HEDL-SA-1189-FP

CONF-770622--7

A CORRELATION FOR BORON CARBIDE HELIUM RELEASE IN FAST REACTORS

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April, 1977

To be presented at the 1977 Annual Meeting of The American Nuclear Society, June 12-17, 1977 in New York, N. Y.

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I. INTRODUCTION

Boron carbide possesses excellent nuclear characteristics for fast reactor absorber applications. This attribute, in conjunction with its commercial availability, acceptable physical and chemical properties, and comparative irradiation stability have identified boron carbide as the reference absorber material for use in Liquid Metal Fast Breeder Reactor (LMFBR) control rod systems.

Neutron capture in boron carbide is accomplished primarily through (n,α) reactions involving the ^{10}B isotope; thus, helium gas is produced during irradiation of this material. Most of this helium remains in the boron carbide pellet, but some is released to the control pin plenum. The quantity released is dependent on material characteristics and irradiation conditions. Continuous release with irradiation can cause potentially high gas pressures to build up in control pins with small plena. Characterization of this helium release behavior is, therefore, essential for accurate design and safety analyses of LMFBR control elements.

Extensive testing has been conducted[1,2] to characterize the irradiation performance of boron carbide, including helium release behavior, in fast reactor systems over a broad range of temperature and burnup. The objective of this work was to develop a helium release correlation representative of the data that could be extrapolated to conditions beyond the range of the data. A final objective was to define the method of applying the correlation under varying irradiation conditions.

II. DATA

The test parameters required in formulating the helium release correlation are burnup (10^{20} captures/cm^3), irradiation temperature (°K), and volumetric helium release (cm^3 He/cm^3 B_4C). Helium release quantities refer to standard pressure and temperature conditions. Test data were obtained from two types of experiments: instrumented and noninstrumented. In instrumented tests, data acquisition is continuous as gas pressure and temperature are constantly monitored in each specimen subcapsule. Specimen size is generally quite restricted in instrumented tests, while relatively large numbers of pellets can be included in noninstrumented test pins. With noninstrumented experiments, however, temperatures must be calculated and some uncertainty is necessarily associated with reported values. Also, only end-of-test helium release values are obtained. These are derived by puncturing the test pin after irradiation and measuring the volume of helium present in the capsule plenum. Specimen burnup is obtained by fusing selected pellets to determine the quantity of helium retained in the material. The retained quantity combined with the released quantity defines the total amount of helium produced during irradiation, and therefore the burnup incurred.

An important goal of the HEDL irradiation test program has been to determine the effects of material parameters on the irradiation performance of boron carbide. Consequently, a varied spectrum of material types have been included in the program test matrix. Major parameters investigated include pellet density, grain size, and stoichiometry (boron-to-carbon ratio). In deriving this helium release correlation, only data from specimens that were representative of FFTF reference boron carbide were used in the analyses. Nominally, this refers to material with pellet densities of 90-94% of theoretical, average grain sizes in the range of 10-20 microns, and a boron-to-carbon ratio of ~4.

A total of 61 data points were used in developing the correlation. Each data point typically represents the integrated behavior of multiple test pellets: thus, results from many hundreds of specimens are actually included in the derivation . Irradiation temperatures in this data base ranged from ~755°K (900°F) to ~1255°K (1800°F). Maximum specimen burnup reached was 82×10^{20} captures/cm^3, which represents nearly twice the burnup anticipated in first-core FFTF control rods.

III. ANALYTICAL FORMULATION

Helium release in fast reactor environments can be represented reasonably well by the product of independent burnup and temperature related functions. Such a form was chosen for this correlation. Burnup dependency of helium release over the range of data has been well defined by instrumented test results[(2)], where gas pressures in pin plena were continuously monitored during irradiation. The observed behavior reflects an initially high release rate, followed by a lower, relatively constant release rate. At burnup levels greater than ~50×10^{20} captures/cm^3, the release rate once again slowly increases. This behavior can be described by an asymptotic low burnup function of the form, $x^2/(a + bx^2)$, multiplied by an exponential high-burnup function, $\exp(cx)$, where x is burnup and the remaining terms are constants.

