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HELIUM RETENTION AND ITS EFFECT ON BORON CARBIDE IRRADIATED IN A FAST REACTOR

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ABSTRACT

The principal parameters affecting performance of boron carbide as a neutron absorber material are its helium retention and swelling during irradiation. Experiments in the EBR-II reactor at temperatures of 540-1000°C and to burnups of 80×10^{20} captures/cm^3 of boron carbide have been performed. Data indicate pellet swelling and integrity relate directly with helium retention.

INTRODUCTION

Boron carbide is, or is being considered for, the neutron absorber in most of the world's breeder reactor control systems.(1) Its usefulness is derived from excellent nuclear characteristics, stability in the reactor environment, and the relative ease of fabrication by standard ceramic processing techniques. The major deterrent to the use of boron carbide is the fact that an alpha particle (helium nucleus) is formed each time a neutron is captured. The helium is either retained in the material and causes swelling, or is released to the pin plenum where potentially high pressures can build up. The retention or release of helium is dependent on irradiation conditions and the material characteristics of the boron carbide.

Numerous irradiation experiments have been performed in the EBR-II fast reactor in the course of development of boron carbide control pins for the Fast Flux Test Facility reactor. The objectives of these experiments were primarily to determine the gas release and swelling characteristics of boron carbide. The range of temperatures (540°C-1000°C) and ^{10}B burnups (to 80×10^{20} captures/cm^3) studied were at or beyond those predicted for FFTF control rods, and are consistent with other breeder reactor requirements.

DESCRIPTION OF THE IRRADIATION EXPERIMENTS

Gas release and swelling data have been derived from fifty-five EBR-II irradiation test units. Thirty-seven of these were irradiations of boron carbide pellets contained in sealed stainless steel pins, and eighteen were performed in capsules with temperature and pressure instrumentation. The pin tests were composed of 34 cm or 15 cm columns of individual pellets. The instrumented capsules contained 2.5 cm columns of four individual pellets. Tables 1, 2, and 3 contain the pertinent experiment design matrices.

The materials used in the irradiations were of two basic types. Reference materials were comprised of 92% ±2% theoretically dense pellets having B/C ratios of 4.0 ±.1% and were formed by conventional hot-pressing. Non-reference materials incorporated variations in grain size, stoichiometry, composition, and processing. The effect of the ^{10}B isotopic content in the boron is not considered a variable other than in determining the rate of burnup.

HELIUM BEHAVIOR CHARACTERIZATION

In order to provide pressure buildup data for control pin design, and to understand the irradiation behavior of boron carbide itself, a knowledge of where and how the helium is distributed in the pin and the boron carbide is required. During the irradiation process, the majority of the helium generated by the $^{10}B\ (n,\alpha)\ ^{7}Li$ + 2.79 MeV reaction is imbedded within the boron carbide pellet. However, some of the atoms near "free" surfaces are immediately recoiled out of the material. The thermal conditions, neutron irradiation damage, and the boron carbide structure are involved in any subsequent retention and/or release of the helium from the pellets. As a routine post-irradiation measurement, laser puncturing of the plenum is performed on all pins to determine the volume and composition of the gas. Together with the total burnup of the pellets a gas release fraction is determined.

The eighteen capsule type irradiations featured continuous pressure monitoring by means of strain gauge pressure transducers. The transducers had absolute accuracies of ±0.5 psi. Since gas temperature varied in the line from the capsule to the transducer, the effective volume-averaged temperature had to be determined. This was accomplished by taking "null" pressure readings prior to inserting the experiment in the reactor, and then again just after full reactor power was attained.

Figure 1 shows the typical isothermal gas release behavior as a function of burnup. Generally the peak fractional gas release occurs at values between 5 and 15×10^{20} captures/cm^3.

The value of the continuous gas release data is apparent when the isothermal gas release behavior is considered as a function of burnup. In Figure 2 the continuous gas release behavior of an 820°C instrumented capsule is shown together with the data from pins. The fractional gas release is defined as that portion of the total gas generated which has escaped from the boron carbide at a particular burnup level, e.g. if the total burnup is 60×10^{20} captures/cm^3, and only 45×10^{20} helium atoms remain in the pellet, the gas release fraction is 0.25.

In the neighborhood of 760°C there is an apparent maximum in the gas release vs. temperature relationship (Figure 3). Throughout the burnup range studied this condition is valid. No studies have been performed to isolate the cause of this behavior. However, it is theorized that this phenomenon is due to subtle microstructural effects.

The general gas release-temperature relationship, sans the 760°C phenomenon, is represented as increasing gas release with increasing temperatures. Figure 3 shows the relationship with the 760°C peaking superimposed. After about 20×10^{20} captures/cm^3 there is less than 5% gas release from 560°C irradiations.

