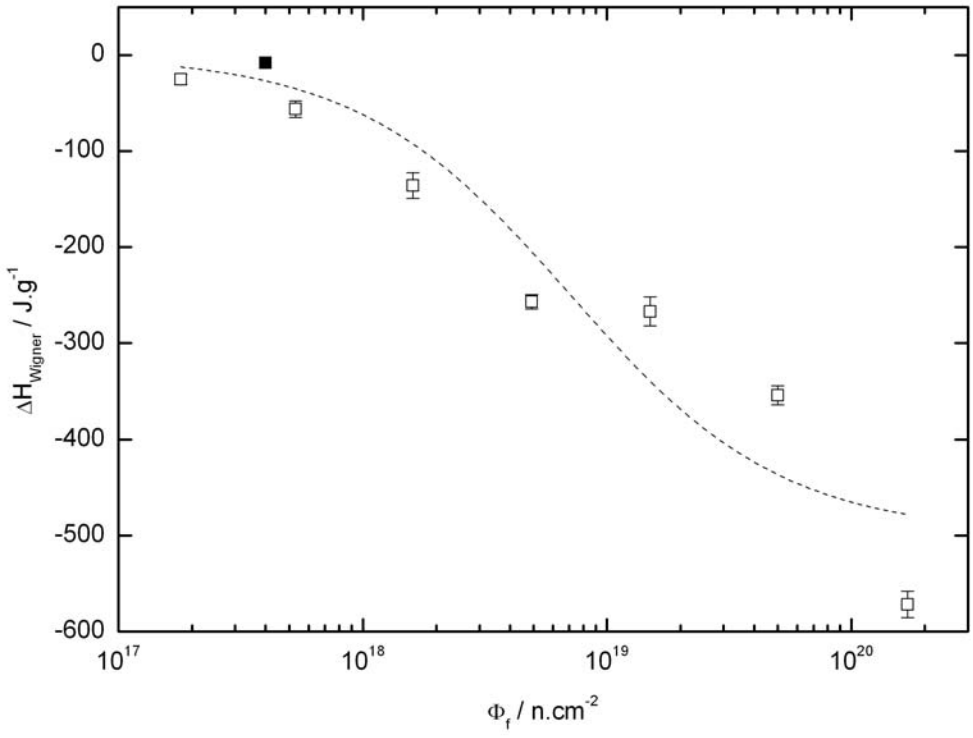

Fig. 3. Wigner energy released between 25 °C and 525 °C at 10 °C.min^{-1} as a function of fast-neutron fluence. □ – experimental data, ■ – previous work [1]

A low-angle section of a set of XRD patterns taken at 25 °C at the beginning of the DSC/XRD scans is shown in Figure 4. The 002 graphite diffraction peak at ~14 deg is very sharp in samples a through e and its position is such that the *c* lattice parameter increases in the same order. The 002 peak is missing in samples f and g, being replaced by a single broad peak at ~12.2 deg (*c* = 0.778 nm). This correlates well with the different DSC plot character in samples f and g. Hence, a fast-neutron fluence of somewhere between 1.5 x 10^{19} $n.cm^{-2}$ (e) and 5 x 10^{19} $n.cm^{-2}$ (f) was sufficient to render the original graphite crystal structure amorphous. The amorphous character of a sample increases with the number of atoms displaced

from their normal lattice positions (dpa). A rough estimate from Fig. 4 puts the dpa of samples f and g at 0.1. The dpa values for all samples calculated from Eq. 3 and Eq. 4 (with h_F = 14 eV) are given in Table I. The agreement for samples a through c is surprisingly good but deteriorates progressively for samples d through g. Considering the character of the quantities involved in Eq. 3 and Eq. 4, it seems most likely that the simple model presented by Eq. 4 breaks down at higher fast-neutron fluence values, *i.e.*, h_F is no longer constant and independent of dpa. In fact there are indications that the h_F decreases substantially for dpa > 0.1 [1]. This would explain the observed underestimation of dpa at higher fluence values by Eq. 4. Taking the dpa from Eq. 3 and substituting back to Eq. 4 yields h_F values of between 5.7 eV (d) and 0.4 eV (g).