The observed temperature dependency reflects generally increasing release with increasing temperature, with a superimposed peak at ∿1050°K (1430°F). The formulation selected for the temperature dependence was a negative exponential [d exp (-f/θ)] combined with an additive Gaussian function. The term, θ, represents temperature in this expression, and the remaining terms are constants. By the use of this Gaussian function, the data are better fit at temperatures near 1050°K (1430°F).

The complete form of the helium release expression is then

$$R\ (cm^3\ He/cm^3\ B_4C) = B * T$$

$$B = \frac{x^2}{a + b\,x^2} * \exp\,(cx)$$

$$T = d * \exp\,(-f/T) + g * \exp - \left(\frac{h - \theta}{k}\right)^2$$

where x is burnup (10^{20} captures/cm^3), θ is temperature (°K), and a, b, c, d, f, g, h, and k are constants.

IV. HELIUM RELEASE CORRELATION

The data were fit by regression analysis to the described functions using the LEARN code.[3] The resulting correlation provides a good fit through the data. Analytically, it is expressed as:

$$R\ (cm^3\ He/cm^3\ B_4C) = BT$$

$$B = \frac{x^2 \exp\,(0.0116x)}{0.688 + 0.0184\,x^2}$$

$$T = 2.62 \exp\left(-\frac{2778}{\theta}\right) + 0.13 \exp - \left(\frac{1056 - \theta}{111}\right)^2$$

where x is burnup in units of 10^{20} captures/cm^3 and θ is temperature in °K. Fractional helium release can be obtained by dividing the volumetric release value by the volume produced (3.722x).

The fit of the correlation to the data is shown in a three-dimensional view in Figure 1. Data points are represented by the arrow tips. There is noticeable scatter in the data, but they are generally well fit by the correlation. The standard error of estimate of the correlation is ±5.4 cm^3 He/cm^3 B_4C.

In Figure 2, the predicted helium release quantities based on the correlation derived here are plotted against the measured values for all the data used in the analysis. The dashed lines represent the standard error of estimate of the correlation. This figure once again serves to illustrate the generally good agreement between measured and predicted behavior. Thus, application of the correlation within the range of data results in high confidence predictions of the helium release behavior.

It should be noted that a good fit to the data was obtained, but high temperature extrapolation resulted in a calculated release rate that exceeded the generation rate at 2000°K (3500°F). The coefficients d and f were modified slightly to accommodate the necessary shape of the high temperature portion of the expression while the lower temperature (<1350°K/1970°F) shape of the function was maintained to give a good fit to the data.

Graphic representation of the burnup and temperature functions of the helium release correlation is presented in Figure 3. The product of the two functions at appropriate burnup and temperature values gives the predicted helium release quantity in units of cm^3 (STP) He/cm^3 B_4C for the given conditions. Helium release curves at various temperatures are shown in Figure 4. It is seen that the release rate approximately equals the production rate at 3500°F and 200×10^{20} captures/cm^3.

V. APPLICATION

For steady-state irradiation conditions (constant temperature and burnup rate), applications of the helium release correlation is direct and straightforward. In actual control rod operation, however, individual absorber sections are exposed to frequently varying irradiation conditions as the control rod is moved axially within the reactor core to maintain reactivity control. Thus specific boron carbide pellets may experience operation at substantially different temperatures during their service lifetime. The question, therefore, arises on how the helium release correlation should be applied under these changing conditions. Recent results obtained in an instrumented irradiation experiment provided valuable insight to this situation. The experiment possessed the capability to control the coolant flow rate past the specimen subcapsule train, and, in effect, the temperature rise in the coolant stream. Specimen temperatures were thus varied ∿200°F from their normal levels on two separate occasions for several days duration. The results are shown schematically in Figure 5. When the specimen temperature was lowered, helium release behavior during this period corresponded to the release rate that would be projected by the correlation at this new temperature. When the temperature was returned to its normal level, however, the helium release rate increased rapidly until total helium release corresponded to the value that reflected steady operation at this temperature. Subsequent helium release behavior was then consistent with continued operation at this normal temperature as predicted by the correlation. Similar results were obtained on both occasions when the temperature was varied. Application of the correlation should be consistent with this type of behavior. Short time steps should be selected to avoid large temperature fluctuations between calculation instances when applying the correlation to design analyses.

