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EFFECT OF TEMPERATURE ON RADIATION-INDUCED CONTRACTION OF REACTOR GRAPHITE*

by

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I. INTRODUCTION

In the simpler cases of graphite used as a structural element in reactors, dimensional instability can gradually affect the mechanical configuration and cause operational problems. In reactor types utilizing massive graphite bars, differential distortion may cause bar rupture due to stress variation in excess of the graphite rupture limit.

At the last Carbon Conference, we pointed out the need for a better understanding of radiation induced dimensional changes in graphite and presented the data that existed at that time.(1) Since then we have accumulated significantly more data and have improved our knowledge of exposure parameters of the high flux reactors used for accelerated testing. Also, some information has been developed on the effect of neutron spectra and the convertability of neutron exposure parameters between reactors. This latter information will be given in another Hanford paper at this conference(2). In this paper we will review the distortion behavior of graphite as a function of temperature of irradiation.

II. BACKGROUND

There are considerable differences in the distortion behavior of different graphite types. Slide 1, which was presented in our earlier paper, illustrates the physical distortion of KC, CSF, and TSGBF graphites through long exposures at low temperatures. It is seen that the direction transverse to extrusion expands on irradiation at low temperatures while in the direction parallel with extrusion contracts.

KC is Kendall coke graphite. graphitized at 2800° C.
TSGBF is Texas Lockport coke graphite, gas baked and F-purified at 2450° C.
CSF is Cleves coke graphite, graphitized at 2800° C and F-purified.
Needle coke graphites are graphitized at temperatures of 2800° C and higher. They are structurally similar to KC.

Exposures are given in either MWD/AT of fuel or nvt of neutrons with energies in excess of 0.18 mev. The conversion between MWD/AT and nvt is not constant but depends on the spectra of the reactor positions considered. For the Experimental Gas-Cooled Reactor, 1 MWD/AT = 1.3×10^{17} nvt, $E > 0.18$ for exposures generated in core positions of water moderated testing reactors. Since the conversion constant varies and is not fully determined it is risky to utilize the constant indiscriminately.

III. GENERALIZED BEHAVIOR

Taking CSF graphite as a reference, Slide 2 shows the effect of a broad temperature spectrum on the contraction rate per 10^{21} nvt, $E > 0.18$ mev. The much considered instability at high irradiation temperatures is seen to be relatively negligible compared to the effect of temperature below 300° C. From the greater than 4% expansion in the perpendicular direction at 30° C, the rate changes to negative and then proceeds through a hump around 600-800° C. In the parallel direction, the distortion becomes less negative as the temperature of irradiation increases joining the perpendicular rate at high temperatures. ~~If one considers the interaction of parallel and perpendicular behavior, two~~ curves representing the hypothetical behavior of " CALCULATED perpendicular" and " CALCULATED parallel" directions can be calculated. Using an orientation ratio of 2.3:1 and an effective binder contraction rate of 0.45 per 10^{21} which is not temperature dependent, the curves show a similar generalized behavior to the measured points except that the parallel rate is much larger than formerly. The binder contraction is applied only to the perpendicular distortion since it is hypothesized that intrinsic keying of the planer configuration would probably not allow additional parallel contraction due to the binder.

IV. CONTRACTION AT HIGH TEMPERATURES

We have studied contraction of needle-coke base graphite and CSF graphite at high temperatures in the General Electric Test Reactor. The testing was performed at high specific flux intensities such that the equivalent of 15,000-20,000 EGCR MWD/AT was accumulated in only a few months. Actual peak exposures were 2.3 x 10^{21} nvt, $E > 0.18$ mev. In these experiments we used a graphite sample arrangement as shown in Slide 3. The samples were made into quarter-round shapes and assembled four in a group, then arranged longitudinally in the capsule. One CSF reference samples was included in each group.

The samples were self-heated by gamma heating of the reactor and careful heat transfer design was used to obtain the desired irradiation temperatures. Slide 4 shows the generalized behavior of needle-coke graphite and CSF graphite as a function of the temperature of irradiation. The needle-coke graphite used was of the type fulfilling the previously advanced criteria for high temperature dimensional stability in the transverse direction to extrusion. The base coke structure was highly developed, the graphitization temperatures was over 2800° C and the particle size was as small as practical for proper fabrication. In the perpendicular direction a reduction of about 40 percent in contraction rate was achieved over the CSF reference rate.

In the parallel direction both the CSF and needle coke graphite contracted at the same rate and at a rate substantially higher than did the perpendicular direction. Unfortunately, no data exists for parallel samples exposed at temperatures in excess of 750° C.

