HEDL-SA-788

CONF-750607--45

HIGH BURNUP INSTRUMENTED IRRADIATION OF BORON CARBIDE (YY02 EXPERIMENT)

A. L. Pitner

INTRODUCTION

Control elements in the Fast Flux Test Facility (FFTF) and Liquid Metal Fast Breeder Reactors (LMFBR's) will employ boron carbide in pellet form as the absorber material. Neutron absorption in boron carbide is accomplished primarily through the ^{10}B (n,α) ^{7}Li reaction; consequently, helium gas is generated during irradiation. Some of this gas is released from the boron carbide structure to the control pin plenum causing gas pressure buildup in the pin. High gas pressures can strain the control pin cladding and potentially even cause rupture, so a knowledge of the gas release behavior from boron carbide during irradiation is necessary for accurate design and safety analyses of LMFBR control elements.

A small fraction (<0.5%) of the neutron absorption reactions in boron carbide also produces tritium through the ^{10}B (n,t) 2α reaction. Although this reaction constitutes a relatively small part of the total neutron capture events, it produces significant quantities of tritium in an LMFBR.(1) Low-exposure data indicate that most of the tritium is retained within the boron carbide.(2) Higher exposure data on tritium retention are required to determine the degree of tritium cleanup equipment required in future LMFBR plants where control elements will experience high burnups.

A third critical performance parameter that requires characterization is irradiation-induced swelling of the boron carbide pellets. This swelling reduces the pellet-clad gap and causes operating temperatures to decrease, which affects subsequent helium release and swelling behavior. Should B_4C swelling close the gap between the pellets and cladding, additional pellet swelling could be transmitted to the cladding causing considerable strain and possible rupture.

A primary objective of the YY02 experiment[4] was to obtain irradiation performance data on boron carbide with regard to the above critical parameters to representative FFTF and LMFBR control rod burnup levels. The experiment was instrumented to continuously monitor irradiation temperatures and gas release from the boron carbide specimens. The test was successful in obtaining boron carbide irradiation data at burnup levels more than twice that previously reported.[5,6]

TEST DESCRIPTION

The experiment is shown schematically in Figure 1. The test contained eighteen boron carbide specimens all located within the core of EBR-II. Each specimen was comprised of four pellets 0.3-inch diameter x 0.3-inch long, doubly encapsulated in stainless steel. Each specimen holder was equipped with a chromel-alumel thermocouple located coaxially within a 1/8-inch diameter pressure tube. Pressure-measuring transducers were located approximately thirty feet above the specimens in the terminal box. Sensor signals were continuously monitored by a data acquisition computer, converted into engineering units, and printed out in hardcopy form daily.

The effects of several material variables were investigated in the experiment. The baseline material was patterned after FFTF reference boron carbide and consisted of enriched (92% ^{10}B) B_4C pellets of 92% TD with a nominal grain size of 15 microns. Variations from this baseline material included enrichment (20, 50, and 92% ^{10}B), pellet density (80, 92, and 99% TD), grain size ($\sim$8, 15, and 28 microns), and stoichiometry ($B_{4.0}C$, $B_{4.3}C$, and $B_{6.5}C$). Although natural boron carbide ($\sim$20% ^{10}B) will be used in FFTF control elements, enriched material was employed in this test to simulate FFTF reaction rates in the lower flux of EBR-II. Specimen temperatures ranged from 1200 to 1600°F and the burnup range was 21 to 82 x 10^{20} captures/cm^3. There are approximately 10^{23} boron atoms per cubic centimeter of boron carbide so the highest exposure represents about 8% burnup of the boron. The complete test matrix is given in Table I.

The experiment was irradiated in the EBR-II for 355 operating days. Instrumentation functioned well during the test with all thermocouples and all but two pressure transducers remaining operative throughout the irradiation. Temperatures, in general, remained within 50°F of the time-averaged values reported in Table I. Out-of-reactor control transducers submitted to similar pressure-history conditions as those in the experiment indicated no change in calibration, thereby testifying to the confidence in measured gas release quantities.

Burnup values were determined by fusion analysis of pellet segments. Measurement of the retained gas in conjunction with the quantity released during irradiation defines the total quantity of helium generated, and therefore, the burnup.

IRRADIATION RESULTS

Pellet Integrity

For the most part, the irradiated pellets were recovered intact with good physical integrity, particularly pellets representative of FFTF reference boron carbide. Figure 2 shows reference pellets irradiated to 81×10^{20} captures/cm^3 at 1345°F, which were recovered in very sound condition. Some of the specimens did show deterioration as a result of irradiation exposure. Low temperature (1175°F) natural B_4C pellets were found to be quite friable; high density (99% TD) pellets exhibited some visible cracking; and $B_{6.5}C$ pellets were fragmented. The remaining specimens, however, retained relatively good integrity throughout the irradiation.

Considerable variation in microstructure was noted during examination of irradiated specimens. Figure 3 shows micrographs of reference material before and after irradiation to 66×10^{20} captures/cm^3 at 1460°F. This irradiated microstructure is representative of that seen in many of the specimens. A significant degree of microcracking that appears to follow grain boundaries was generated during irradiation. However, even with this considerable disruption of microstructure, the pellets remained sound on a macro-scale.

The effect of irradiation on the microstructure of natural B_4C was very temperature-sensitive, as shown in Figure 4. Irradiation at 1175°F caused extensive microcracking and loss of strength as evidenced by the degree of pullout that occurred during sample preparation, even though the burnup of this specimen was only 21 x 10^{20} captures/cm^3. This lack of strength was also apparent on a bulk scale by the tendency of the pellets to crumble when handled. Identical material irradiated at 1280°F (∿100°F hotter), however, showed considerably less microcracking and pullout, as shown in Figure 4C. These pellets also exhibited good structural integrity on a macro-scale.

Material of 50% ^{10}B enrichment maintained the best integrity of all specimens in the test. The microstructure of a pellet irradiated to 50 x 10^{20} captures/cm^3 at 1240°F, as shown in Figure 5, shows only a slight degree of microcracking, nearly undetectable at 100X magnification. Another 50% ^{10}B specimen irradiated to the same burnup at 1405°F exhibited a very similar microstructure. The bulk pellets were also in excellent physical condition. These pellets varied from other specimens in that they had a relatively small grain size (∿10 microns) and received an additional 5-hour heat treatment at 2200°C following hot-pressing. This additional heat treatment may have enhanced the pellet structure and increased resistance to in-reactor degradation. It should be remembered, however, that because of the lower enrichment level, these specimens experienced less burnup than fully enriched (92% ^{10}B) specimens.

In general, microcracking in irradiated boron carbide specimens appeared to decrease in severity with increasing temperature from ∿1200 to 1450°F, and then worsen again at higher irradiation temperatures. This microcracking behavior, in general, is qualitatively consistent with helium retention behavior in this temperature range.

Helium Release

Typical helium release behavior measured in the test is shown in Figure 6. The solid line represents volumetric gas release, while the broken line corresponds to fractional release based on the total quantity of helium generated at any specific burnup. These results are for $B_{4.0}C$ pellets of 92% ^{10}B and 92% TD with a nominal grain size of 15 microns irradiated at 1460°F, but are generally representative of helium release behavior observed in most of the specimens. Following an initial high release rate, helium release decreases significantly (after a burnup of ∿8 x 10^{20} captures/cm^3), remains relatively constant (to ∿30 x 10^{20} captures/cm^3), and then begins increasing again slowly at high burnups. The decrease and subsequent increase in release rate are attributed to the development and saturation of helium trapping sites in the boron carbide lattice.

Helium release behavior of the four reference B_4C specimens irradiated at four separate temperatures is shown in Figure 7. It is seen that helium release increases with temperature from 1285°F to 1460°F, but is less at 1510°F than at 1460°F. This is consistent with results obtained in earlier tests.(5) The numbers in parentheses at the end of each curve in Figure 7 represent fractional release values at the end of the irradiation. Intermediate fractional release quantities can be determined based on the helium generation rate of 3.72 cm^3 (STP) per 10^{20} captures/cm^3.