SWELLING

The swelling data involves measurements from approximately one-thousand pellets from pin irradiations and seventy-two capsule pellets. In each case pre-irradiation and post-irradiation measurements were obtained using the same measuring device. The measuring equipment used can accurately reproduce measurements to ±0.0002 in.

In general, swelling increased with burnup throughout the range studied (Figure 4). An inverse temperature relationship was observed in that the greatest swelling occurred at the lowest temperature. An apparent trend in the data is that swelling is directly related to the amount of helium retained in the pellet.

GAS RELEASE AND SWELLING CORRELATIONS

An empirical relationship which best describes the gas release data was generated for the "reference" material using LEARN,[2] a multiple non-linear regression code. Figure 5 is a three dimensional plot of the gas release behavior with the data shown as arrows. The degree of correlation is very good.

An empirical swelling correlation has also been developed. Figure 6 shows the three dimensional view of the swelling data superimposed on the correlation plot. Again, the goodness of fit is evident.

NON-REFERENCE MATERIAL BEHAVIOR

The non-reference materials tested show some interesting helium release and swelling patterns. Variables studied included pellet density, boron to carbon ratios, boron carbide grain size, pellet fabrication technique and impurity additions. Some of these variables did not affect gas release and swelling to any great extent, but boron to carbon ratio variations, and the introduction of impurities did. The material with B/C ratios generally exhibited very high gas release, with gross cracking in the pellets (Figure 7). This high gas release and cracking was unexpected. High boron content was obtained by addition of boron powder to the boron carbide powder prior to hot-pressing. Non-uniform distribution of the boron on a sub-grain size scale probably resulted from this fabrication scheme. Efforts to determine the nature and exact cause of this behavior are in progress.

Atypical gas release seen in pellets containing impurities, and the decrease in fractional gas release with burnup, are related to the boron carbide structure. Examination of these and other B_4C specimens by transmission electron microscopy[3-5] show clearly the form and concentration of the platelike cavities found (Figure 8).

Consistent with the many theories[(6-9)] of gas release and swelling, the added nuclei introduced by impurities, and the additional heterogeneous nuclei associated with lithium or lithium compounds, can cause less gas release and less swelling. Figure 9 shows the typical structure of a high burnup material containing impurities for comparison with Figure 8. The gas release of material with the impurity addition is compared to the reference material in Figure 10.

Slightly less gas release was observed in material with larger grain size ($\sim$30 μ) compared to the small ($\sim$10 μ) grain size. Density effect on gas release was minimal. However, at very high densities cracking was observed.

SUMMARY AND CONCLUSIONS

Empirical correlations describing helium release and swelling of boron carbide have been formulated from numerous tests at temperatures from 540°C to 1000°C and burnups to 80×10^{20} captures/cm^3. The following observations have been made:

1. Total helium release from boron carbide continuously increases with burnup while fractional release goes through a maximum at low exposure and then decreases.
2. Fractional gas release increases with temperature with a superimposed maximum at 760°C.
3. Swelling varies directly with burnup and inversely with temperature with a superimposed minimum at 760°C.
4. The swelling is directly related to helium retention in the material.
5. Gas release is less and swelling about the same for boron carbide with impurities added.
6. High B/C ratio material exhibits high gas release and gross cracking.
7. Pellet density and grain size variations only slightly affect performance.

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TABLE 1

Reference Material Pin Irradiations

Stoichiometry	-	B/C = 4
Pellet Density	-	90-94% of theoretical
Fabrication Technique	-	Hot Pressing ∿2150°C
Average Pellet Irradiation Temperatures	-	540-900°C

TABLE 2

Non-Reference Material Pin Irradiations

Stoichiometry	-	B/C = 3.9-6.5
Pellet Density	-	80, 90-94, 99
Fabrication Techniques	-	Hot Pressing; Cold Pressing and Sintering
Grain Size	-	10, 15, 30 micron
Impurity Additions	-	0-1% Fe
Average Pellet Irradiation Temperatures	-	600-1000°C

TABLE 3

Instrumented Capsule Irradiations

^{10}B Enrichment	-	20, 50, 92%
Pellet Density	-	80, 92, 99% T.D.
Grain Size	-	10, 15, 30 micron
Stoichiometry	-	B/C = 4.0, 4.3, 6.5
Temperature	-	650-870°C