TSGBF graphite irradiated earlier in the Materials Testing Reactor but without the benefit of accurate exposure determination had a high perpendicular contraction rate; however, considerable error may exist due to exposure uncertainty.

V. NEEDLE-COKE GRAPHITE

In Slide 4, the generalized behavior of graphite as a function of temperature was presented. The generalized curves were obtained from two high temperature irradiation experiments that were performed to determine the differences between needle-coke graphites and CSF. The lower temperature experiment operated in the range 450-850° C and produced the data shown in Slide 5. It is noted that the CSF perpendicular rate is greater at all temperatures than the needle-coke graphite rate but the needle-coke rate is the same as CSF in the direction parallel with extrusion. Another important result of this experiment was the determination of the lack of rate sensitivity due to sample selection position for parallel samples from the 16" x 16" extrusion. The parallel samples were taken from positions at the center, midway, and at the outside edge of the extrusion. Prior to experimentation, it was expected that discernable differences would exist in the contraction rate due to greater orientation of the crystallites near the edge than at the center.

Slide 6 presents an extension of the results to 1200° C but with the elimination of parallel data. The improvement of needle-coke graphite over CSF is still evident but not as greater as at lower temperatures. In this experiment, several different types of needle-coke graphite were tested that were produced by different vendors. From the results it is not possible to statistically discern differences in contraction rate between the different needle-coke graphites. This is not considered significant, however, because the variations in processing were small.

VI. SUMMARY

Graphite displays a temperature sensitive radiation damage characteristic. While the direction parallel with extrusion always contracts under irradiation, the direction perpendicular to extrusion both expands and contracts depending on the temperature of irradiation. Irradiation at temperatures less than 300° C results in large expansions in the perpendicular direction and at higher temperatures, a contraction although much smaller in magnitude. The behavior of needle-coke graphite and CSF graphite was examined over moderate exposures in the GETR and results showed needle-coke graphites to be less contracting than CSF. Details of contraction show a minimum contraction rate per 10^{21} nvt at 600-800° C for both graphite types.

Certain limitations must be placed on the data presented. They are:

1. The region of 100-450° C is still scantily developed, particularly in the case of parallel contraction.
2. Although the exposures for the high temperature data were moderately high, no knowledge presently exists of the continuing rate at higher exposures. We believe that there is adequate justification for reasonable extrapolation.
3. Neutron intensity dependence has been studied at low temperatures but work remains to be done at elevated temperatures. A strong but unlikely intensity dependence could modify conclusions on these data.

Further work is planned to extend the exposures to the equivalent of 100,000 MWD/AT or until saturation of damage occurs. Also, information in the temperature range of 100-400° C is being further developed.