Figure 8 shows the effect of ^{10}B enrichment (or the effect of reaction rate) on helium release. A direct comparison at 1400°F was not possible since no 20% or 92% specimens were irradiated at this temperature. The 92% ^{10}B curve shown for 1400°F was interpolated from the curves shown in Figure 7. No significant variation in helium release behavior is noted for the three enrichment levels investigated in this test. Another natural B_4C specimen irradiated at 1175°F released only 9 cm^3 (STP) He/cm^3 B_4C at 21 x 10^{20} captures/cm^3.

Helium release behavior of boron carbide pellets of 80, 92, and 99% TD is shown in Figure 9 for irradiation temperatures of ∿1350 and 1510°F. Pellet density appears to have essentially no effect on helium release behavior under the irradiation conditions of this test.

The effect of grain size on helium release is shown in Figure 10. At 1350°F, specimens with nominal grain sizes of 15 and 28 microns behave nearly identically. Above 1500°F, however, helium release appears to vary inversely with grain size. Pellets with a nominal grain size of 8 microns release twice as much helium as those with a nominal grain size of 28 microns at

1520°F. On an absolute scale, however, the difference in behavior is not large: fractional release values for the 28 micron and 8 micron specimens are 9% and 19%, respectively.

The effect of stoichiometry on helium release behavior is quite dramatic, as shown in Figure 11. Helium is released from $B_{6.5}C$ pellets in a burst-like manner at low exposures, and far exceeds the release from $B_{4.0}C$ specimens at higher burnups. At a burnup of 65 x 10^{20} captures/cm^3 and a temperature of ~1490°F, helium release from the $B_{6.5}C$ specimen is nearly five times that from the $B_{4.0}C$ specimen. Helium release from the $B_{4.3}C$ specimen is intermediate to that from the $B_{4.0}C$ and $B_{6.5}C$ specimens. Another $B_{6.5}C$ specimen irradiated at 1410°F released 165 cm^3 (STP) He/cm^3 B_4C at 81 x 10^{20} captures/cm^3. Release fractions for the $B_{6.5}C$ specimens at the end of the irradiation were 54% and 72% at 1410°F and 1495°F, respectively.

Tritium Retention

Results of tritium retention measurements are given in Table II. The fractional retention values quoted in the table are based on measured tritium content and estimated tritium production rates. There is considerable uncertainty in each of these quantities, and the combined uncertainty in the fractional retention value is estimated as ±40% (relative) at the 95% confidence level. Because of the large uncertainty, definite trends are difficult to identify. A general observation, however, appears to be decreasing retention with increasing irradiation temperature. This is noticeable in the reference specimens and large-grained (28 μ) specimens where direct temperature comparisons can be made. It is also noted that for FFTF reference type boron carbide, more than half the tritium generated is typically retained within the pellet.

Swelling

The swelling of FFTF reference boron carbide is shown in Figure 12 as a function of burnup for temperatures of 1250-1550°F. Swelling results obtained in previous lower-exposure tests(5) are included in the figure as the open points, and YY02 swelling data are represented by the closed points. It is apparent that swelling increases with increasing exposure. There is some scatter in the 1250-1350°F data, but the higher temperature data are quite consistent and agree well with the lower-exposure results from previous tests. Swelling at 1350-1450°F is notably less than at the other temperatures considered here. In general, diametral pellet growth is less than 4% at 80 x 10^{20} captures/cm^3 for FFTF reference boron carbide at 1250-1550°F.

An interesting observation noted in the YY02 test results was the inverse relationship between pellet swelling and helium release, or alternatively, the direct dependency between pellet swelling and helium retention. This behavior is shown in Figure 13, which includes swelling data from all crack-free boron carbide specimens in the test at a burnup of ~75 x 10^{20} captures/cm^3. The data are quite consistent when viewed in this respect. Thus, it appears that irradiation-induced swelling in boron carbide pellets is at least partially due to helium production and retention.

SUMMARY AND CONCLUSIONS

Boron carbide pellets were irradiated in the EBR-II at temperatures of 1200-1600°F to burnups as high as 80 x 10^{20} captures/cm^3. Temperature and gas release from each specimen were continuously monitored during the irradiation. In general, helium release was observed to be high during initial irradiation, then decreased after a burnup of ~8 x 10^{20} captures/cm^3. The release rate remained relatively low until burnup exceeded ~30 x 10^{20} captures/cm^3, then began increasing slowly. Helium release fractions at the end of the test were generally small, 10-20% for most of the specimens. Material variables of ^{10}B enrichment, pellet density, and grain size had little effect on helium release. $B_{6.5}C$ specimens, however, showed quite high helium release: 54% and 72% at 1410°F and 1495°F, respectively.

Pellet integrity of most specimens was good after the irradiation. $B_{6.5}C$ specimens were fragmented, high density (99% TD) pellets were cracked, and natural B_4C pellets irradiated at low temperature (1175°F) were friable, but the remainder of the eighteen test specimens were basically sound.

Microstructure degradation appeared to decrease with increasing temperature from 1175°F to 1450°F, but then worsened again at higher temperatures. Bulk pellets retained good physical strength even when significant microcracking was observed in the microstructure. Fine-grained pellets that received additional heat treatment following hot-pressing exhibited the best structural integrity after irradiation.

Measurement of tritium retention in boron carbide indicated decreasing retention with increasing irradiation temperature. For FFTF reference boron carbide, tritium retention generally exceeded 50% of the calculated production quantity.

Diametral swelling of boron carbide pellets increased with increasing irradiation exposure. Measured swelling of FFTF reference B_4C specimens agreed with trends indicated by previous lower-exposure test results, and was typically less than 4% (diametral) at 80 x 10^{20} captures/cm^3. In general, microstructural degradation and bulk pellet swelling were greatest for those specimens that retained the greatest fraction of helium.

This test demonstrated the capability of FFTF reference boron carbide to perform well to burnups of 80 x 10^{20} captures/cm^3, well beyond the maximum burnup level of $\sim$50 x 10^{20} captures/cm^3 anticipated for first core control elements.

References

1. T. J. Kabele, "Parametric Study of Tritium Distribution in FFTF," HEDL-TME-74-6 (1974).
2. Neutron Absorber Technology Staff, "A Compilation of Boron Carbide Design Support Data for LMFBR Control Elements," HEDL-TME-75-19 (1975).

4. A. L. Pitner, "The YY02 Instrumented Irradiation of LMFBR Absorber Material," Trans. Am. Nucl. Soc., 17, 188 (1973).
5. J. A. Basmajian et. al., "Irradiation Effects in Boron Carbide Pellets Irradiated in Fast Neutron Spectra," Nucl. Technol., 16, 238 (1972).
6. R. E. Dahl and J. W. Bennett, "IWGFR Specialists Meeting on Absorbing Materials and Control Rods for Fast Reactors -- Summary Report," HEDL-TME-73-91 (1973).

TABLE I

EXPERIMENT TEST MATRIX

Specimen Type	% ^{10}B	% T.D.	$\overline{G.S.}$ (μ)	B:C	Burnup (10^{20} capt/cm^3)	Temperature (°F)
Reference	92	92	15	4.0	81	1285
	92	92	15	4.0	81	1345
	92	92	15	4.0	66	1460
	92	92	15	4.0	66	1510
Pellet Density	92	80	13	4.0	71	1355
	92	80	13	4.0	71	1505
	92	99	12	4.0	82	1360
	92	99	12	4.0	82	1570
^{10}B Enrichment	20	92	12	4.0	21	1175
	20	92	12	4.0	21	1280
	50	92	10	4.0	50	1240
	50	92	10	4.0	50	1405
Grain Size	92	92	8	4.0	72	1525
	92	92	28	4.0	72	1360
	92	92	28	4.0	72	1530
Stoichiometry	92	92	15	4.3	65	1455
	92	92	14	6.5	81	1410
	92	92	14	6.5	67	1495

TABLE II

TRITIUM RETENTION IN BORON CARBIDE

Specimen Type	Burnup (10^{20} capt/cm^3)	Temperature (°F)	Tritium Retention (%)
Reference	81	1285	91
"	80	1345	66
"	80	1345	46
"	66	1510	57
"	66	1510	47
80% TD	71	1505	17
99% TD	82	1360	65
Natural B_4C	21	1280	129
50% ^{10}B	50	1405	96
8μ G.S.	72	1525	38
28μ G.S.	72	1360	69
"	72	1530	39
$B_{4.3}C$	65	1455	37
$B_{6.5}C$	81	1410	51