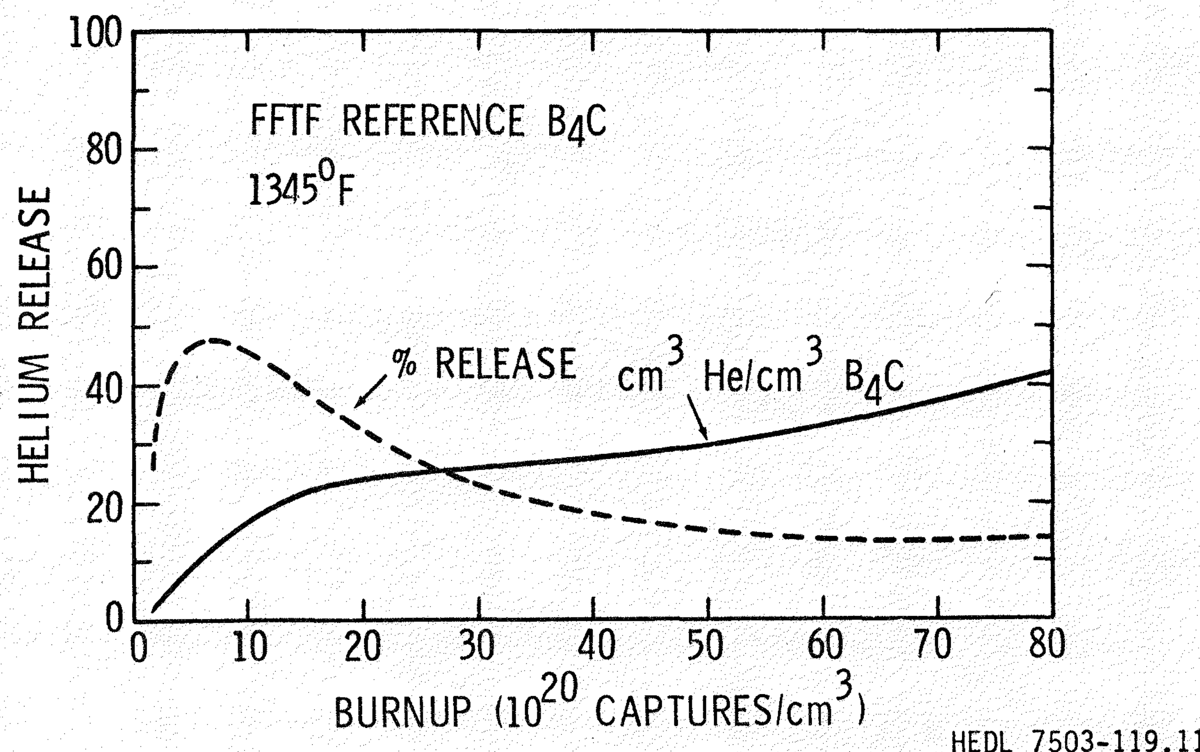


Figure 1. Typical Helium Release Behavior for Isothermal Irradiation

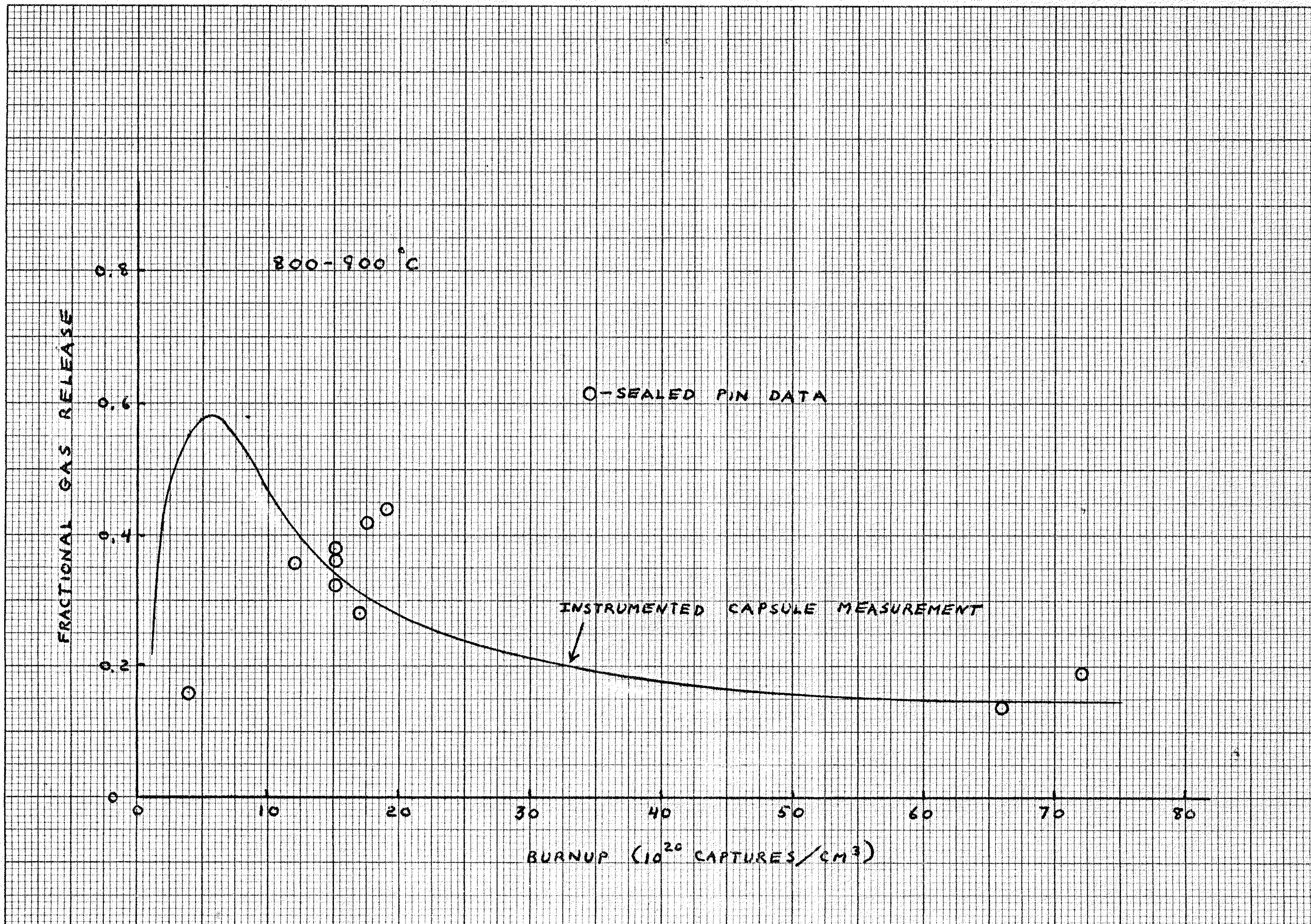