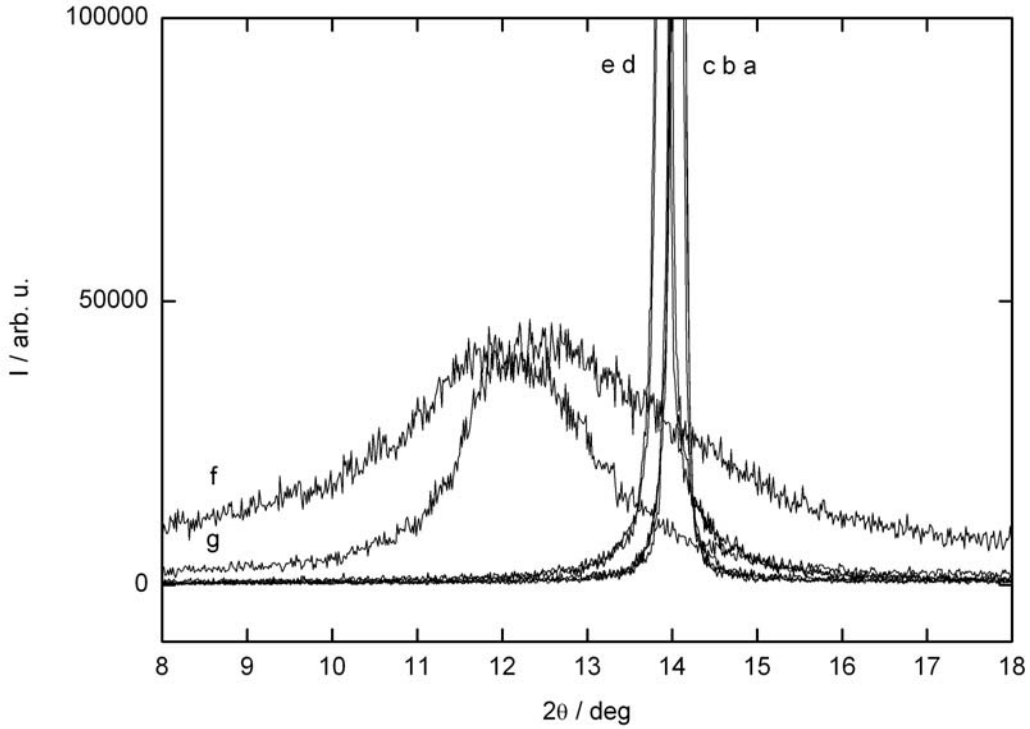

Fig. 4. Section of X-ray diffraction patterns at 25°C in the vicinity of the 002 graphite diffraction peak.

The changes in the graphite *c* lattice parameter obtained as a result of Rietveld refinement using the XRD patterns from samples a through e between 25 °C and 525 °C are shown in Figure 5. (The graphite *a* lattice parameter did not exhibit any measurable changes.) Between 25 °C and 180 – 190 °C, the samples undergo thermal expansion consistent with a microscopic thermal expansion coefficient along the *c* axis of ~26 x 10^{-6} °C^{-1} – in line with the literature values. The break in the curves at 180 – 190 °C, indicating a crystal lattice relaxation, coincides with the inflection point of the rate-of-heat-release peak at 200 °C in the DSC curves. Hence, the Wigner energy release at 200 °C is clearly a result of the lattice relaxation. The effective thermal expansion coefficient of samples d and e above 300 °C is ~12 x 10^{-6} °C^{-1}. Rough linear extrapolation indicates that the *c* lattice parameter of these samples would reach that of sample a at ~1000 °C.