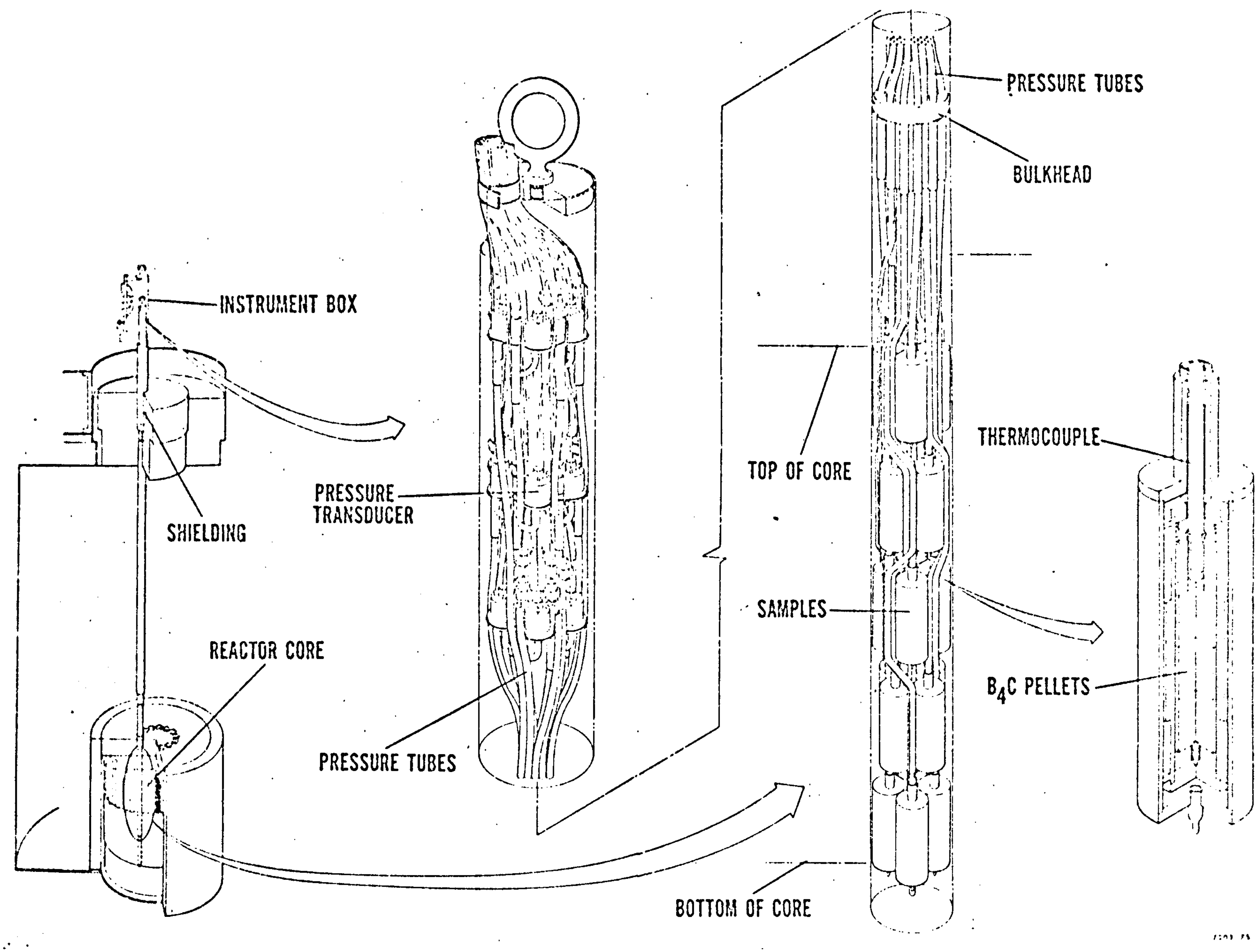


Figure 1. Schematic of YY02 Experiment

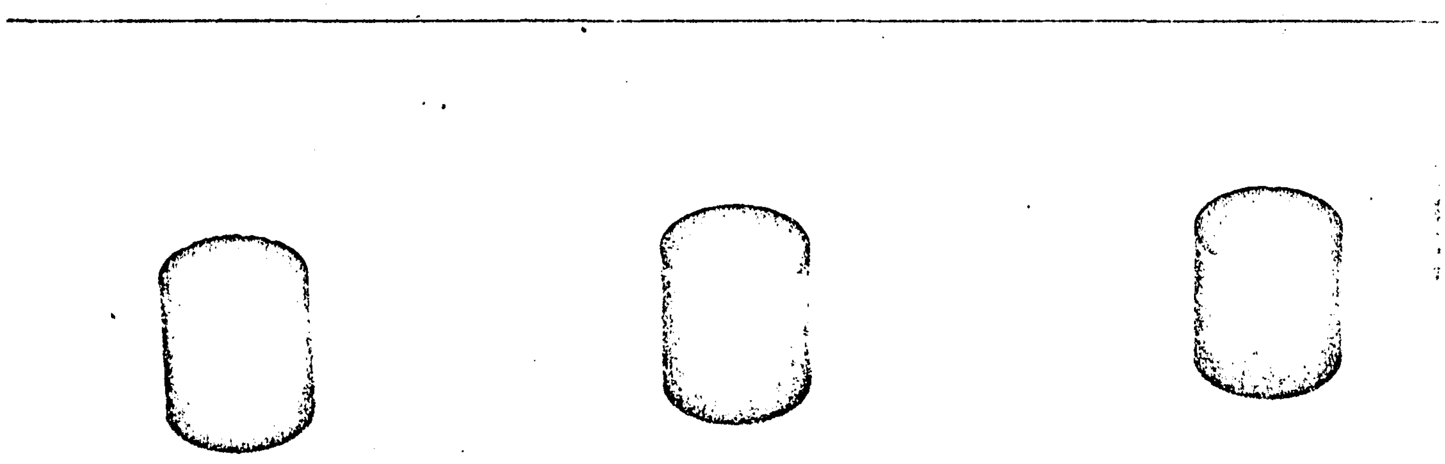
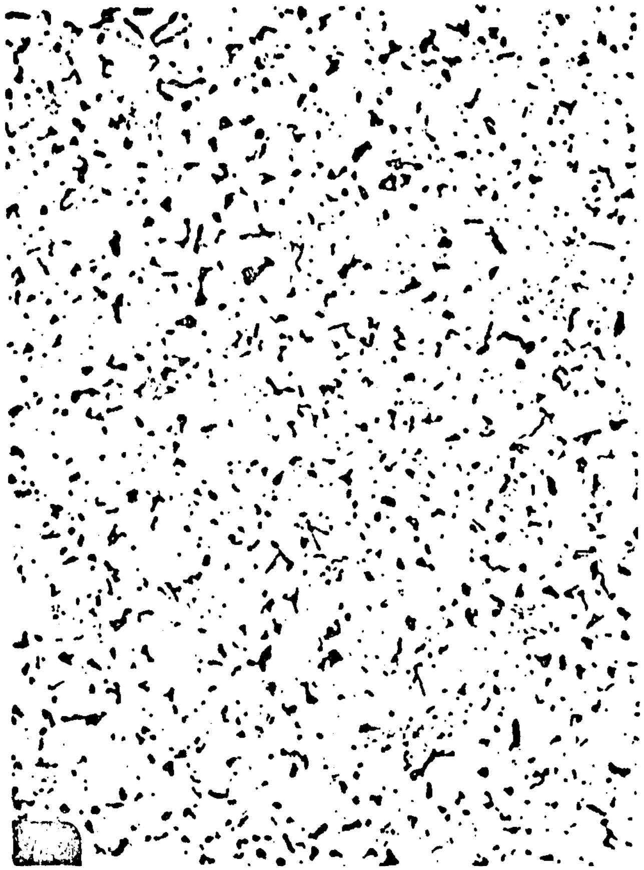
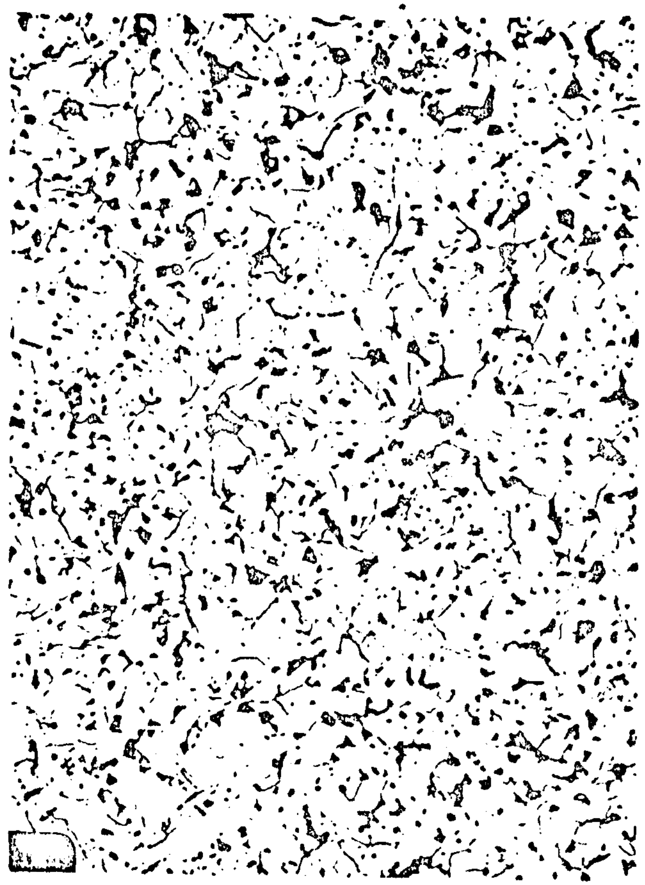


