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EFFECT OF MASSIVE NEUTRON EXPOSURE ON THE DISTORTION OF REACTOR
GRAPHITE

AUTHOR

J. W. Helm and J. M. Davidson

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EFFECT OF MASSIVE NEUTRON EXPOSURE ON THE DISTORTION OF REACTOR GRAPHITE⁺

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ABSTRACT

Distortion of reactor-grade graphites was studied at varying neutron exposures ranging up to 14×10^{21} neutrons per cm^2 (nvt)* at temperatures of irradiation ranging from 425 to 800° C. This exposure level corresponds to approximately 100,000 megawatt days per adjacent ton of fuel (Mwd/At) in a graphite-moderated reactor. A conventional-coke graphite, CSF, and two needle-coke graphites, NC-7 and NC-8, were studied. At all temperatures of irradiation the contraction rate of the samples cut parallel to the extrusion axis increased with increasing neutron exposure. For parallel samples the needle-coke graphites and the CSF graphite contract approximately the same amount. In the transverse direction the rate of contraction at the higher irradiation temperatures appears to be decreasing. Volume contractions derived from the linear contractions are discussed.

INTRODUCTION

Graphite is currently being used as the moderator in a number of production and power reactors. In all these reactors it is used to some degree as a structural component. Radiation-induced dimensional changes of the graphite moderator may therefore distort the reactor core and lead

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*All neutron exposures refer to the number of neutrons with energies greater than 0.18 Mev.

to operational problems. In those reactors using large graphite blocks, differential distortions may also cause rupture of the bars due to stresses or strains exceeding the graphite rupture limit.

The effects of temperature of irradiation on the distortion of nuclear graphites have been previously reported⁽¹⁾ to neutron exposures of 2.3×10^{21} nvt. Since that time considerably more data have been obtained on radiation-induced dimensional changes, and neutron exposures have been extended to a maximum of 14×10^{21} nvt. This exposure is equivalent to lifetime doses in many graphite-moderated reactors. In this paper the effects of these high neutron exposures on graphite distortion will be presented.

BACKGROUND

Different graphite types show considerable differences in distortion behavior as a function of exposure. Fig. 1 presents data on three nuclear-grade graphites irradiated near room temperature. The neutron exposures to the samples have been extended to 11,000 Mwd/At since these data were presented at the last two Conferences on Carbon.^(1,2) The properties and manufacturing parameters of these graphites have been described previously.^(1,3) The expansion observed in the transverse direction is continuing except in the case of the KC graphite. The flattening may be partly due to an increase in temperature in the facility where the irradiations are carried out. The contraction in the parallel direction continues at approximately the same rate.

The rate of contraction of two graphite types at irradiation temperatures from 450 to 1200° C has been reported⁽¹⁾ for exposures to a maximum of 2.3×10^{21} nvt. The contraction rate decreased with increasing temperature and went through a minimum at approximately 800° C. The needle-coke graphites, NC-7 and VC, contracted at a slightly lower rate than the conventional-coke graphite, CSF, in the samples cut transverse to the extrusion axis. There was no significant difference between the two types parallel to the extrusion direction.

EXPERIMENTAL

Irradiation experiments are being conducted in the high-flux testing reactors on the distortion of several types of graphite. Two needle-coke graphites, NC-7 and NC-8, and a conventional-coke graphite, CSF, are being studied in a series of experiments designated as H-3. The NC-7 graphite, with a density of 1.70 g/cm³, is a prototype material for the moderator of the Experimental Gas Cooled Reactor (EGCR), and was manufactured from a Continental-Lake Charles needle-coke. The production-run material for the EGCR moderator, NC-8, was also manufactured from a Continental-Lake Charles needle coke, and has a density of 1.72 g/cm³. The transverse samples of the EGCR graphites are taken from the outer 4 inches of the bar, with the long axis of the sample perpendicular to the edge of the 17 by 17 in. extrusion. Three orientations of the parallel samples are taken, the first at the cross-sectional center of the extrusion, the second half-way between the center and the edge, and the third at the edge

of the bar. These are designated as N1, N2, and N3 respectively for the NC-7 parallel samples, and as P1, P2, and P3 respectively for the NC-8 samples. The transverse samples of the CSF graphite are taken along the cross-sectional center line of the 4 by 4 in. extrusion. The parallel samples are taken along the extrusion axis half-way between the center line and the extrusion edge.

The H-3 experiments are being irradiated in the E-7 position of the General Electric Test Reactor (GETR). A cross-sectional view of the reactor is shown in Fig. 2. The GETR is a light-water-moderated reactor using fully enriched uranium fuel. In the E-7 test position neutron exposures of 2.5×10^{21} nvt are generated in four to five months.

Neutron exposures for these experiments are given in terms of neutrons having energies greater than 0.18 Mev, whereas the exposures for the samples presented in Fig. 1, which were irradiated in a graphite-moderated Hanford reactor, are given in Mwd/At. Conversion between the two units is not constant but varies with the reactor spectrum under consideration. Considerable effort is being expended on methods for measuring neutron fluxes and on a better understanding of the effect of differences in flux spectra on radiation-induced changes in graphite.

The neutron spectrum in the E-7 position of the GETR is shown in Fig. 3. The spectrum, $\phi(u)$, was calculated through the use of the GNU-II computer code modified to include 20 energy groups above 0.18 Mev. GNU-II is a one-dimensional (radial) diffusion code using the continuous slowing down theory.⁽⁴⁾ The dip at lethargy 2.0 is caused by the homogenization of the beryllium located in the same radial increment as E-7. The lethargy, u , is defined as $u = \ln E_0/E$, where E_0 is 10 Mev.