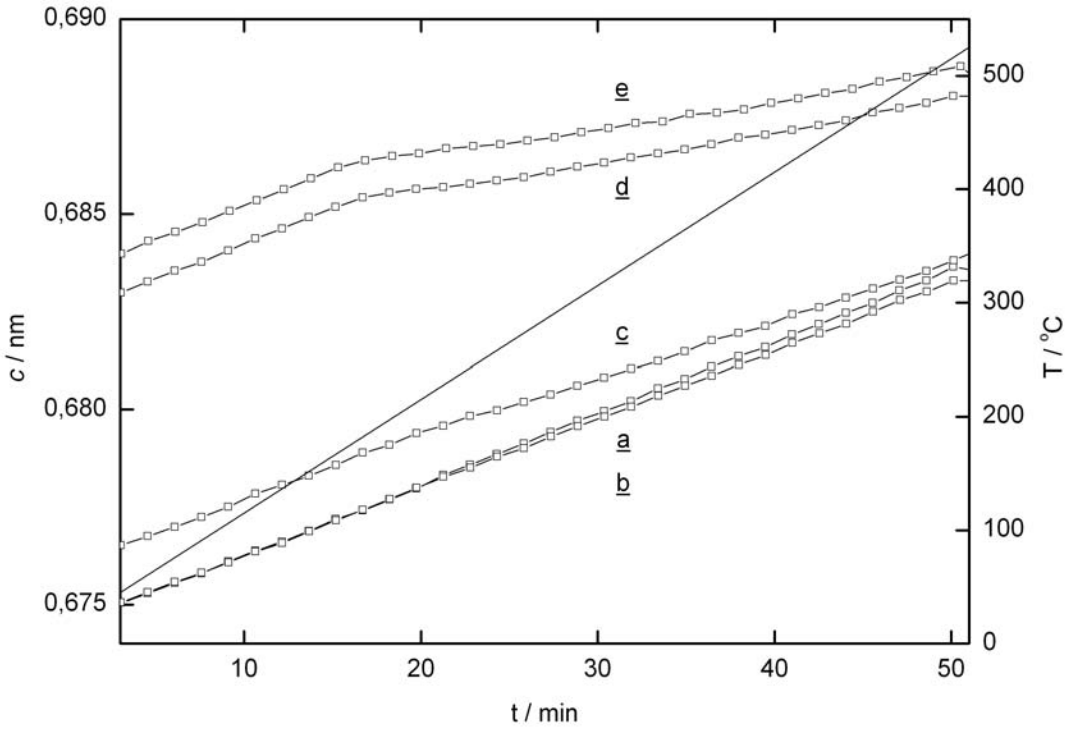

Fig. 5. Graphite c lattice parameter from Rietveld refinement

Results of the C-14 activity measurements in the inner thermal column graphite are shown in Table I and Fig. 6. Each data point is an average of 6 measurements on each sample aliquot. As expected, the activities are significantly higher than the 1 $kBq.g^{-1}$ measured in the outer thermal column and range from 6 $kBq.g^{-1}$ for sample a to 467 $kBq.g^{-1}$ for sample g. Sample d in the middle of the inner thermal column exhibits an unusually high C-14 activity. This would imply a thermal neutron fluence peak in the middle of the inner thermal column – a well-known phenomenon in reactor moderator material, as opposed to the fast-neutron fluence peak necessary to explain the anomalous behavior of the sample d as far as Wigner energy is concerned. The thermal neutron fluence entering the inner thermal column (g) has been estimated at 1.1 x 10^{22} $n.cm^{-2}$. Taking into account only the C-13(n,γ)C-14 reaction, this would correspond to a C-14 activity of ~20 $kBq.g^{-1}$. Consequently, the N-14(n,p)C-14 reaction must be responsible for the majority of the C-14 present in the thermal column. In fact, it has been shown that N-14 initially contained as air in the pores of the graphite (~24-25% porosity) would be sufficient to generate about 30 times more C-14, *i.e.*, ~ 600 $kBq.g^{-1}$. (The inner thermal column has been hermetically sealed in an aluminum liner over the life time of the reactor.) Hence, the total expected C-14 activity of ~620 $kBq.g^{-1}$ is not significantly different from the value measured in sample g.