Figure 2. FFTF Reference B_4C Pellets Irradiated to 81×10^{20} Captures/cm^3 at 1345°F

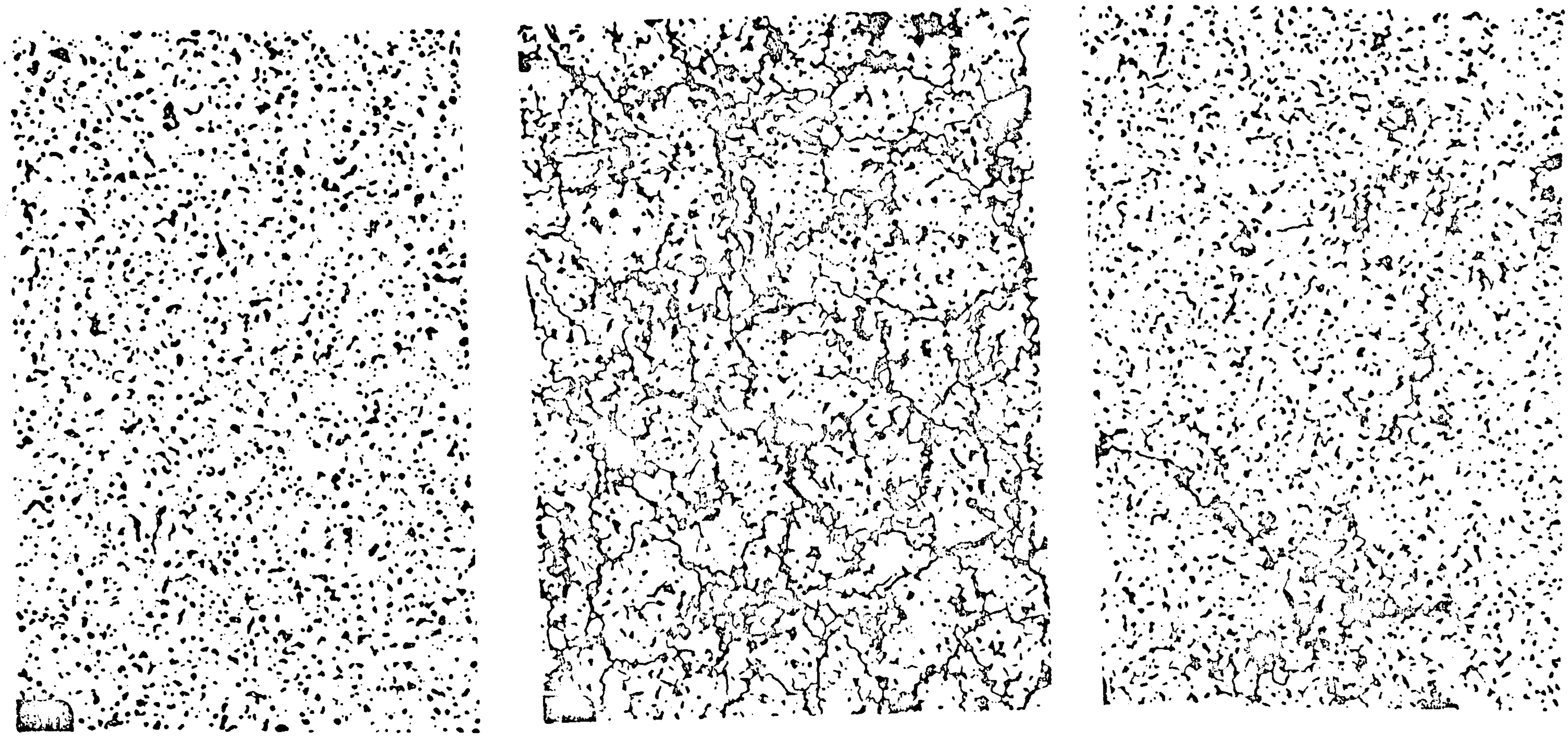


(a)



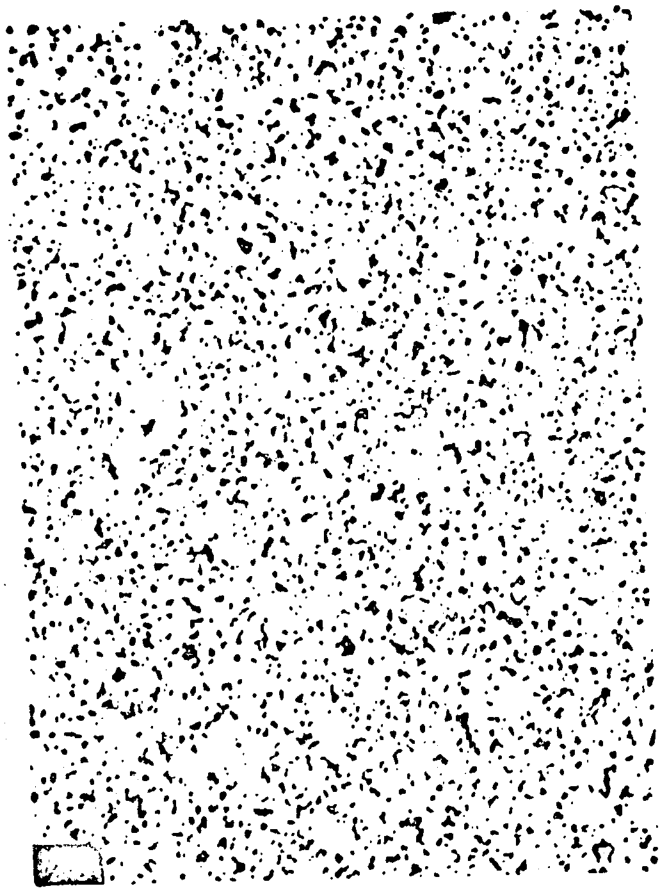
(b)

Figure 3. Microstructure of Reference B_4C (100X)
(a) Unirradicted (b) Irradiated to
66×10^{20} Captures/cm^3 at 1460 °F