Further refinement of the spectrum is being made through use of the 2DKY code. This is a two-dimensional transport-theory code. At present a factor of 1.5×10^{17} nvt, $E > 0.18$ Mev = 1 Mwd/At is being used for converting neutron exposures generated in the E-7 position to exposures expected in the EGCR. Since use of such conversion factors must take into account the flux spectra, they are specific to particular reactors.

The construction details of a typical H-3 capsule prior to assembly have been described previously.⁽¹⁾ The appearance of a capsule after irradiation in the as-opened condition is shown in Fig. 4. Four individual quarter-round samples, 0.5 in. in radius and 3-7/8 in. long, are assembled into a cylinder and slipped into a graphite sample holder. Six such assemblies are arranged along the length of the capsule. The samples are heated solely by the gamma heating in the reactor. The heat transfer paths from the sample to the aluminum shell are calculated by means of an IBM 7090 computer so that the desired irradiation temperature can be achieved as closely as possible. Sample temperatures are measured by means of nine thermocouples.

As will be noted in Fig. 4 some transport of carbon from the samples to the outer can-wall has been observed, particularly at the lower end of the capsule. X-ray analysis shows the deposit to be carbon, and it is probably a form of carbon suboxide. The mechanism for the transfer is not known at the present time.

The primary length measurements on the samples are made from end-to-end. The appearance of the end of a typical irradiated sample is shown in Fig. 5. The ends of the samples show no evidence of gross damage even after irradiation in several capsules.

HIGH TEMPERATURE CONTRACTION

The results from the samples irradiated in the first five H-3 capsules are presented in Figs. 6 through 9. Neutron exposures are based on an average flux calculated by means of the PDQ computer code.⁽⁶⁾ The PDQ code is a two-dimensional diffusion code using continuous slowing-down theory. The average flux was normalized along the capsule length by the use of flux-monitor foils of nickel, iron, and titanium, irradiated in the first two capsules. Counting of the foils irradiated in the remaining three capsules is in progress.

The contraction data for samples taken transverse and parallel to the extrusion axis of the bar are shown on separate figures to reduce the number of curves per graph and thus minimize confusion. Both orientations for one graphite type should be considered together.

The data for the transverse CSF samples are presented in Fig. 6. The peak neutron flux and peak gamma heating occurred at the capsule mid-position so that the samples with the highest neutron exposure are those irradiated at the highest temperature. Over the temperature range studied, 425 to 800° C, a definite temperature effect on distortion continues to be evident. The samples irradiated at the higher temperatures contract at a lower rate. These differences in rate are attributed to the effect of temperature rather than the effect of flux intensity. It appears that the length change of the transverse samples at 800° C has reached a maximum contraction of 0.85 to 0.90 per cent. Conversely the samples irradiated at 475° C show an increasing rate of contraction.

Fig. 7 shows the results from the CSF graphite samples cut parallel to the extrusion axis. The effect of irradiation temperature is not as pronounced. Samples irradiated at 625 to 800° C all show nearly the same contraction behavior. At all irradiation temperatures the contraction rate is increasing with increasing exposure. At 475° C the parallel samples are contracting about 1-1/2 times and at 800° C 2 to 4 times the transverse samples.

Fig. 8 presents the results for the transverse samples of the EGCR needle-coke graphites, NC-7 and NC-8. The effect of irradiation temperature is again evident with the exception that the samples irradiated at 725 to 800° C show the same contraction. There is a significant difference between the NC-7 and NC-8 materials although it had originally been thought they would be quite similar. The NC-7 contracts less than the CSF while the NC-8 contracts about the same or slightly more than the CSF. Since the NC-7 and NC-8 were made from the same raw materials and by the same processes, there are no apparent differences in the manufacturing methods which would indicate there should be differences in distortion behavior.

It is interesting to note that the 800° C sample expanded significantly during the last irradiation period. The sample shows no outward damage or oxidation that might account for this expansion. Both the physical end-to-end length measurements and the optical hole-to-hole length measurements confirm the results shown. The apparent crystallite size as measured by L_c continued to decrease during the last irradiation period.

Results from the NC-7 and NC-8 samples oriented parallel to the extrusion axis are shown in Fig. 9. For the NC-7 there is no temperature effect from 625 to 800° C but the NC-8 shows some effect. The samples irradiated at 800° C contracted less. The contraction behavior of the NC-7 samples is nearly identical to that of the CSF parallel samples. The NC-8 samples contracted less at 800° C than the CSF and about the same amount at the lower temperatures. At all temperatures the contraction rate increases with increasing exposure.

Although a large effect of neutron-flux intensity is not expected, it is possible that part of the effect attributed to temperature may be due to flux intensity differences. However, data presented previously⁽¹⁾ tend to confirm the presence of a temperature dependence on contraction. The contraction data at 800° C were obtained from samples irradiated in two separate experiments. In one experiment the 800° C data were obtained at the position of maximum flux intensity, and in the other at the position of minimum flux intensity. A factor of 2 between the maximum and minimum flux was measured; however, the contraction rates agreed reasonably well.

Further information on the effect of flux intensity will be obtained from two experiments presently underway. The first is a modification to the H-3 experiment so that the position of maximum flux which was formerly operating at 800° C is now operating at 450° C. Thus data will be obtained at 450° C from samples irradiated in fluxes differing by a factor of three. The second experiment is a re-irradiation of twelve samples at a controlled temperature of 650° C in fluxes varying up to a factor of 3.

VOLUME CONTRACTION

Using the data presented in the last four figures an investigation was made into the combined effect of the transverse and parallel contractions on the change in volume. The model used was a sample of square cross-section with both width and thickness taken as transverse directions and length taken as the parallel direction.