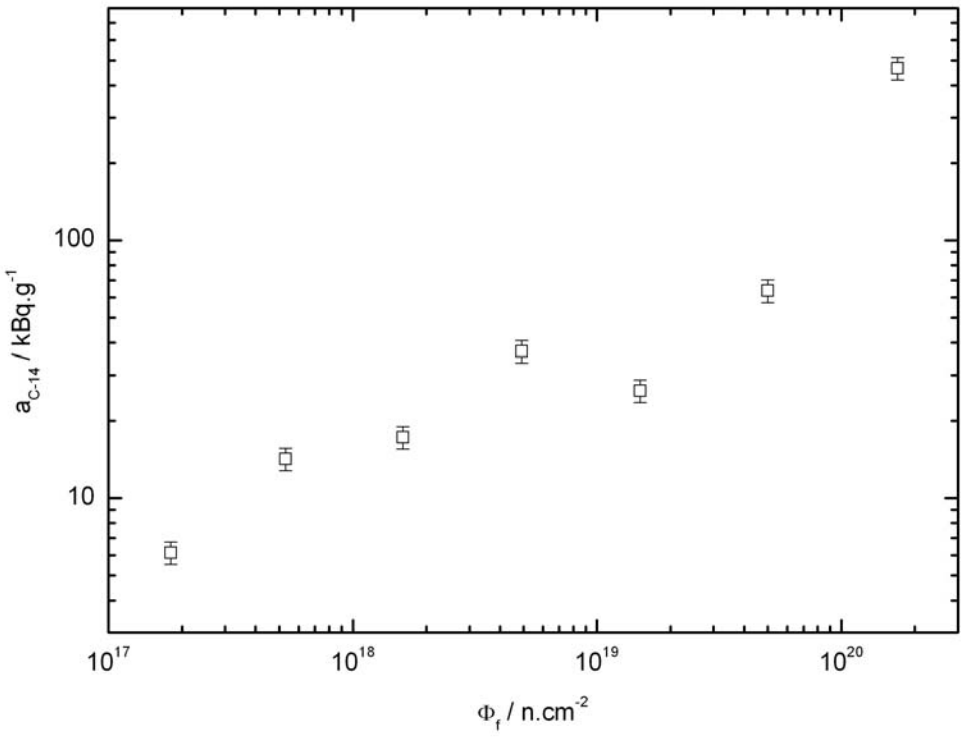

Fig. 6. C-14 activity measured by LSC as a function of fast-neutron fluence

The selection and implementation of any treatment and disposal option for irradiated graphite is determined by the Wigner energy content of the material and the potential for self-heating as well as by the inventory of any activation products, C-14 and H-3 in particular. Even though these two effects are caused by different parts of the neutron spectrum, they are not completely independent and the relationship can get rather complicated. In the case of the ASTRA research reactor graphite, 25 % of the blocks from the far end of the outer thermal column exhibited activities below applicable clearance levels, *e.g.*, 80 Bq.g^{-1} for ^{14}C. Since Wigner energy accumulation was not a concern either, this material has been cleared and will be reused at another facility. The remaining 75 % of the blocks of the outer thermal column could not be cleared because of at least one of the C-14, H-3, Co-60, and Eu-152 activities in excess of clearance levels and will be placed in interim storage on site. No thermal treatment is envisioned since, based on the Wigner energy content of the sample a, $\Delta H_{Wigner} < 25$ J.g^{-1}. The graphite of the inner thermal column exhibits even higher activities and a significant Wigner energy content. Thermal treatment is clearly indicated for at least the first 10 cm of the material. However, since this study had not been completed by the time of the thermal column removal from the reactor, the material from the entire inner thermal column has been treated by heating to 400 ^{o}C for 24 h. To contain the potential small ^{14}C releases, the heating of individual graphite blocks has been performed in a furnace placed inside a hot-cell facility. Our results indicate that this is certainly sufficient for eliminating the Wigner energy problem.