Figure 2. Comparison of Instrumented and Non-Instrumented Gas Release Data

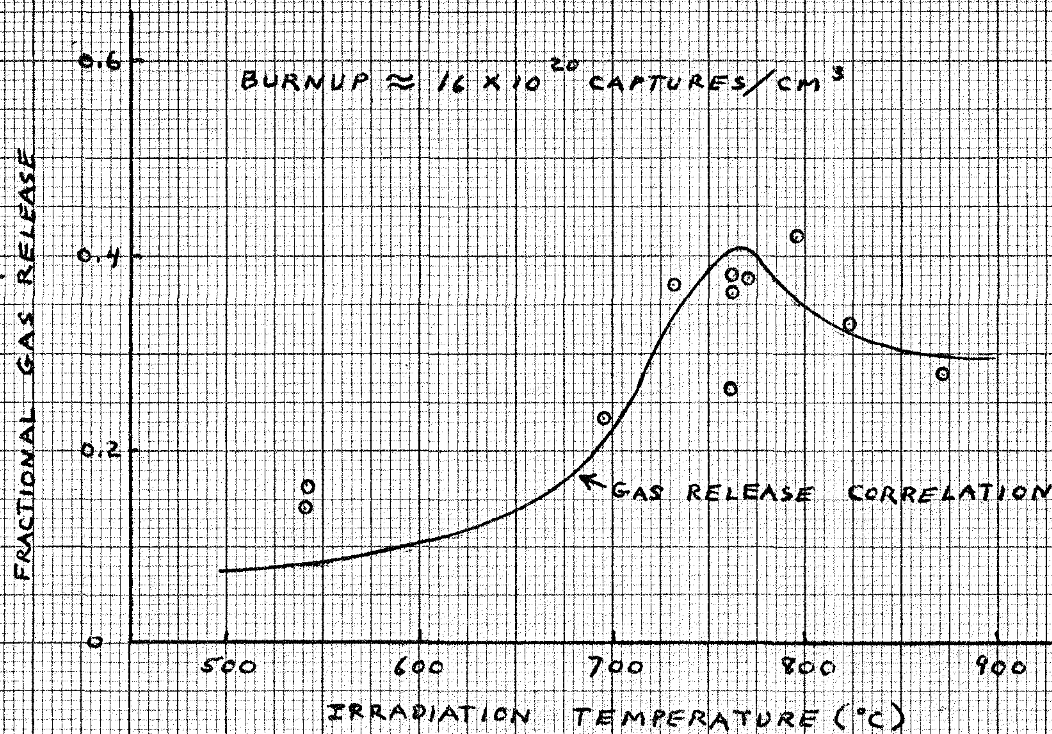