The resultant equation is shown on Fig. 10 with the calculated volume changes. The volume contraction of all three types falls into a narrow band for each irradiation temperature with the greatest divergence at 800° C. The relative contraction among the three types of graphite varies for each temperature but in all cases except at 425° C the needle-coke graphites, NC-7 and NC-8, contract less than the CSF. At 575° C the volume contraction of NC-7 and NC-8 is exactly the same. It appears that NC-8 may be more temperature dependent than the other two types since it contracts at a faster rate at the lower temperatures and at a lower rate at the higher temperatures. The NC-7 volume change at 800° C appears to be starting to saturate. Irradiations of these materials are continuing and as further information is developed these results will be extended to higher exposures.

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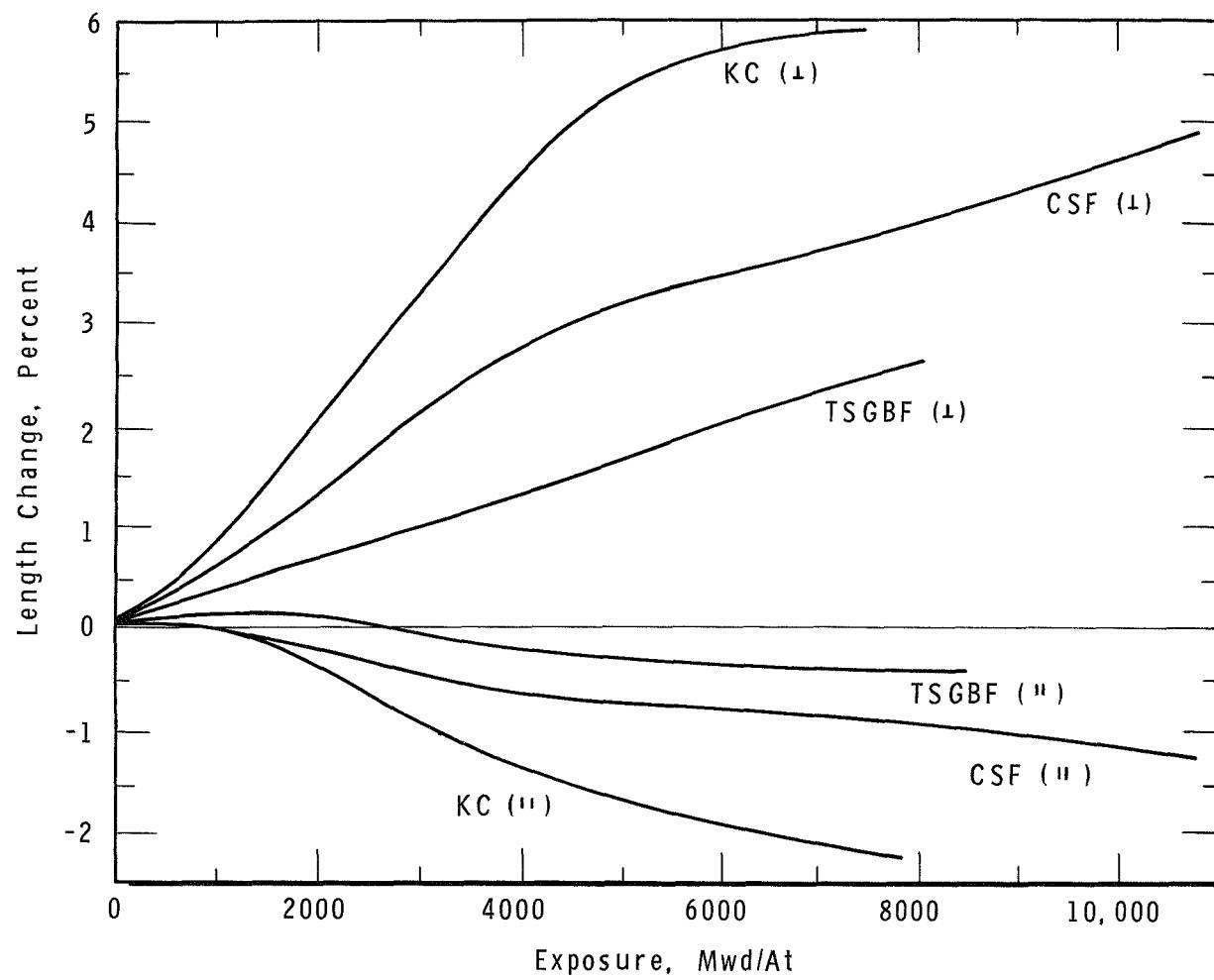