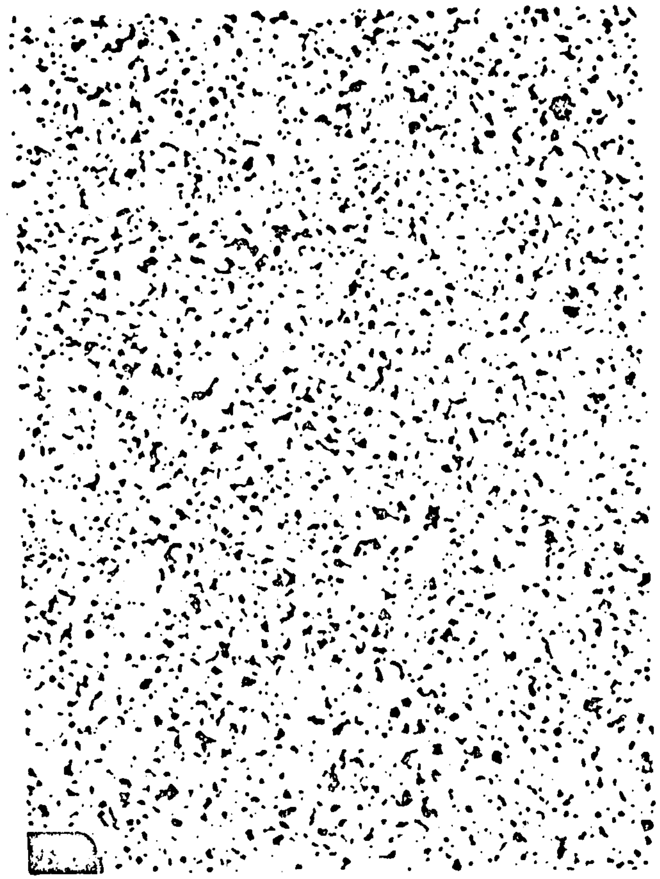


(a) (b) (c)

Figure 4. Microstructure of Natural B_4C (100X)
(a) Unirradiated (b) Irradiated to 21×10^{20} Captures/cm^3 at 1175°F (c) Irradiated to 21×10^{20} Captures/cm^3 at 1280°F



(a)



(b)

Figure 5. Microstructure of 50% ^{10}B Boron Carbide (100X)
(a) Unirradiated (b) Irradiated to 50×10^{20} Captures/cm^3 at 1240°F