Figure 3. Temperature Dependence of Gas Release

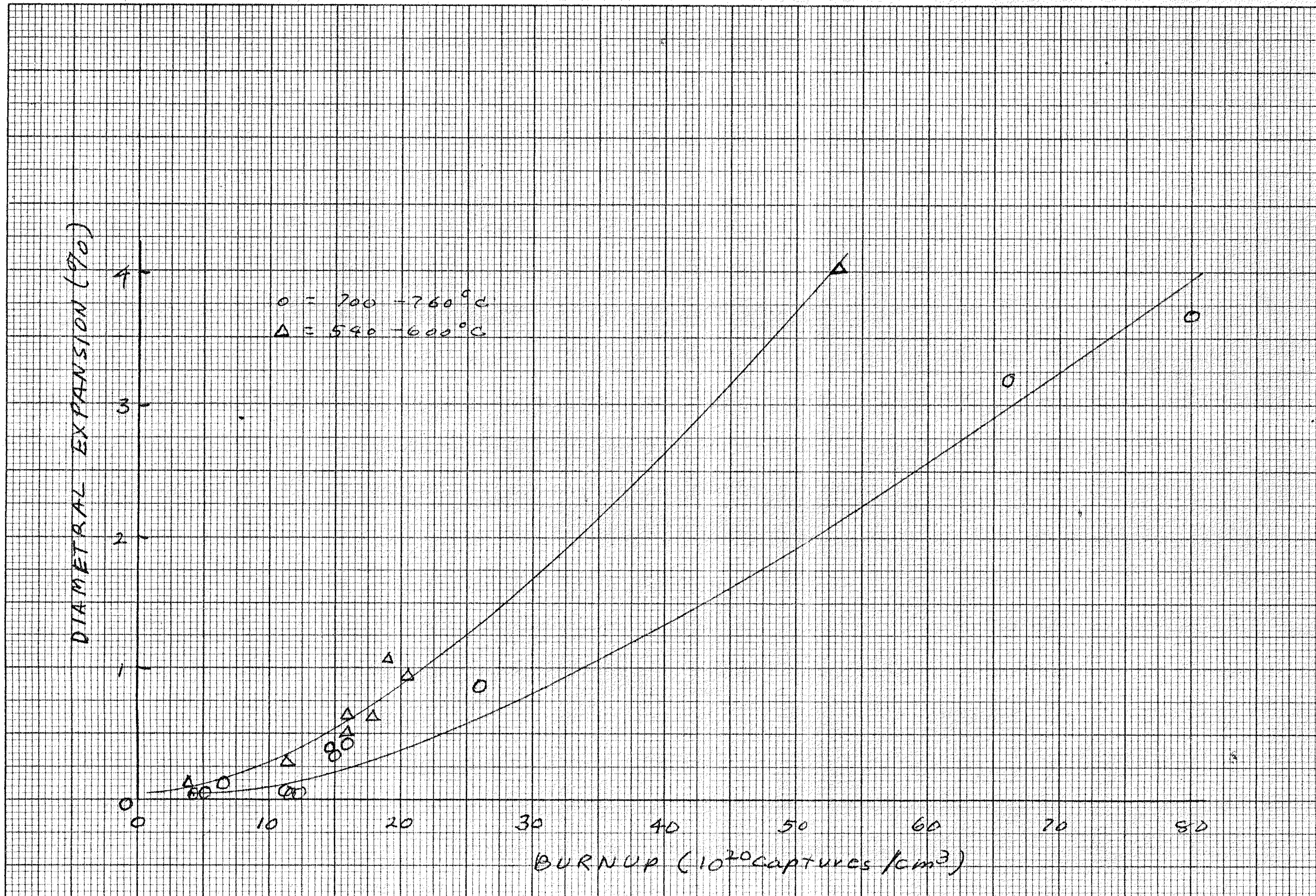


Figure 4. Swelling Behavior of Boron Carbide Pellets