FIGURE 1
GRAPHITE LENGTH CHANGES - 30 C

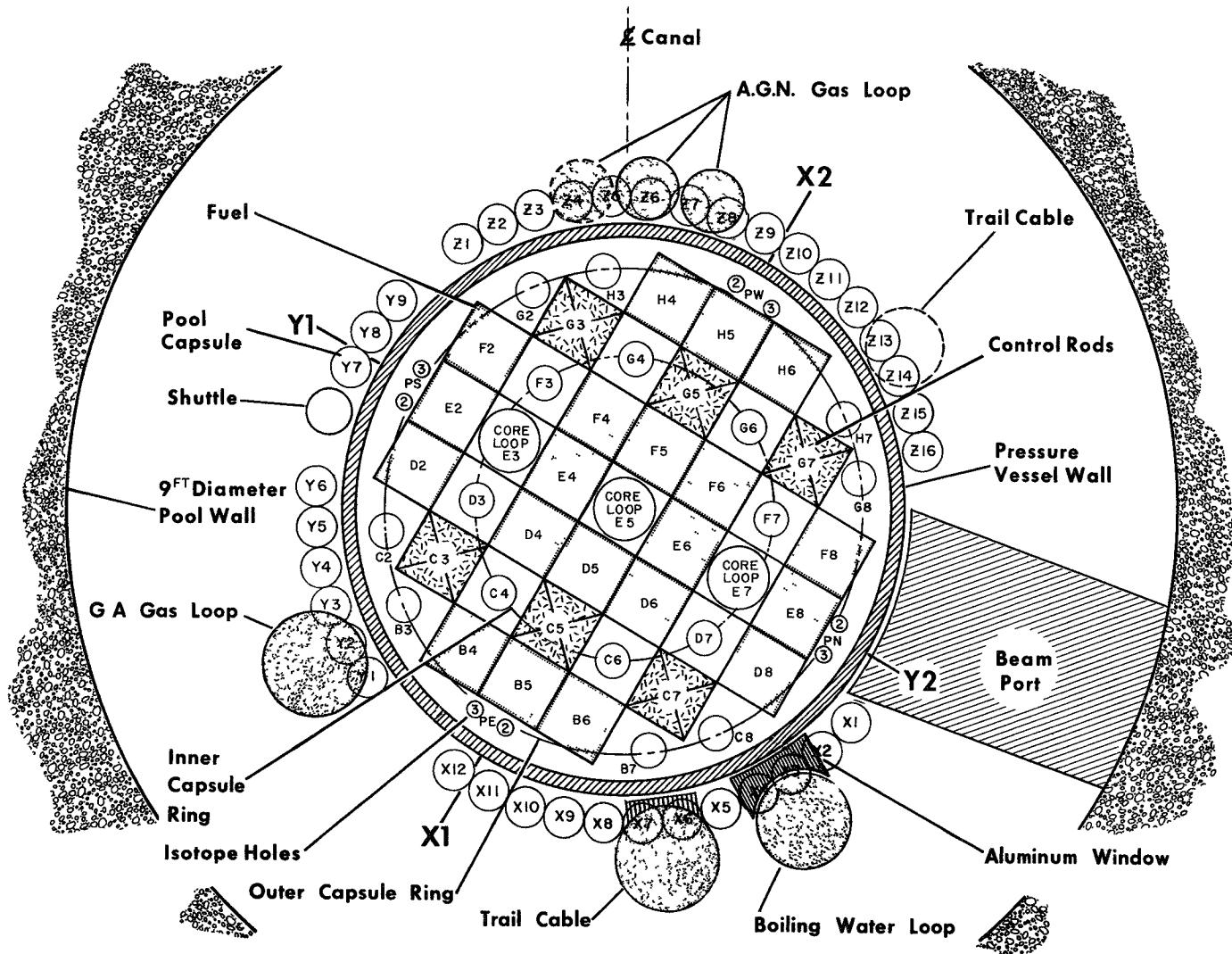


Figure 2. RADIAL LOCATIONS OF GETR EXPERIMENTAL FACILITIES

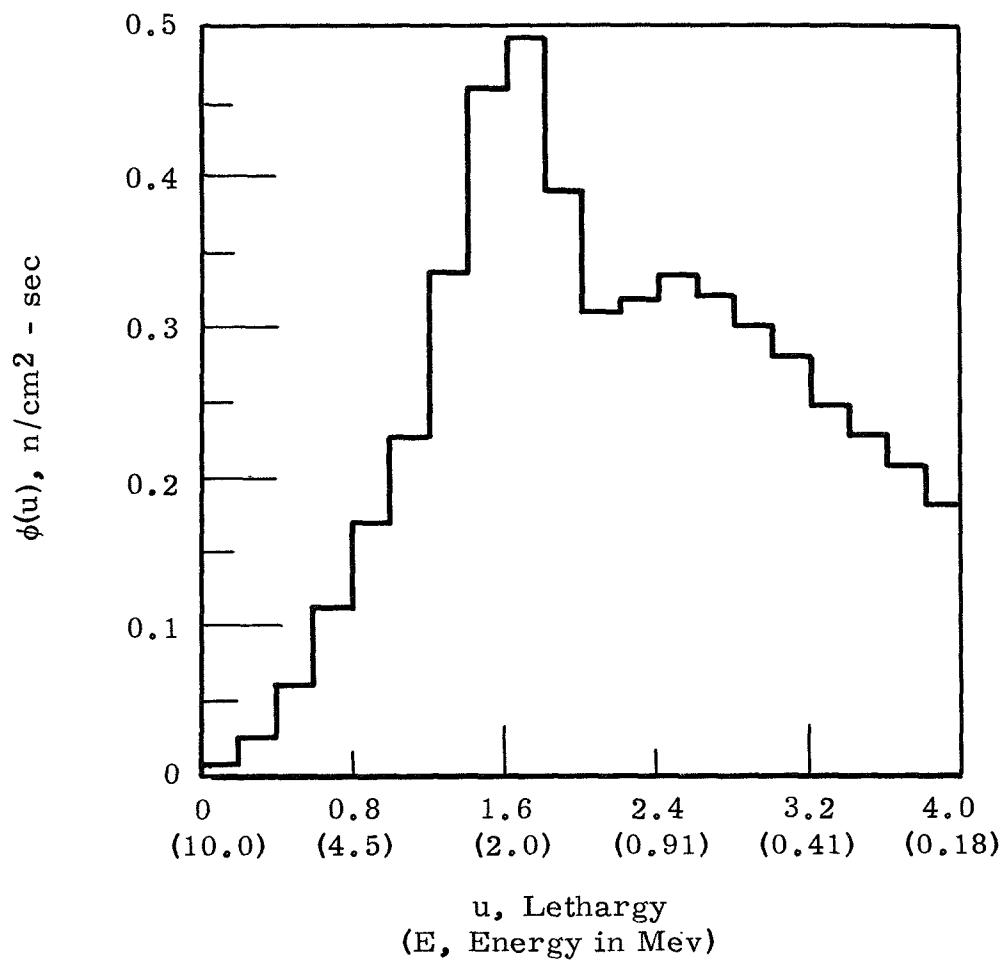


FIGURE 3

NEUTRON SPECTRAL DISTRIBUTION IN E-7
THE INTEGRAL BETWEEN LETHARGY 0 AND
4 HAS BEEN NORMALIZED TO ONE NEUTRON
PER cm^2 PER SECOND