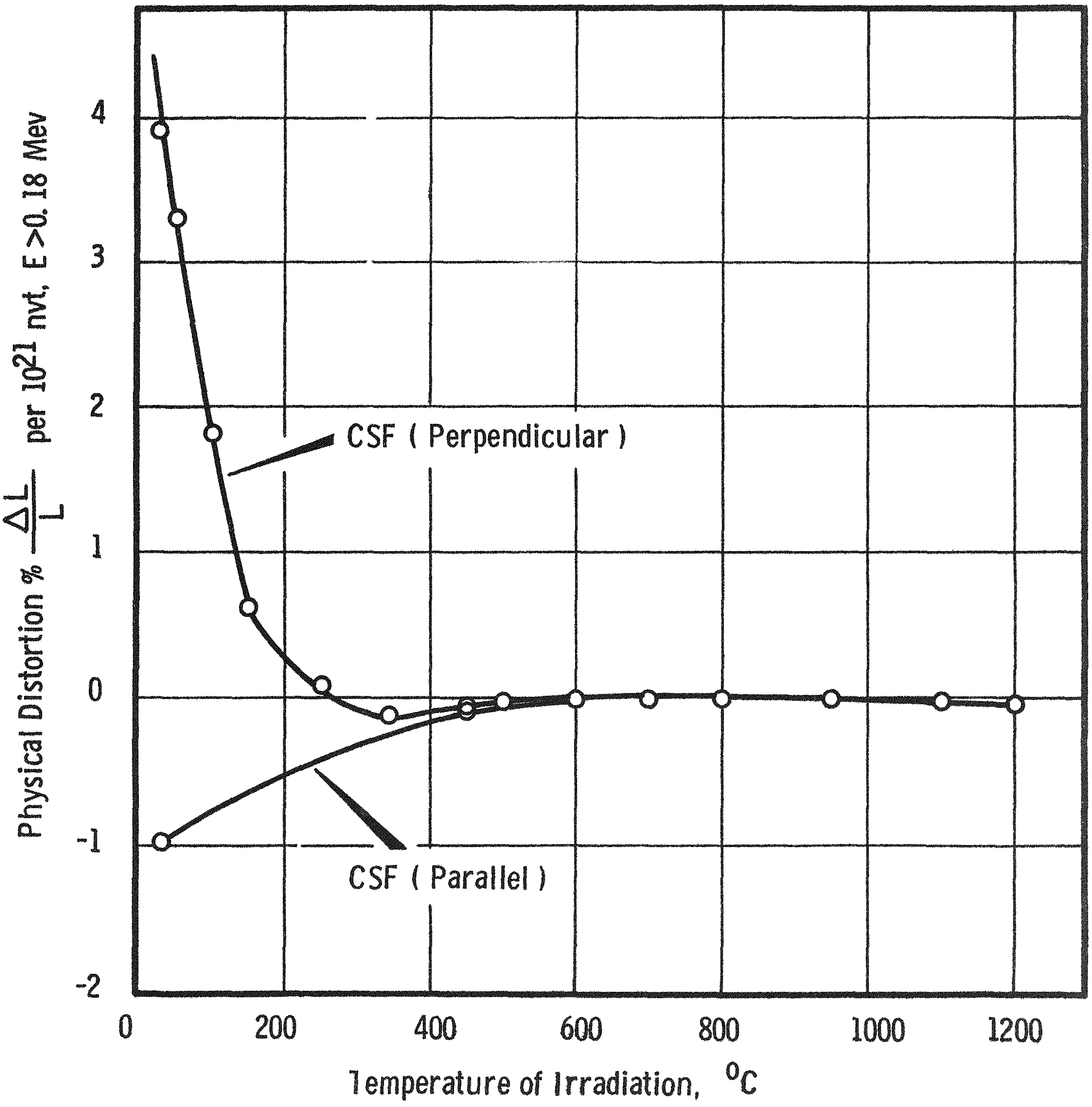
The curves presented should not be considered quantitatively firm since continuing effort is being expended to develop a better understanding and confidence in all segments. The dip between 300 and 400° C is a case in point. We do not have explicit data in that temperature increment although the cip is not unreasonable when our existing data above and below the range is joined. Also, some theoretical justification exists for the dip which will be presented in a later paper[(2)]. The segment 450-1200° C, for the exposure given, is quite adequately backed by data. The data constituting the range 50 to 300° C is technically limited because of its relatively low exposure. The basic fact that temperatures below 300° C cause expansion in the transverse direction is not, however, subject to question. Also, the parallel contraction at low temperatures is fully demonstrated but some qualification is necessary to the rate.

REFERENCES

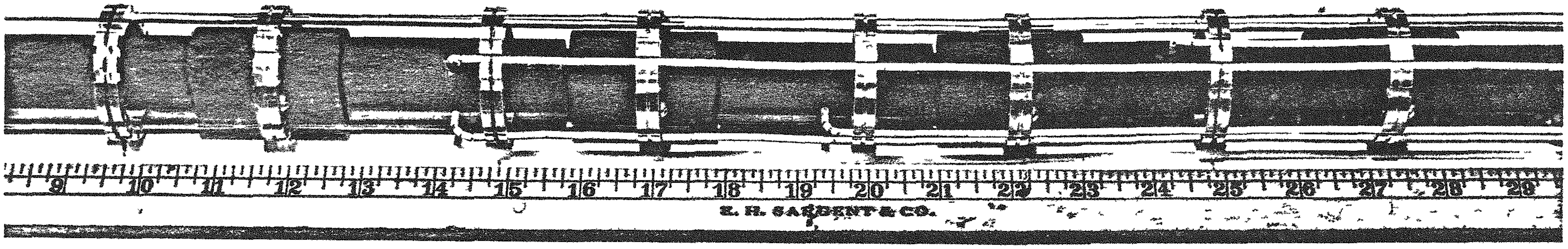
1. DAVIDSON, J. M., WOODRUFF, E. M. and YOSHIKAWA, H. H., High Temperature Radiation Induced Contraction in Graphite, Proc. of the Fourth Carbon Conference, 599-605 (1960).