Fig 5. BORON CARBIDE GAS RELEASE

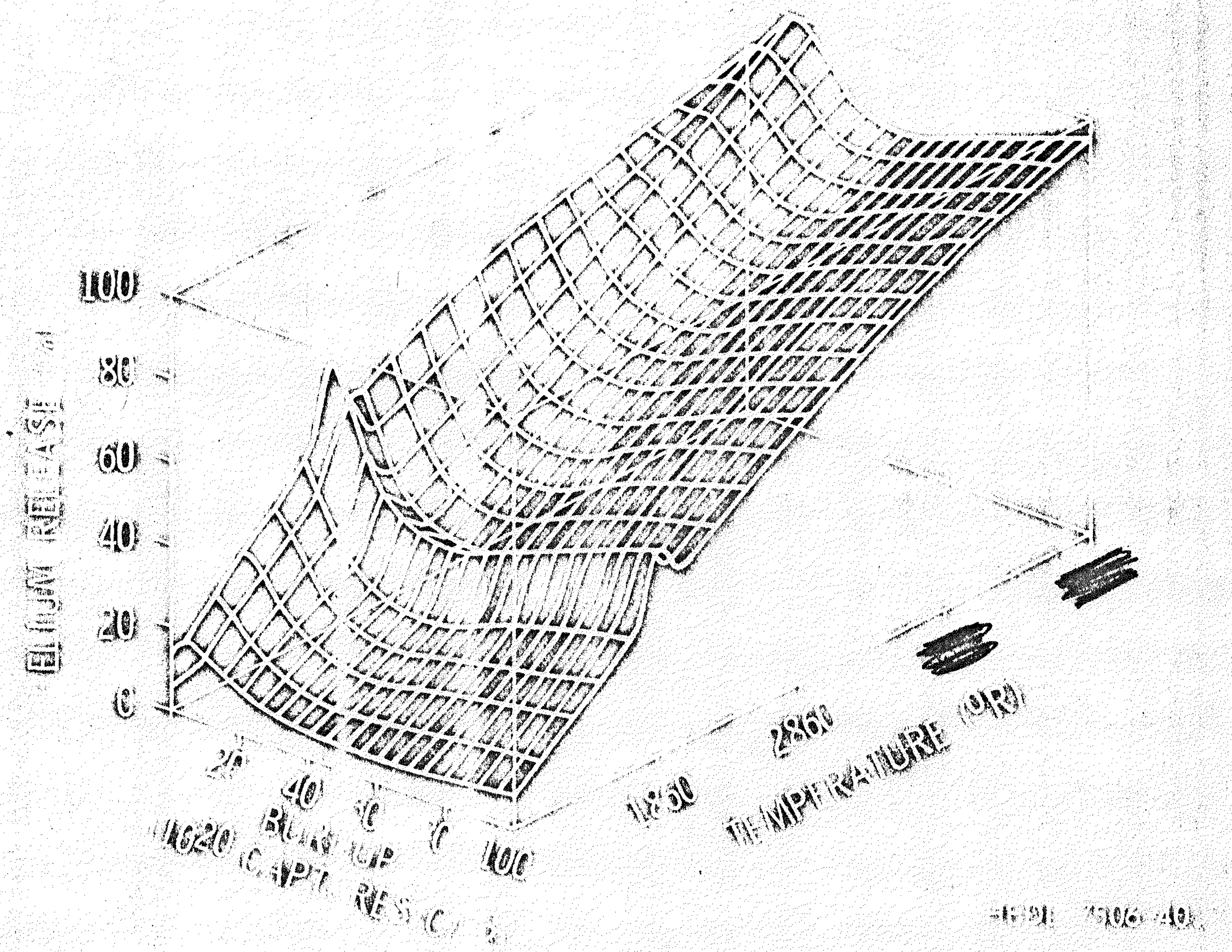
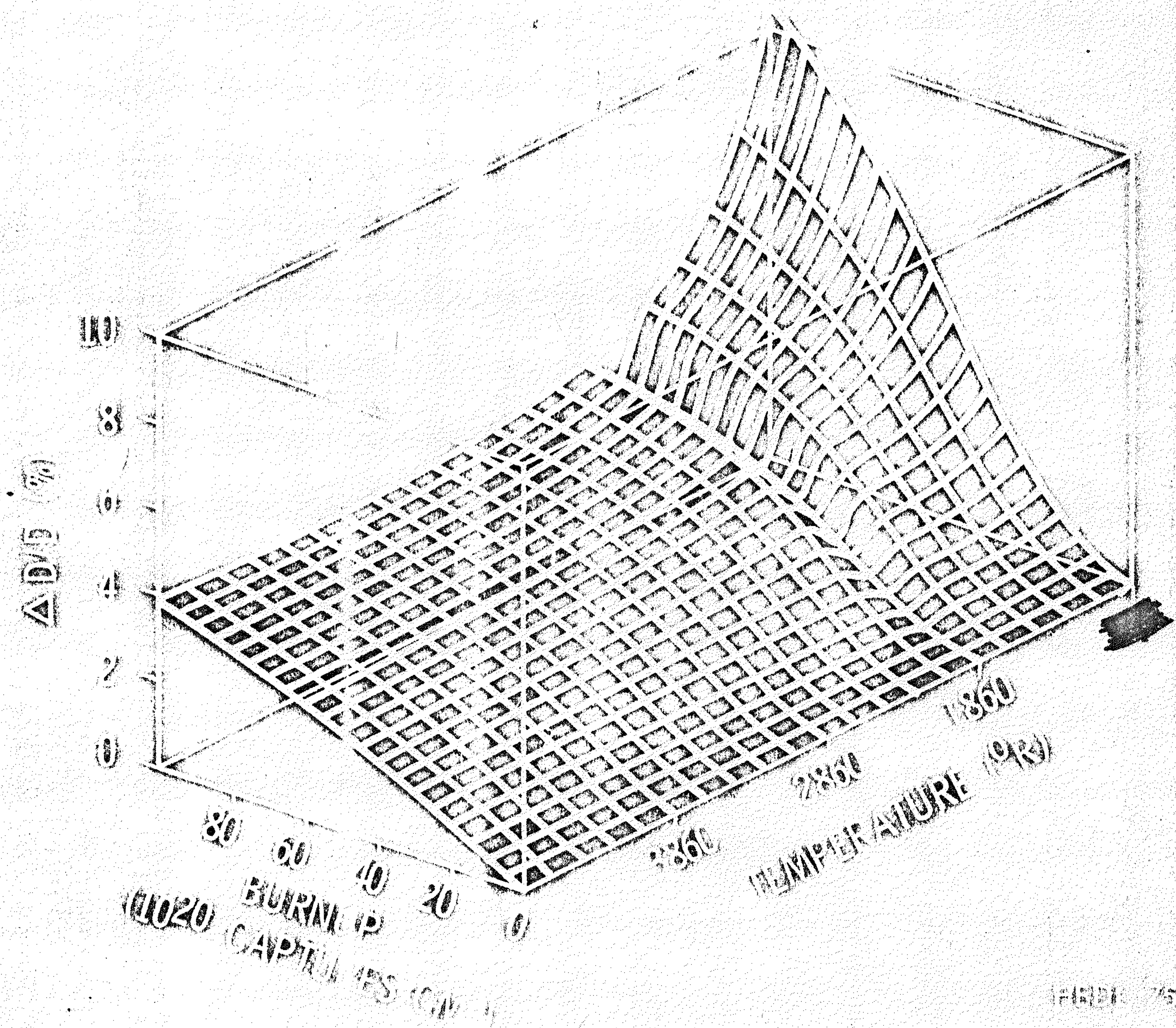