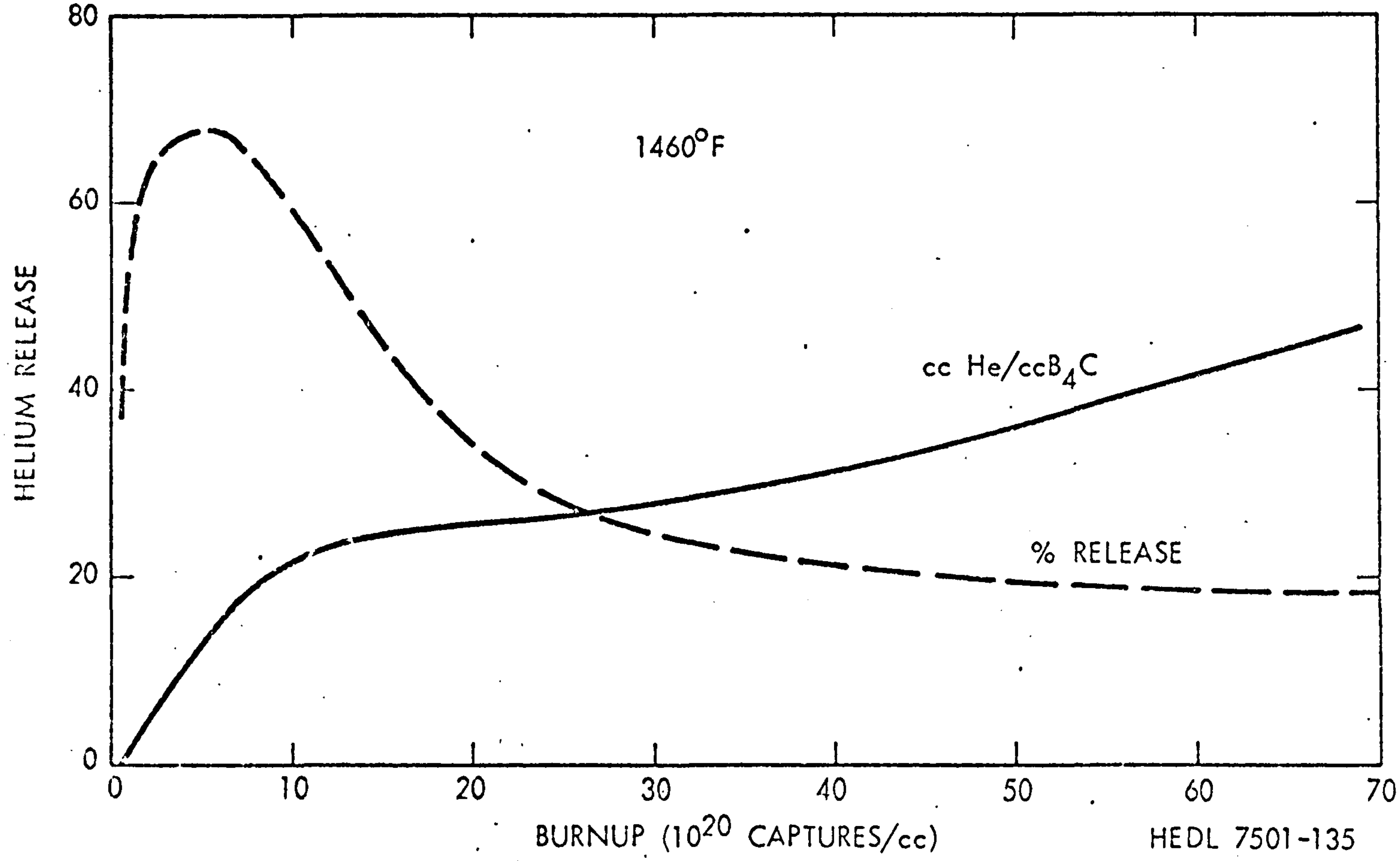


Figure 6. Typical Helium Release Behavior Observed in Test

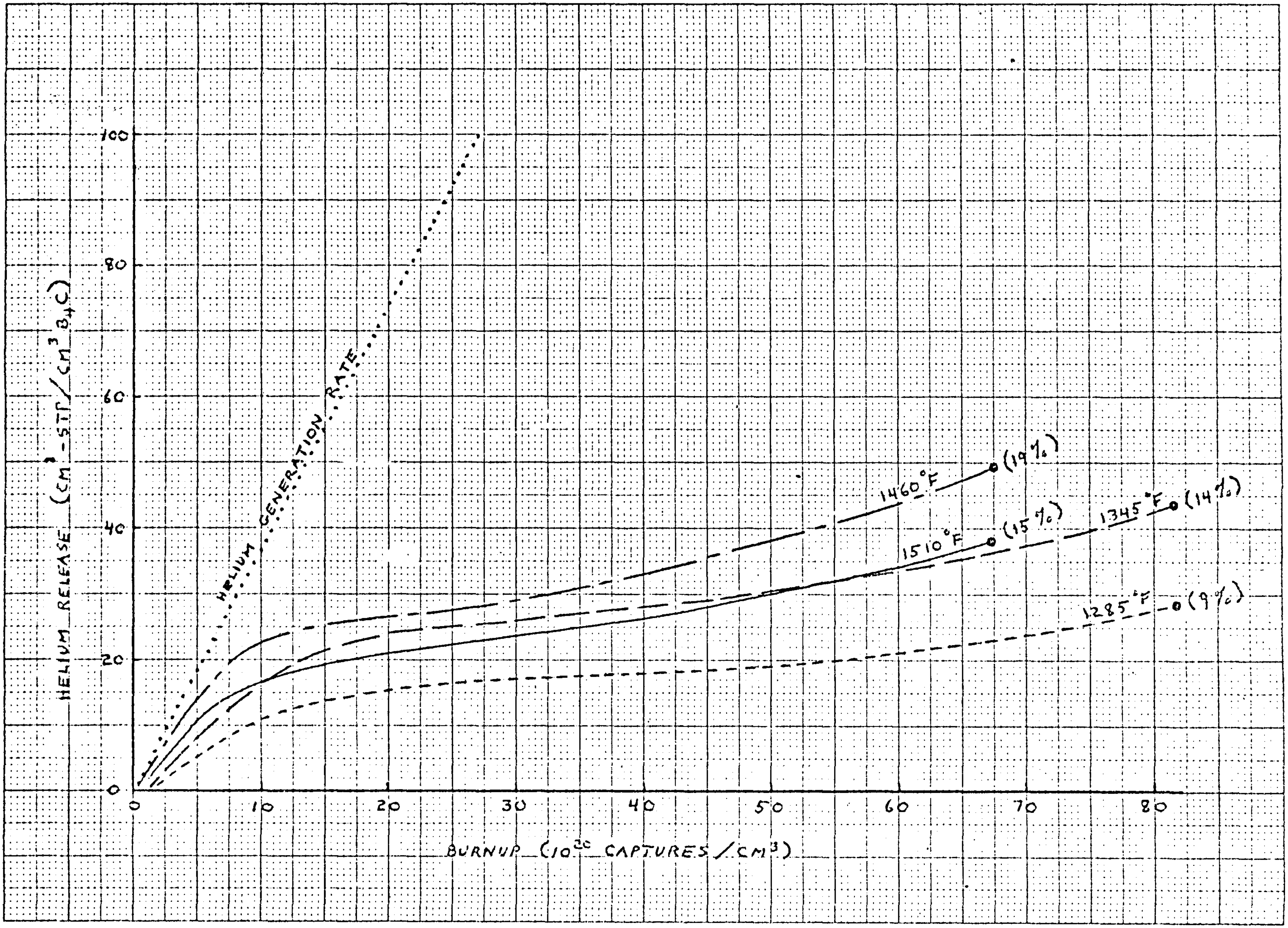


Figure 7. Helium Release From Reference B_4C Specimens

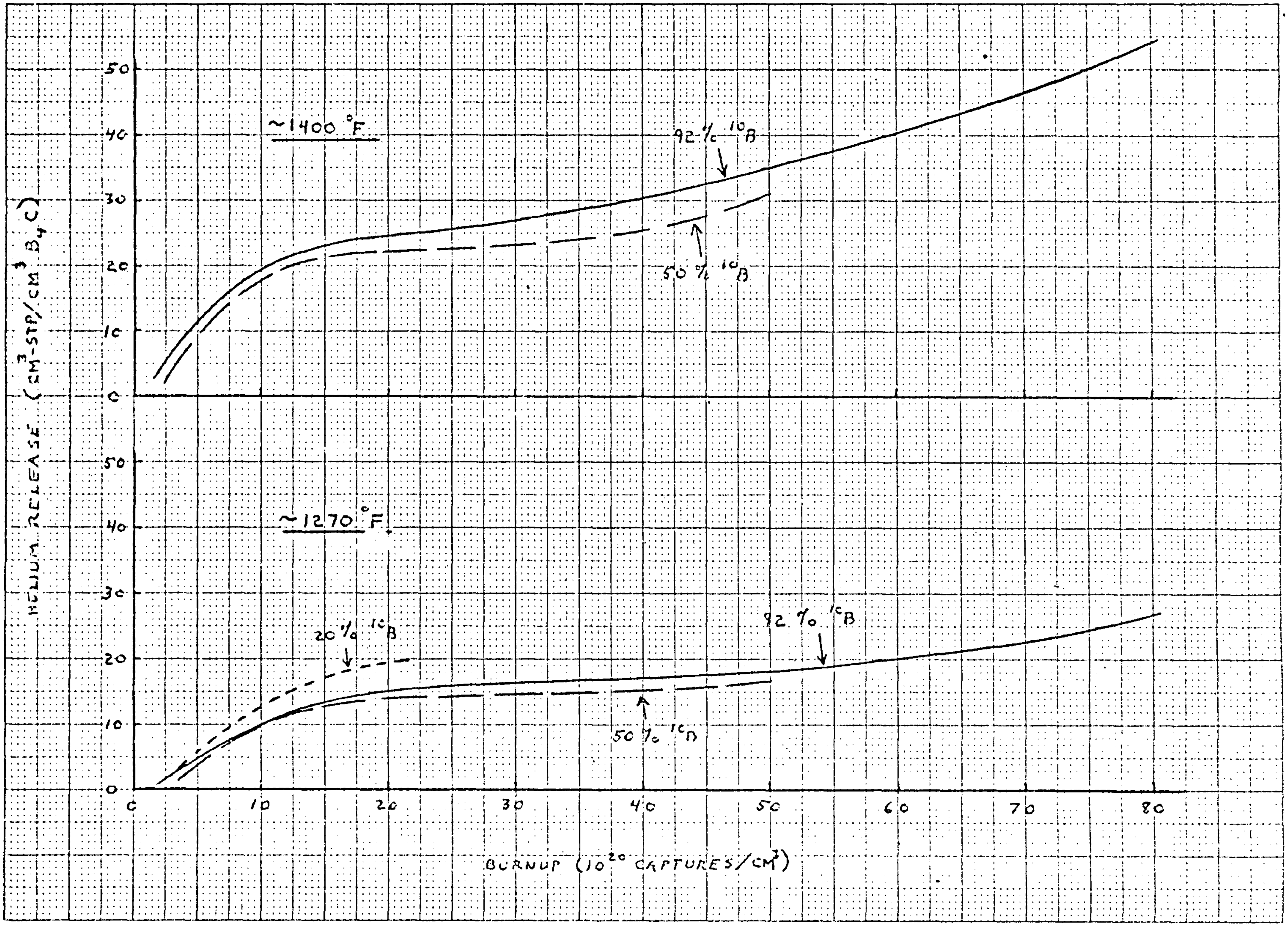