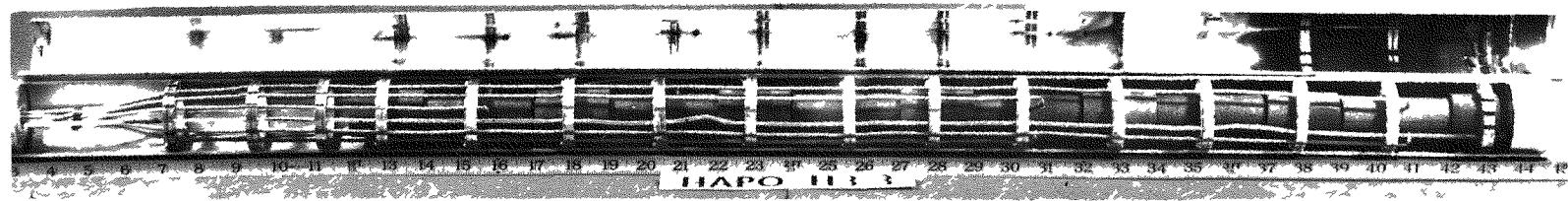


FIGURE 4
POST-IRRADIATION VIEW - GRAPHITE IRRADIATION CAPSULE

51

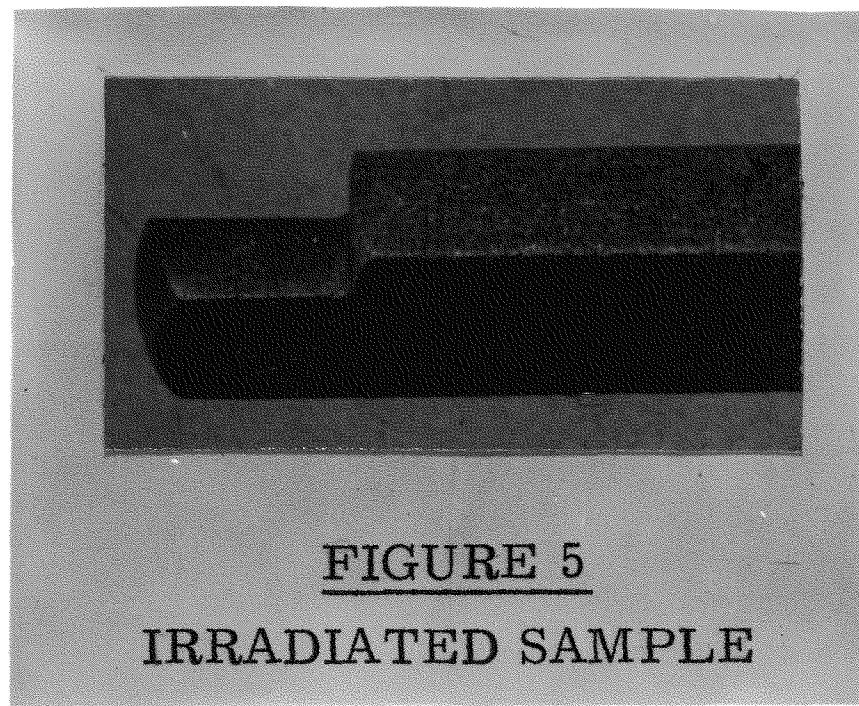


FIGURE 5
IRRADIATED SAMPLE