Fig 4 BORON CARBIDE SWELLING [redacted]



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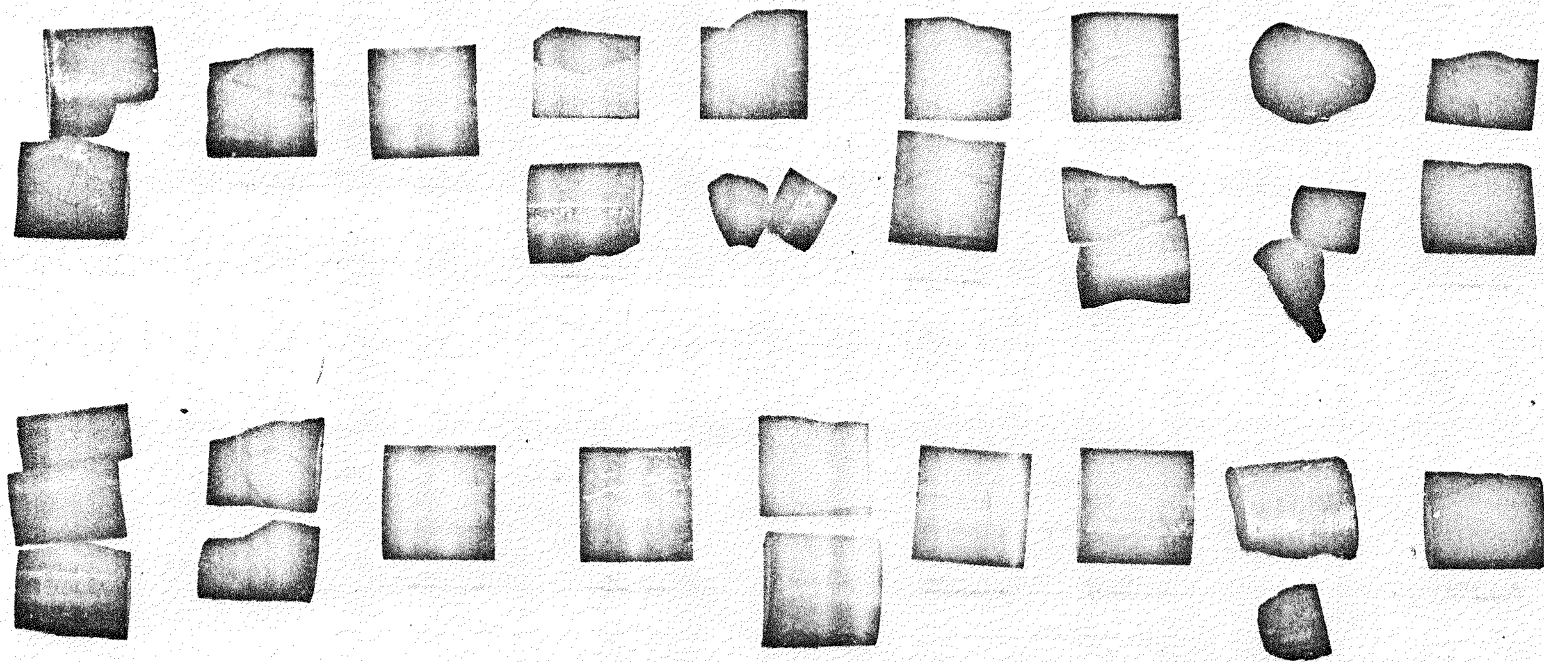