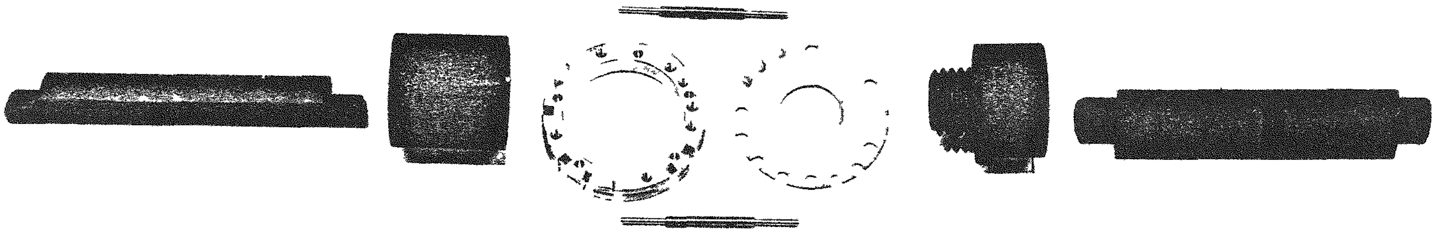
2. de HALAS, D. R. and YOSHIKAWA, H. H., Mechanism of Radiation Damage to Graphite at High Temperatures, Fifth Carbon Conference.