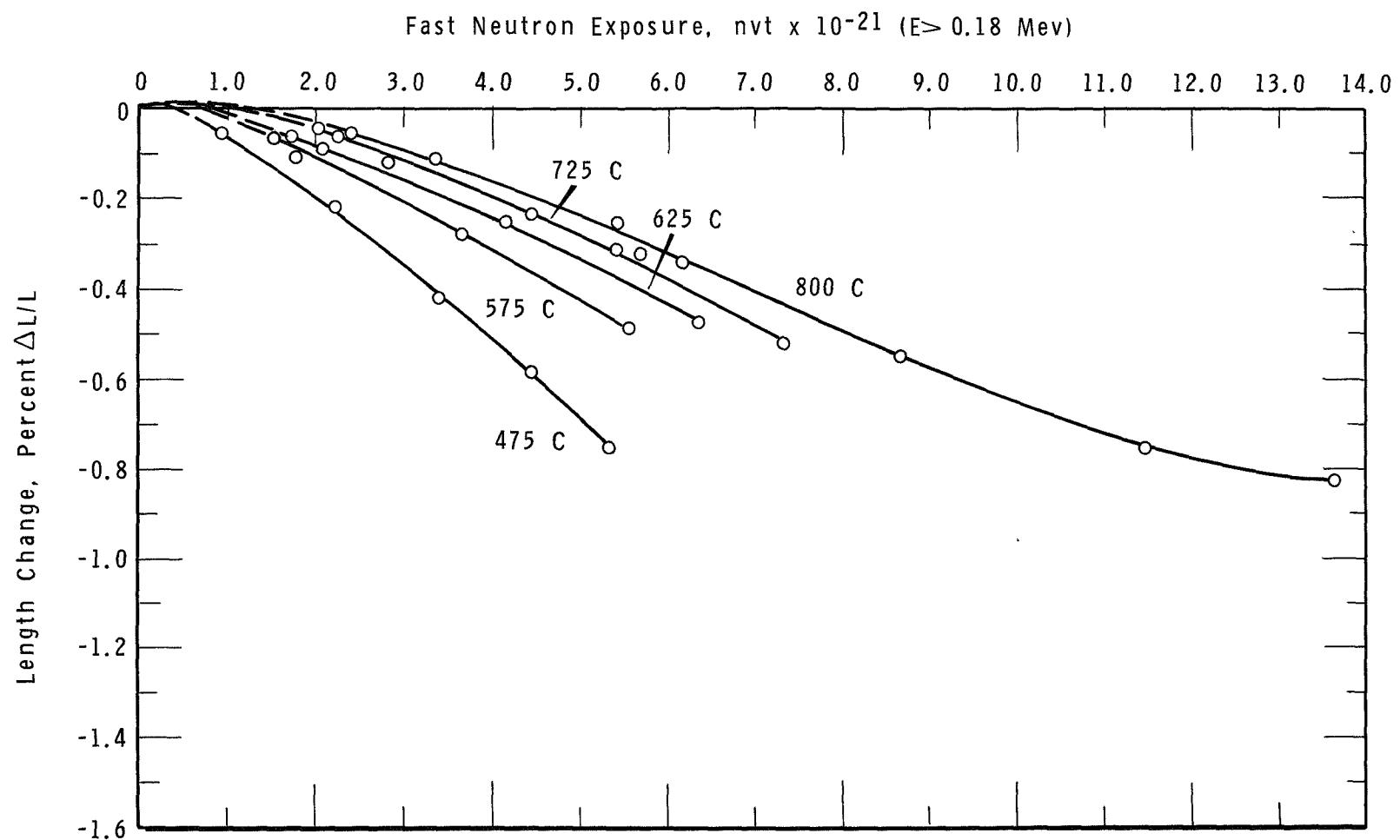


FIGURE 6
CONTRACTION OF CSF GRAPHITE (TRANSVERSE) AS A FUNCTION OF EXPOSURE

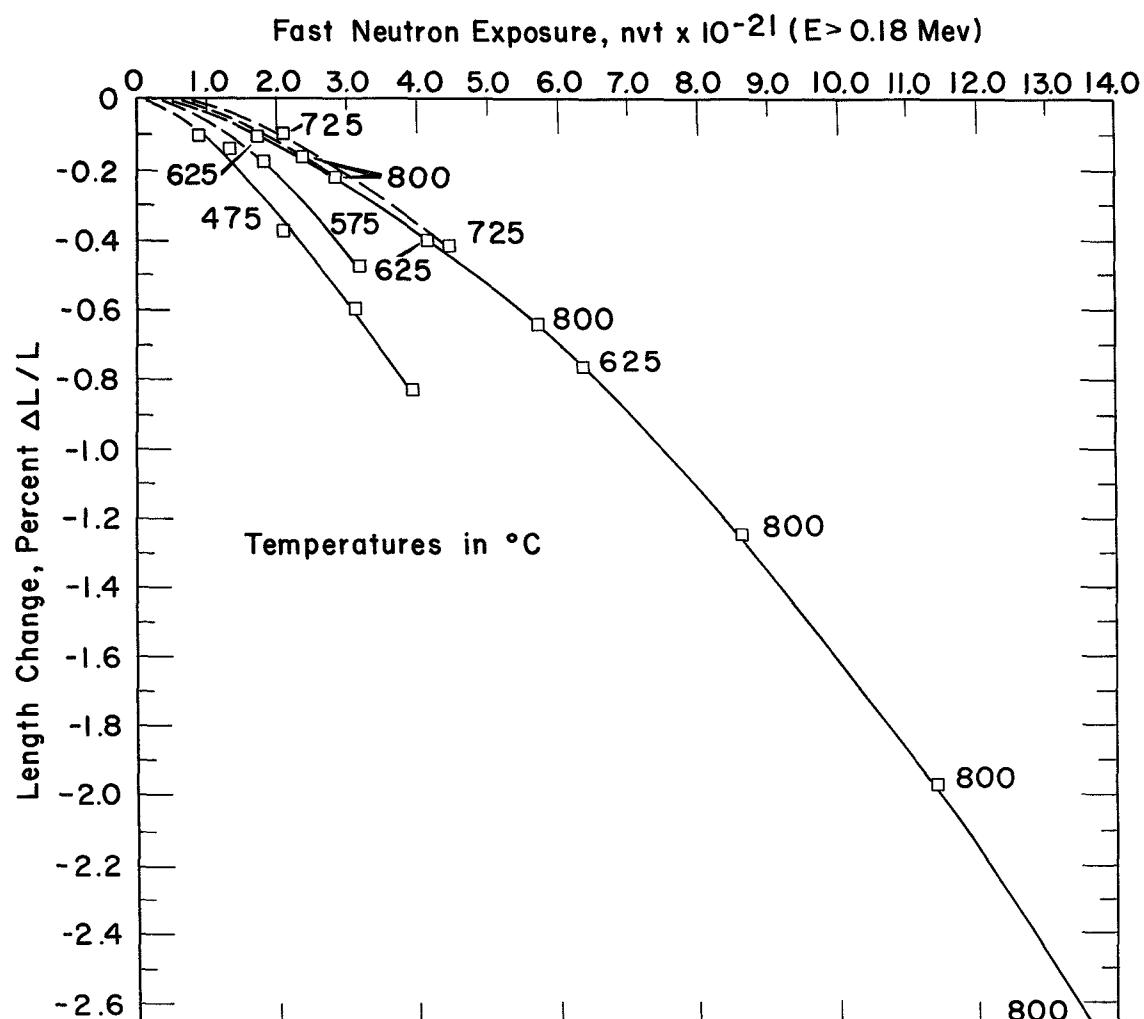


FIGURE 7
CONTRACTION OF CSF GRAPHITE (PARALLEL)