Table I. Sample Characteristics, Measured and Derived Quantities

Sample	d / cm	Φ_f / n.cm^{-2}	dpa (Eq. 3)	ΔH_{Wigner} / J.g^{-1}	dpa (Eq. 4)	ΔT_{max} / ^{o}C (Eq. 2)	*c* at 25 ^{o}C / nm	a_{C-14} / kBq.g^{-1}
g	52.2	1.7×10^{20}	2.0×10^{-1}	-572 ± 14	5.1×10^{-3}	570	0.778[a]	467 ± 47
f	62.0	5.0×10^{19}	5.8×10^{-2}	-354 ± 10	3.1×10^{-3}	350	0.778[a]	64 ± 6
e	71.7	1.5×10^{19}	1.7×10^{-2}	-267 ± 15	2.4×10^{-3}	270	0.684	26 ± 3
d	81.5	4.9×10^{18}	5.6×10^{-3}	-257 ± 7	2.3×10^{-3}	260	0.683	37 ± 4
c	91.3	1.6×10^{18}	1.8×10^{-3}	-136 ± 13	1.2×10^{-3}	140	0.676	17 ± 2
b	101.1	5.3×10^{17}	6.1×10^{-4}	-56 ± 9	5.0×10^{-4}	60	0.675	14 ± 1
a	110.8	1.8×10^{17}	2.1×10^{-4}	-25 ± 5	2.2×10^{-4}	30	0.675	6 ± 1

[a] Calculated from the Bragg equation $\lambda = 2(c/2).\sin\theta$ for $2\theta = 12.2$ deg and $\lambda = 0.082650$ nm.

CONCLUSION

The shape of the DSC rate-of-heat-release curves, *e.g.*, maximum at ca. 200 ^{o}C and a fine structure at higher temperatures, varies with sample position within the inner thermal column, i.e., the distance from the reactor core. The magnitude of the Wigner energy ranges from 25 to 572 J.g^{-1}, the linear Wigner energy accumulation rate is ~7 x 10^{-17} J.g^{-1}/n.cm^{-2}, and the deviation from linearity begins at a fast-neutron fluence of ~8 x 10^{17} n.cm^{2}. Only the first ~10 cm of graphite closest to the reactor core present a hazard with respect to self-heating and subsequent thermal oxidation. Crystal structure of samples closest to the reactor core (fast-neutron fluence > 1.5 -5.0 x 10^{19} n.cm^{-2}) is destroyed – a single amorphous peak is present in the XRD patterns at ~12.2 deg (c = 0.778 nm). Crystal structure of samples farther from the reactor core (fast-neutron fluence < 1.5 -5.0 x 10^{19} n.cm^{-2}) is intact. The dependence of the *c* lattice parameter on temperature between 25 ^{o}C and 200 ^{o}C leads to a microscopic thermal expansion coefficient along the *c* axis of ~26 x 10^{-6} ^{o}C^{-1}. However, at 200 ^{o}C, coinciding with the maximum in the DSC rate-of-heat-release curves, the rate of thermal expansion abruptly decreases indicating a crystal lattice relaxation. Hence, the Wigner energy release at 200 ^{o}C is clearly a result of the lattice relaxation. The ^{14}C activity in the inner thermal column graphite ranges from 6 to 467 kBq.g^{-1}. Prior to interim storage or final disposal, thermal treatment of graphite irradiated under similar conditions to a fast-neutron fluence higher than ~5 x 10^{19} n.cm^{-2} should be considered.

The graphite of the inner thermal column of the ASTRA research reactor has been treated by heating to 400 ^{o}C for 24 h in a hot-cell facility prior to interim storage. Generally, incineration might still be the most reasonable disposal option for irradiated graphite considering the large waste volume reduction factor and the literal disappearance of the Wigner energy issue. However, questions concerning releases of C-14 need to be addressed.

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1. Lexa, D. and Kropf, A. J. (2005). Thermal, Structural, and Radiological Properties of Irradiated Graphite from the ASTRA Research Reactor – Implications for Disposal. *Journal of Nuclear Materials*, in press.