Figure 8. Effect of ¹⁰B Enrichment on Helium Release

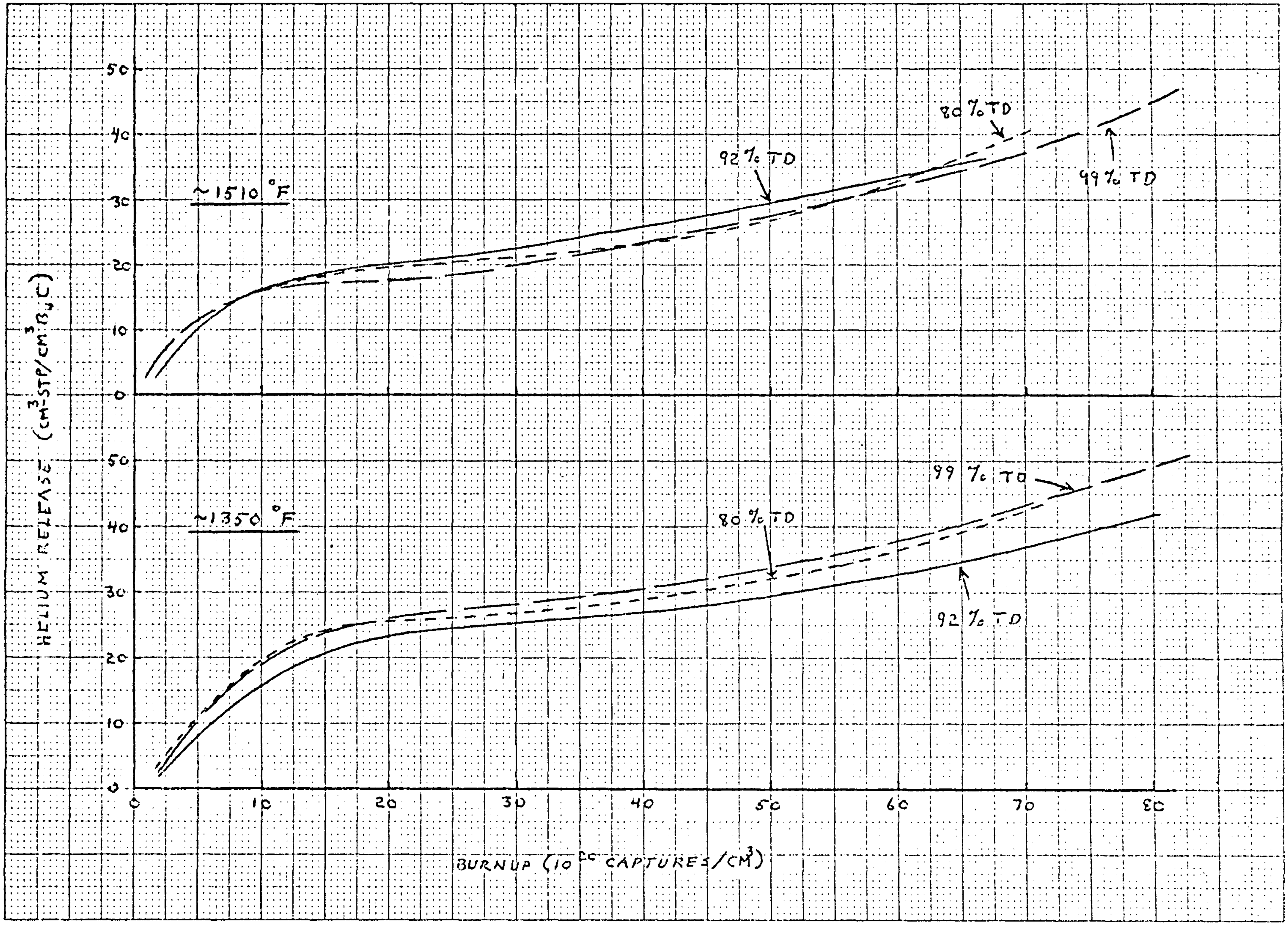


Figure 9. Effect of Pellet Density on Helium Release

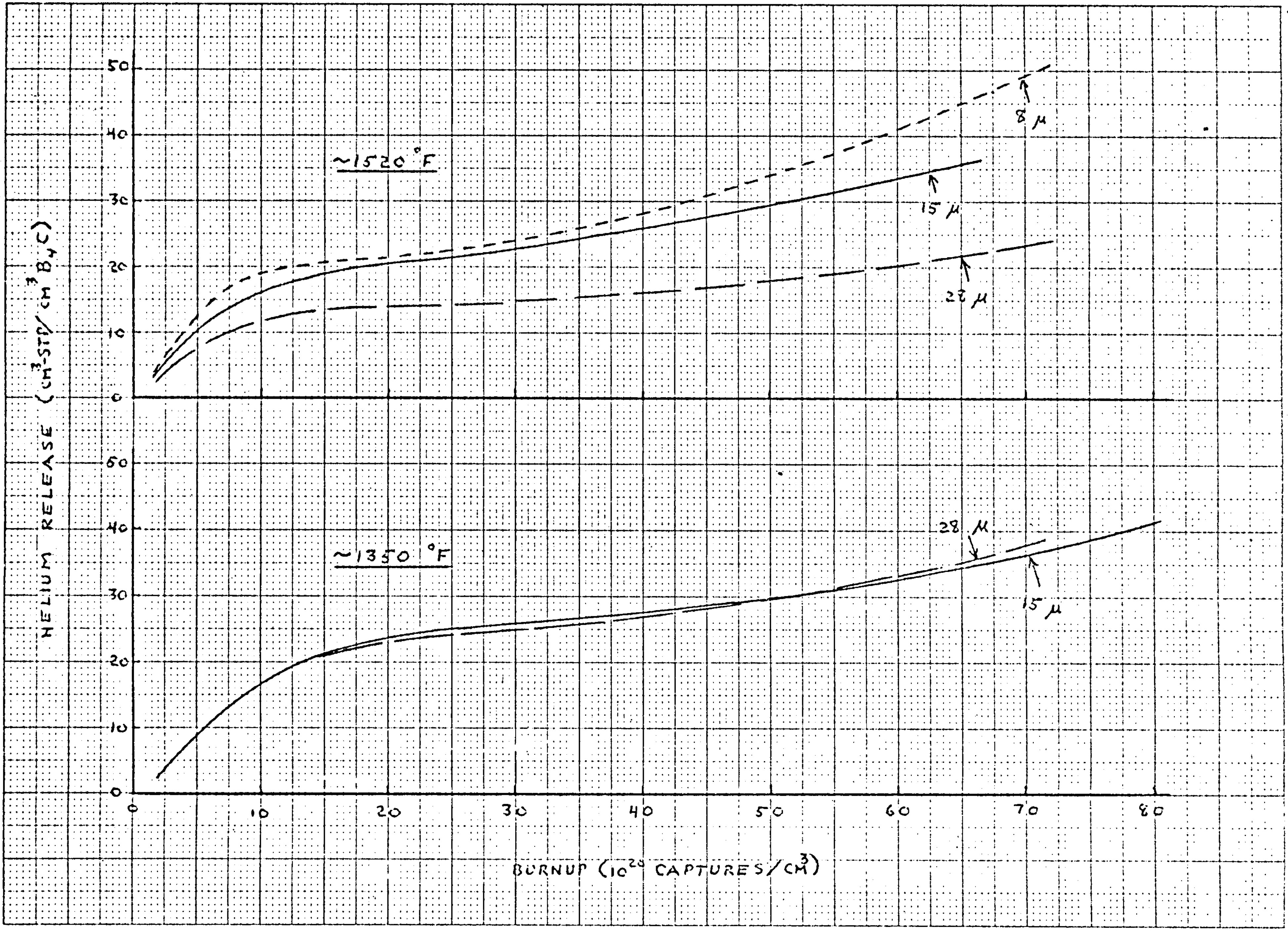


Figure 10. Effect of Grain Size on Helium Release.