AS A FUNCTION OF EXPOSURE

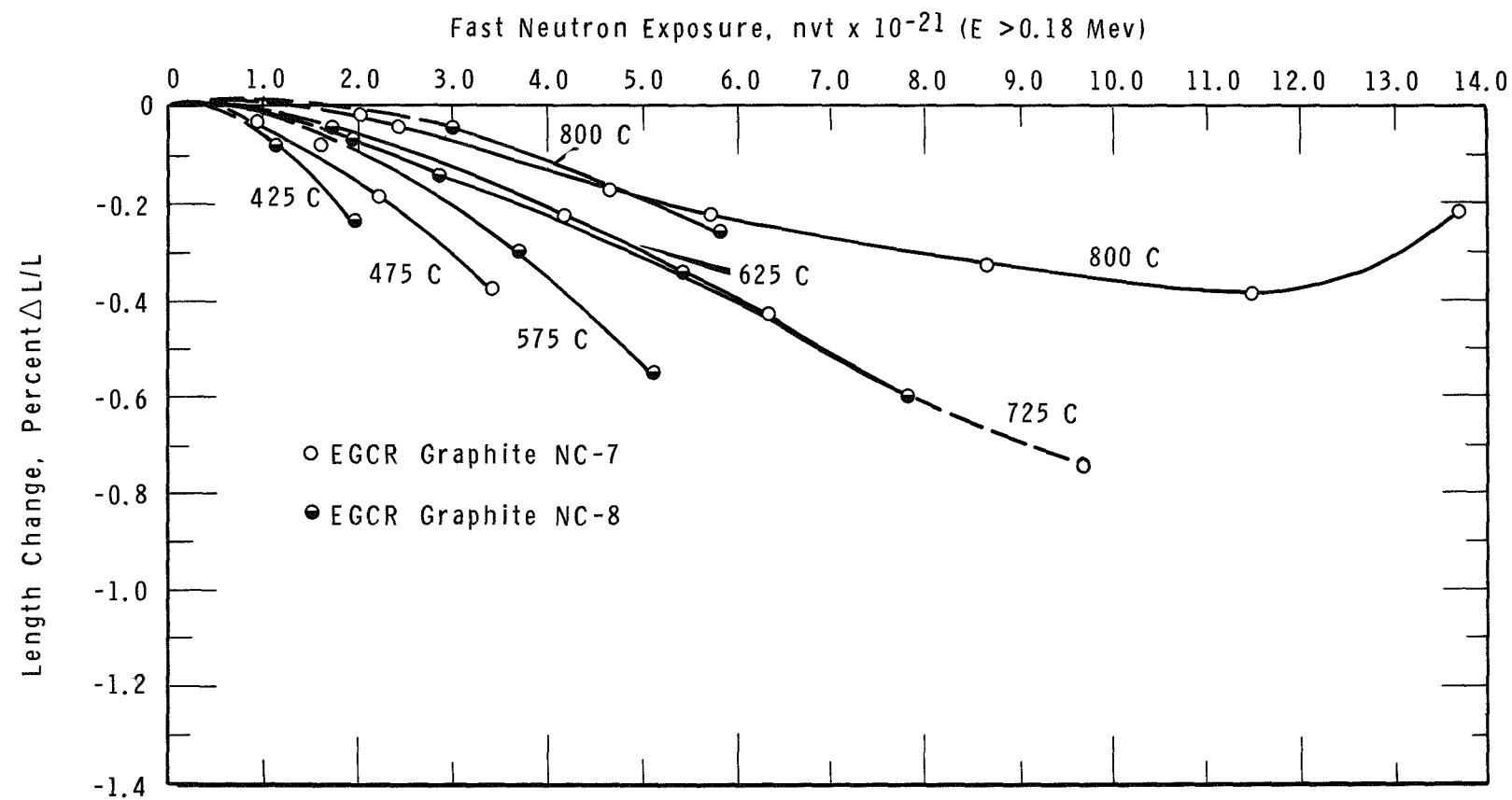


FIGURE 8
CONTRACTION OF EGCR GRAPHITE (TRANSVERSE) AS A FUNCTION OF EXPOSURE

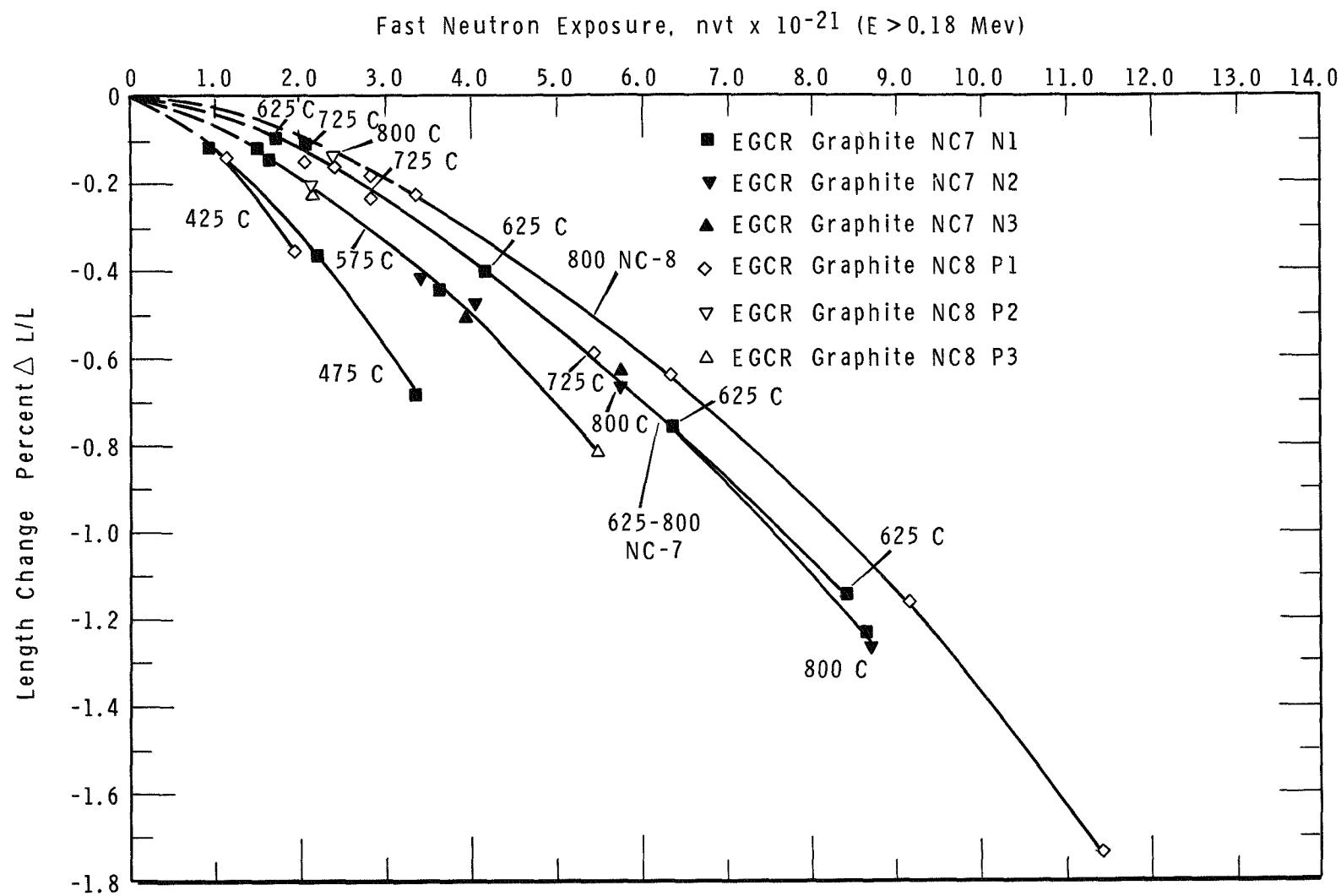