EFFECT OF IRRADIATION TEMPERATURE ON GRAPHITE PHYSICAL DISTORTION

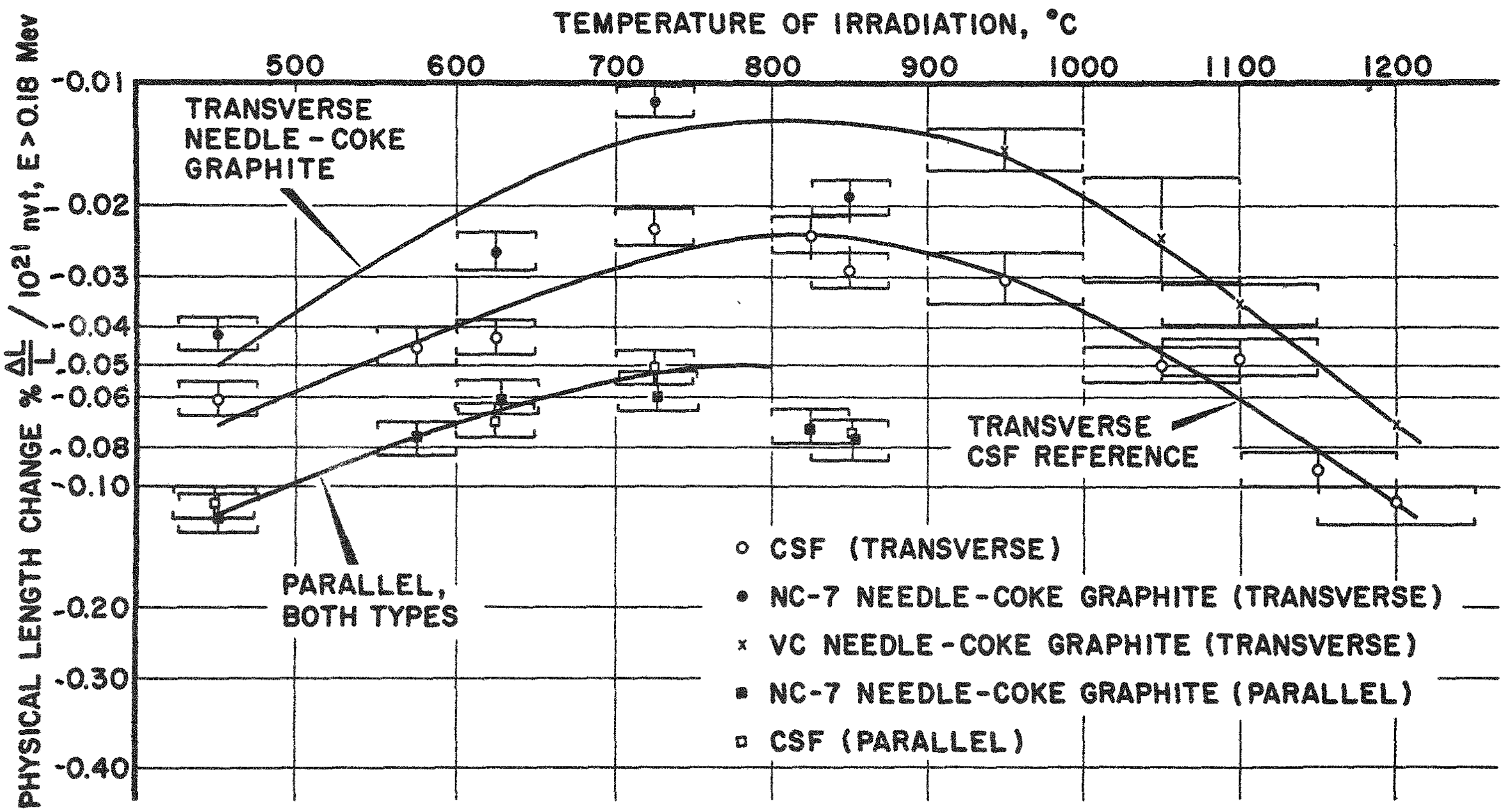


INTERNAL VIEW — HIGH TEMPERATURE
GRAPHITE IRRADIATION CAPSULE — GEH-13-7

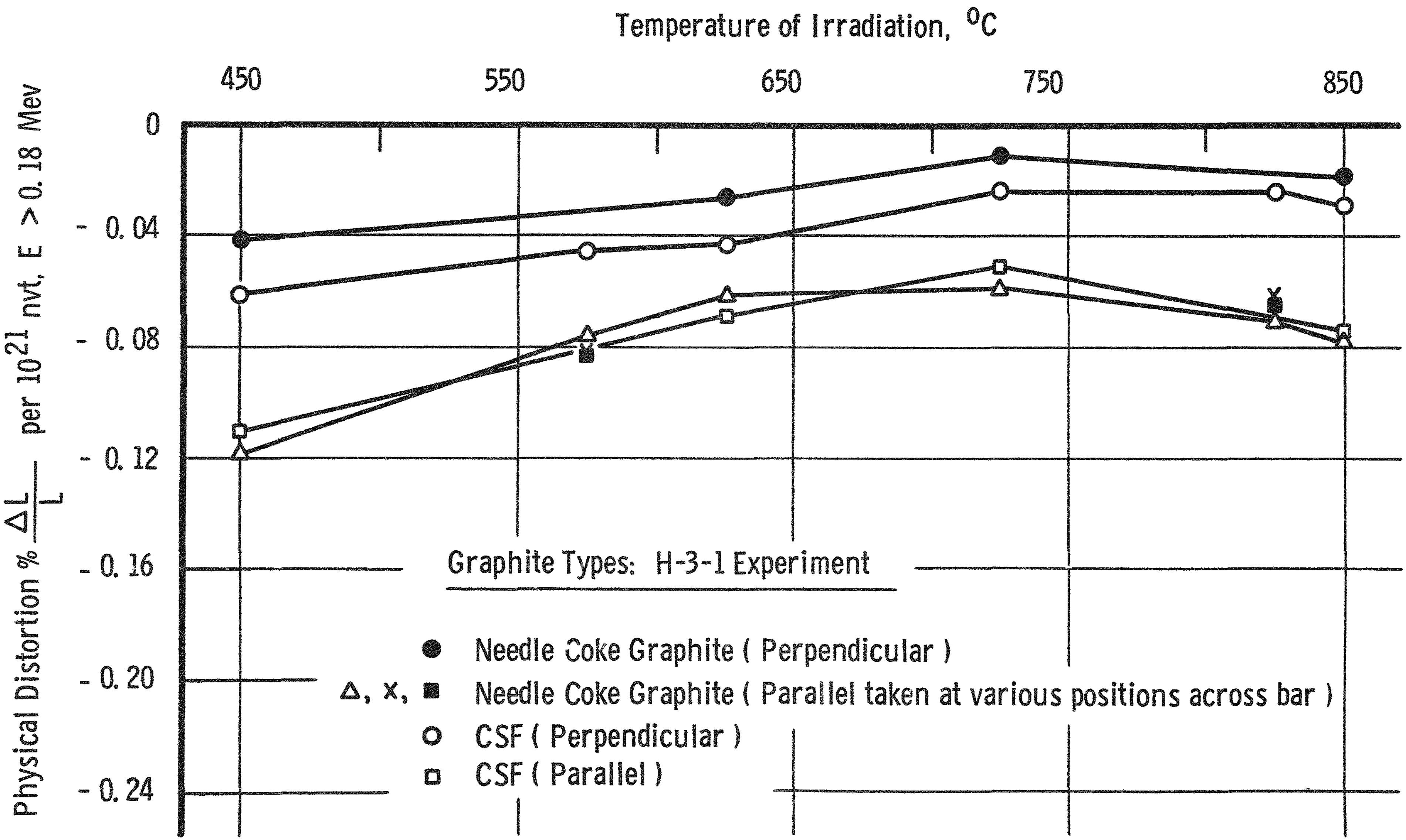




PHYSICAL DISTORTION OF REACTOR GRADE GRAPHITES AT ELEVATED TEMPERATURES



EFFECT OF TEMPERATURE OF IRRADIATION ON PHYSICAL DISTORTION



EFFECT OF TEMPERATURE OF IRRADIATION ON PHYSICAL DISTORTION