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Figure 7 Cracking Observed in High B/c Boron Carbide Pellets

Figure 8. Transmission Electron Micrograph of Reference [redacted] Boron Carbide Material, As-Irradiated

Figure 9 Transmission Electron Micrograph of Boron Carbide Material containing Impurity Additions, As-Irradiated

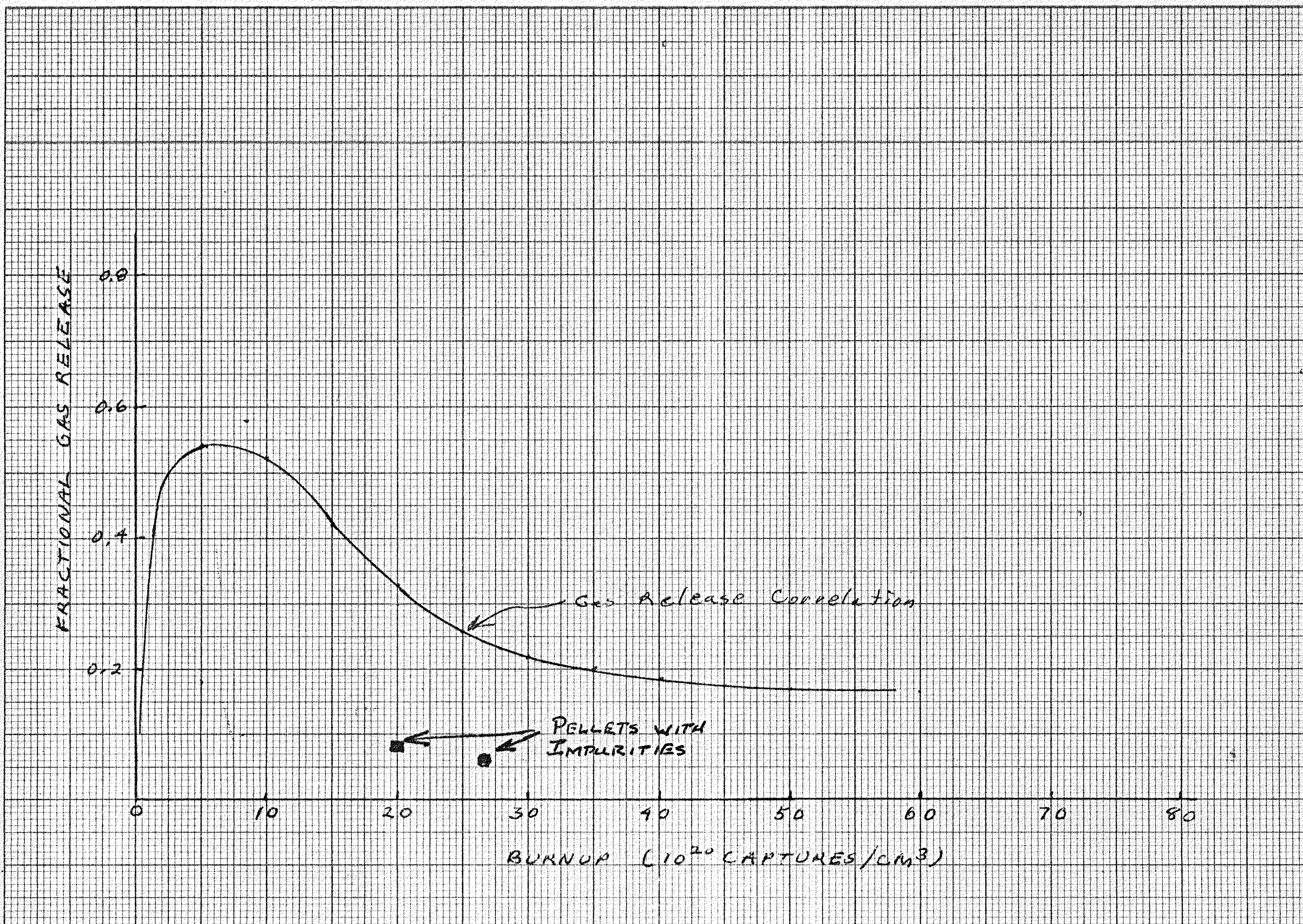

Figure 10. Effect of Impurity Additions on Gas Release