FIGURE 9
CONTRACTION OF EGCR GRAPHITE (PARALLEL) AS A FUNCTION OF EXPOSURE

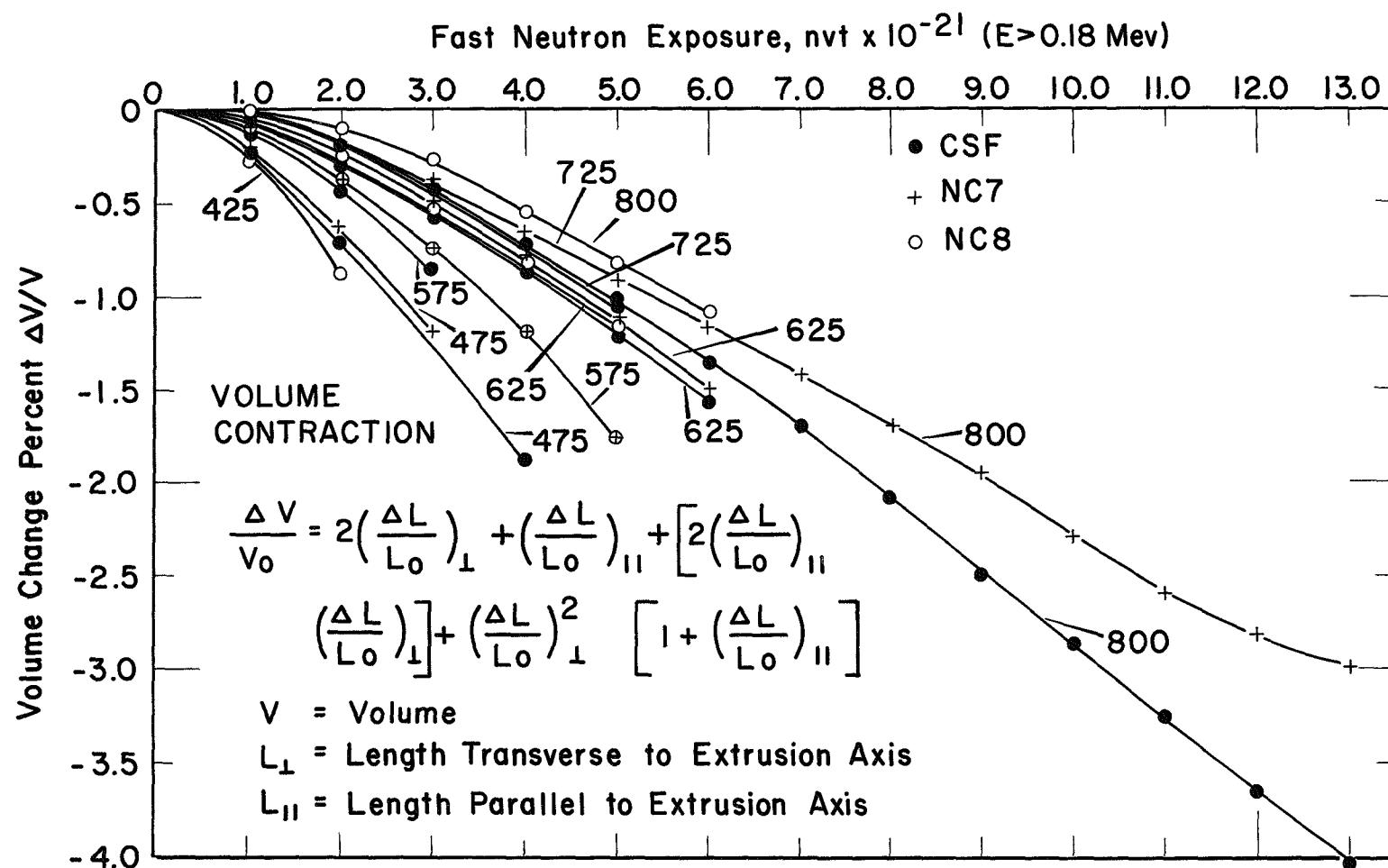


FIGURE 10
 VOLUME CONTRACTION AS A FUNCTION OF EXPOSURE