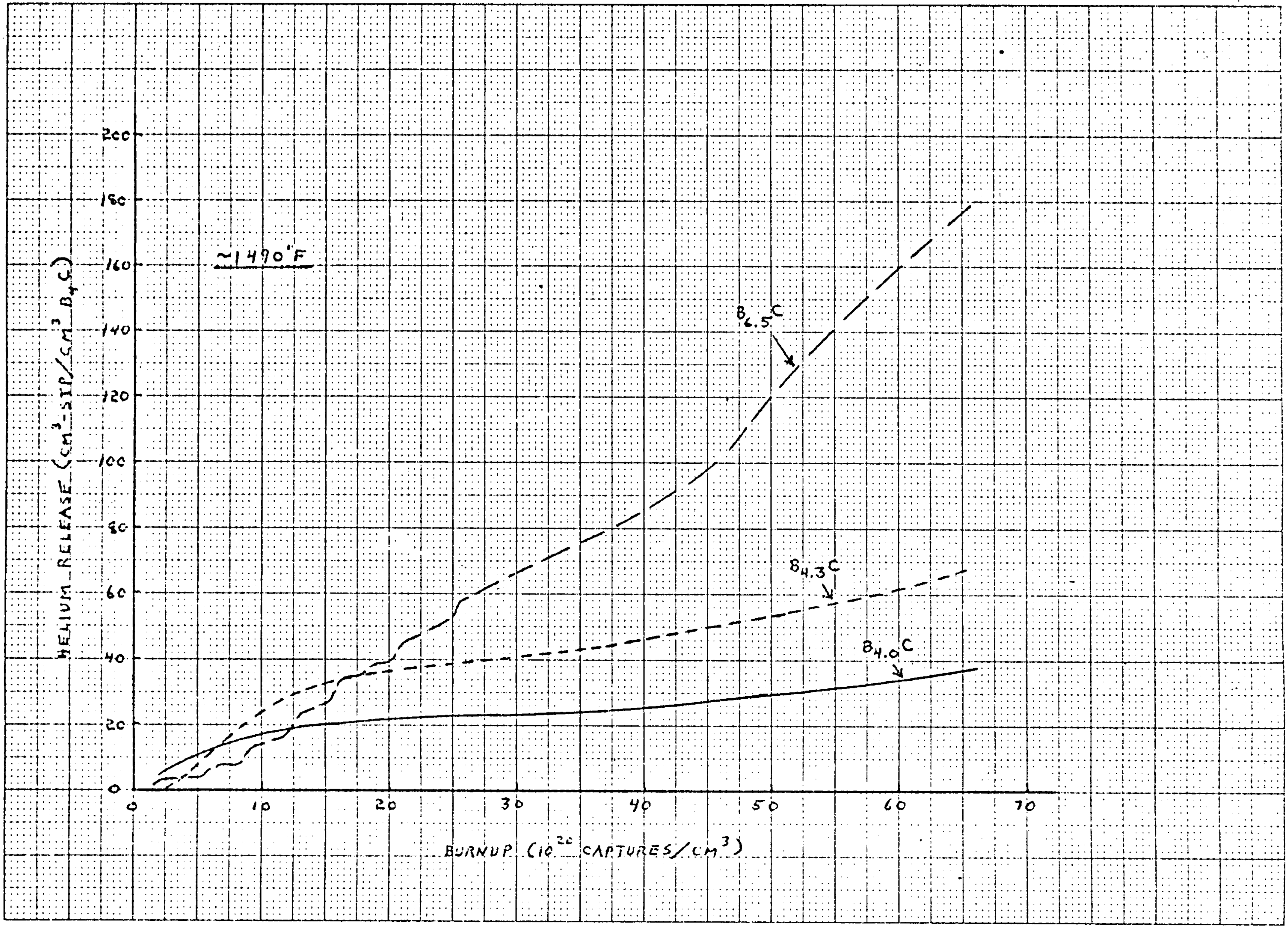


Figure 11. Effect of Stoichiometry on Helium Release

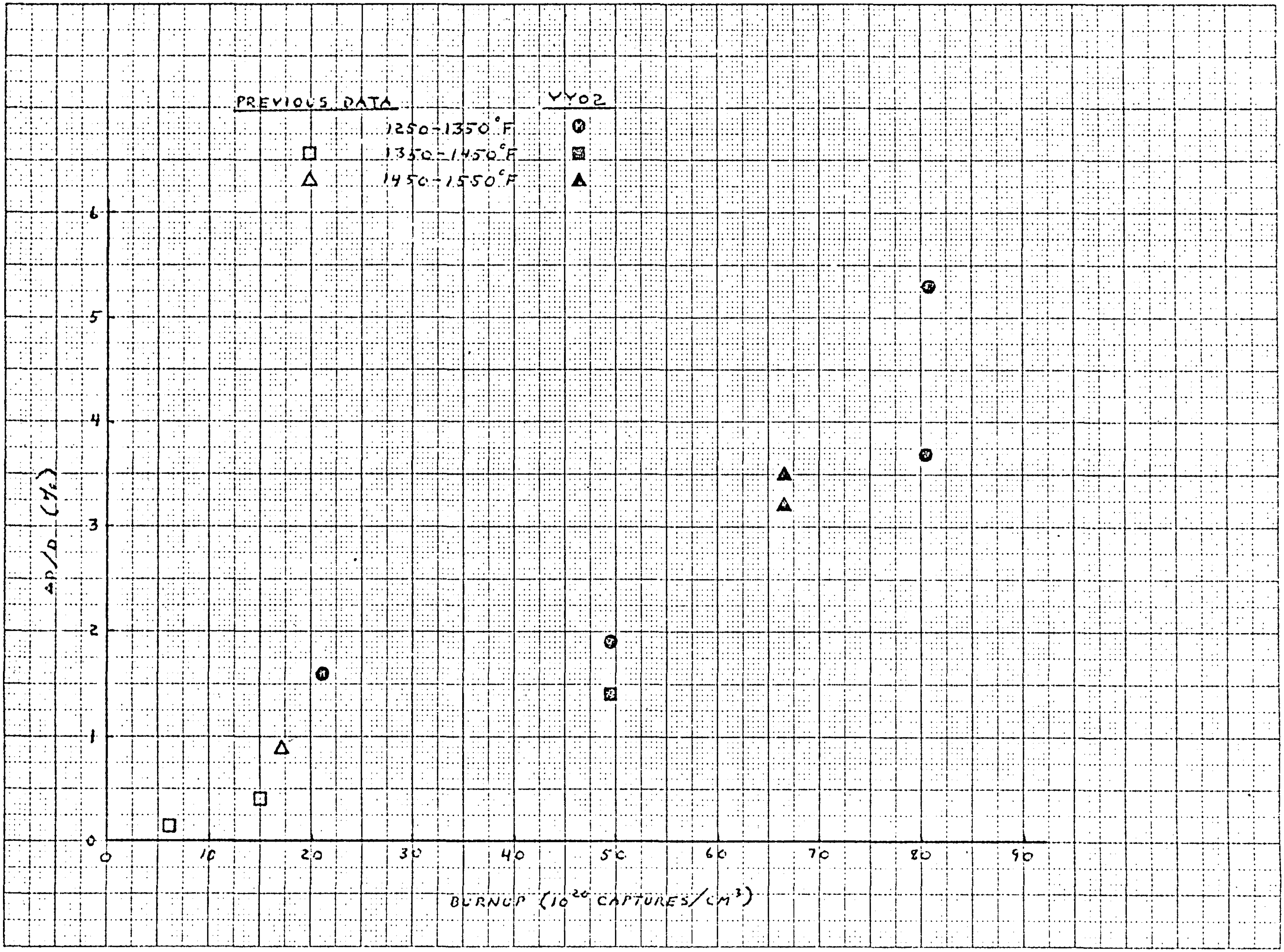


Figure 12. Swelling of FFTF Reference Boron Carbide

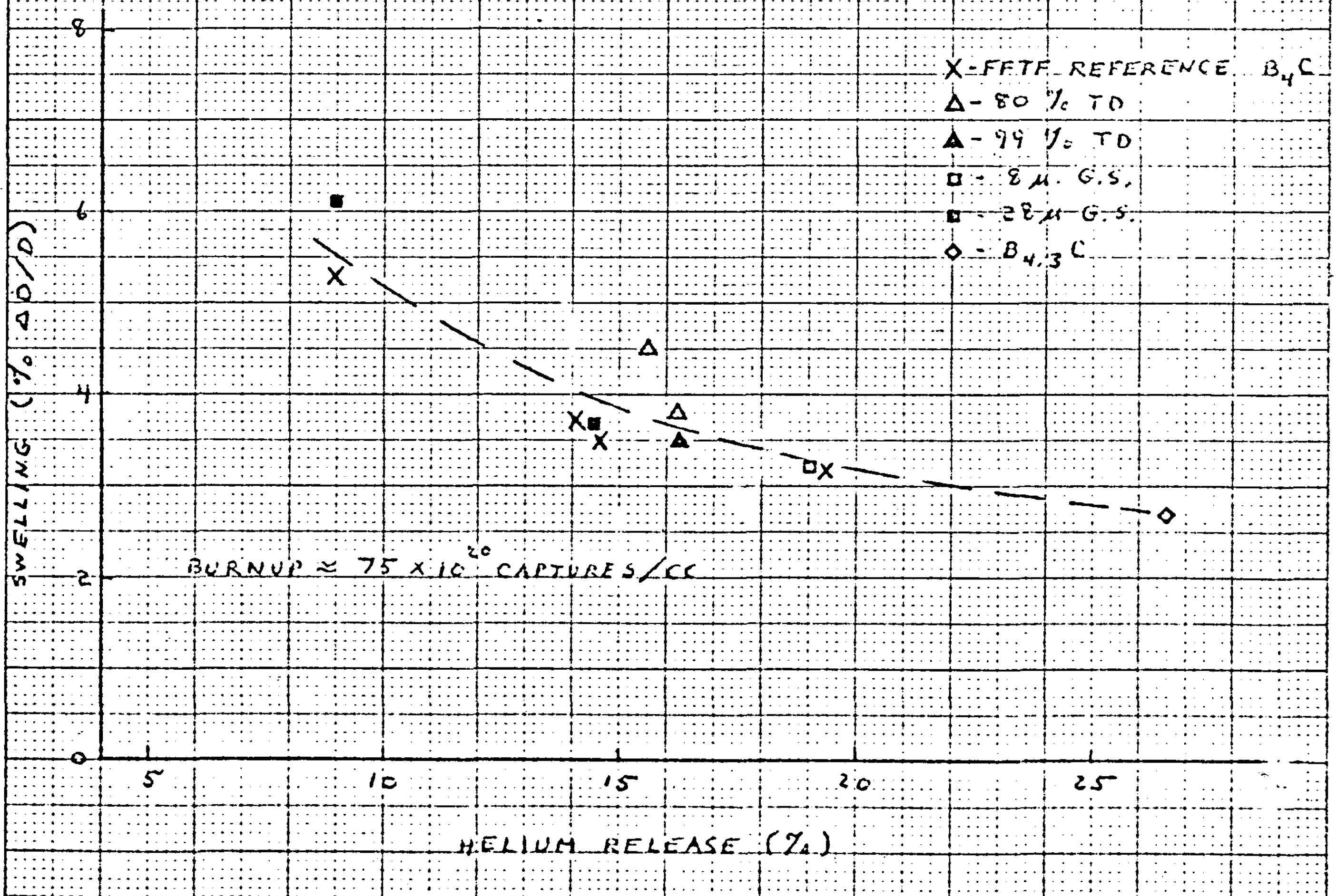


Figure 13. Boron Carbide Swelling vs. Helium Release