VI. CONCLUSIONS

An emperical helium correlation for the helium release from boron carbide has been developed. The correlation provides a good fit to the experimental data in the temperature range from 800°K to 1350°K, and burnup levels up to $80x10^{20}$ captures/cm^3.

The correlation has the capability of extrapolation to 2200°K (3500°F) and $200x10^{20}$ captures/cm^3. In this range the helium release rate will not exceed the generation rate.

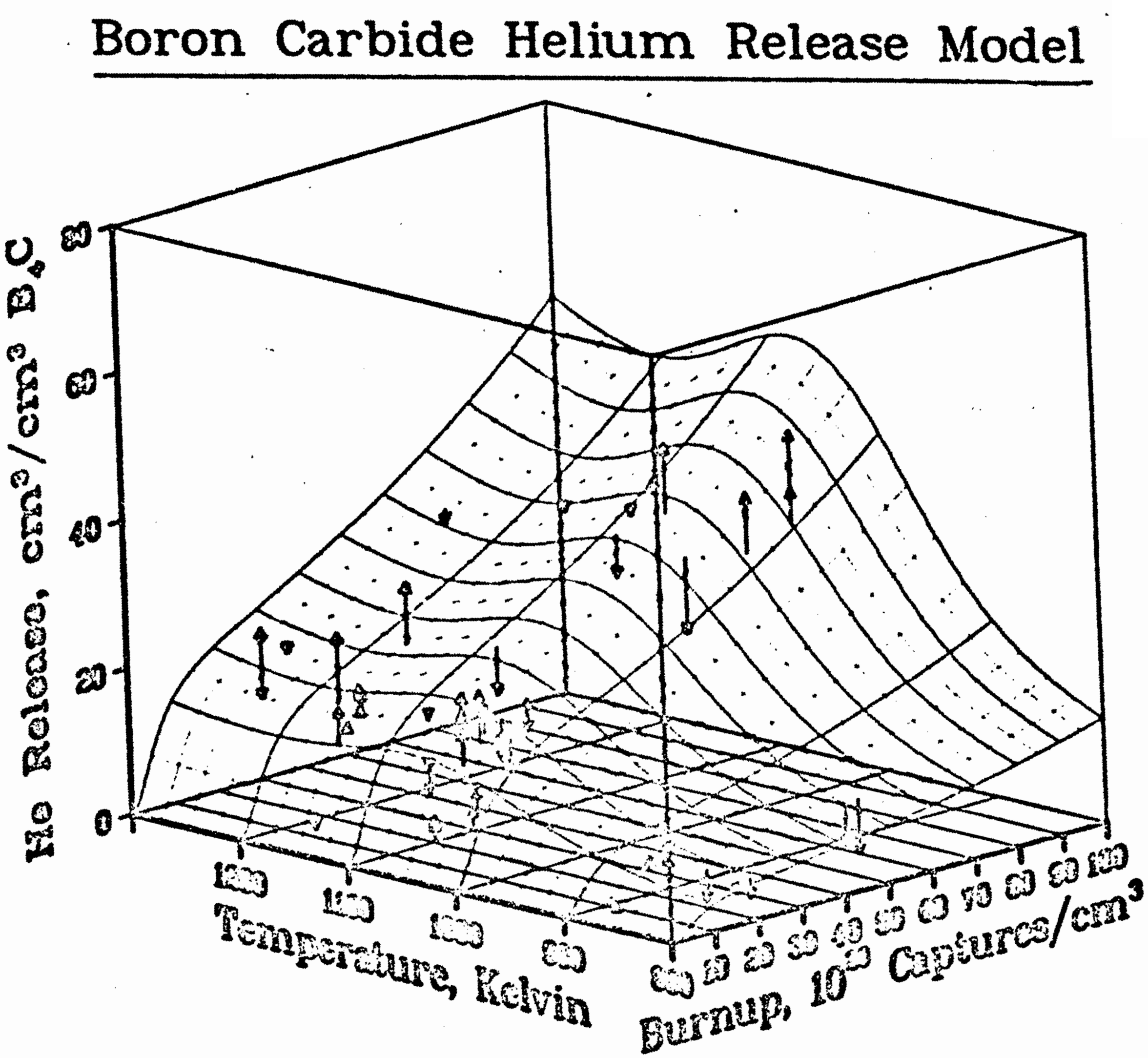

FIGURE 1. Helium Release Correlation and Data Points

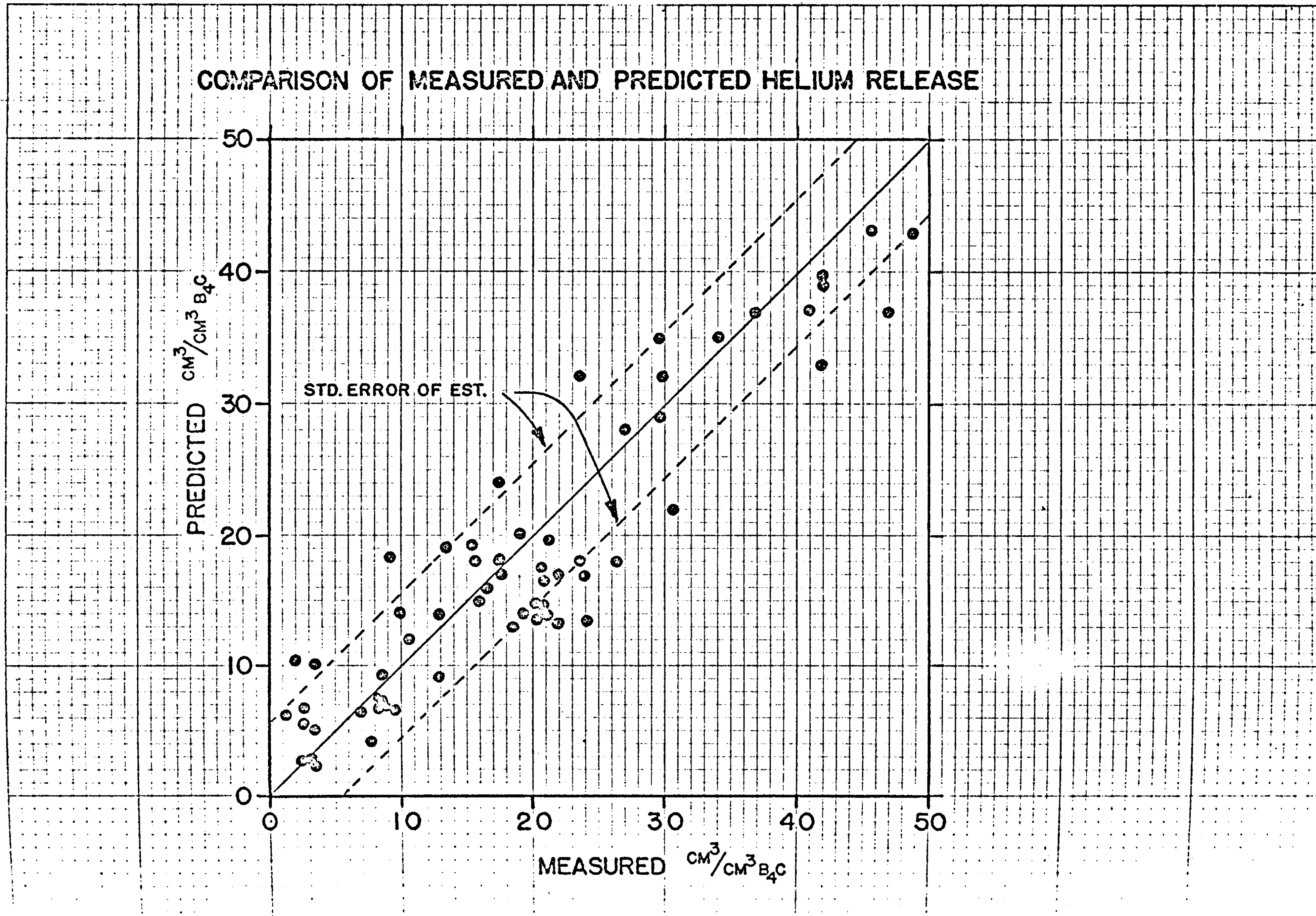
COMPARISON OF MEASURED AND PREDICTED HELIUM RELEASE
STD. ERROR OF EST.
PREDICTED $CM^3/CM^3\ B_4C$
MEASURED $CM^3/CM^3\ B_4C$
0
10
20
30
40
50

FIGURE 2.

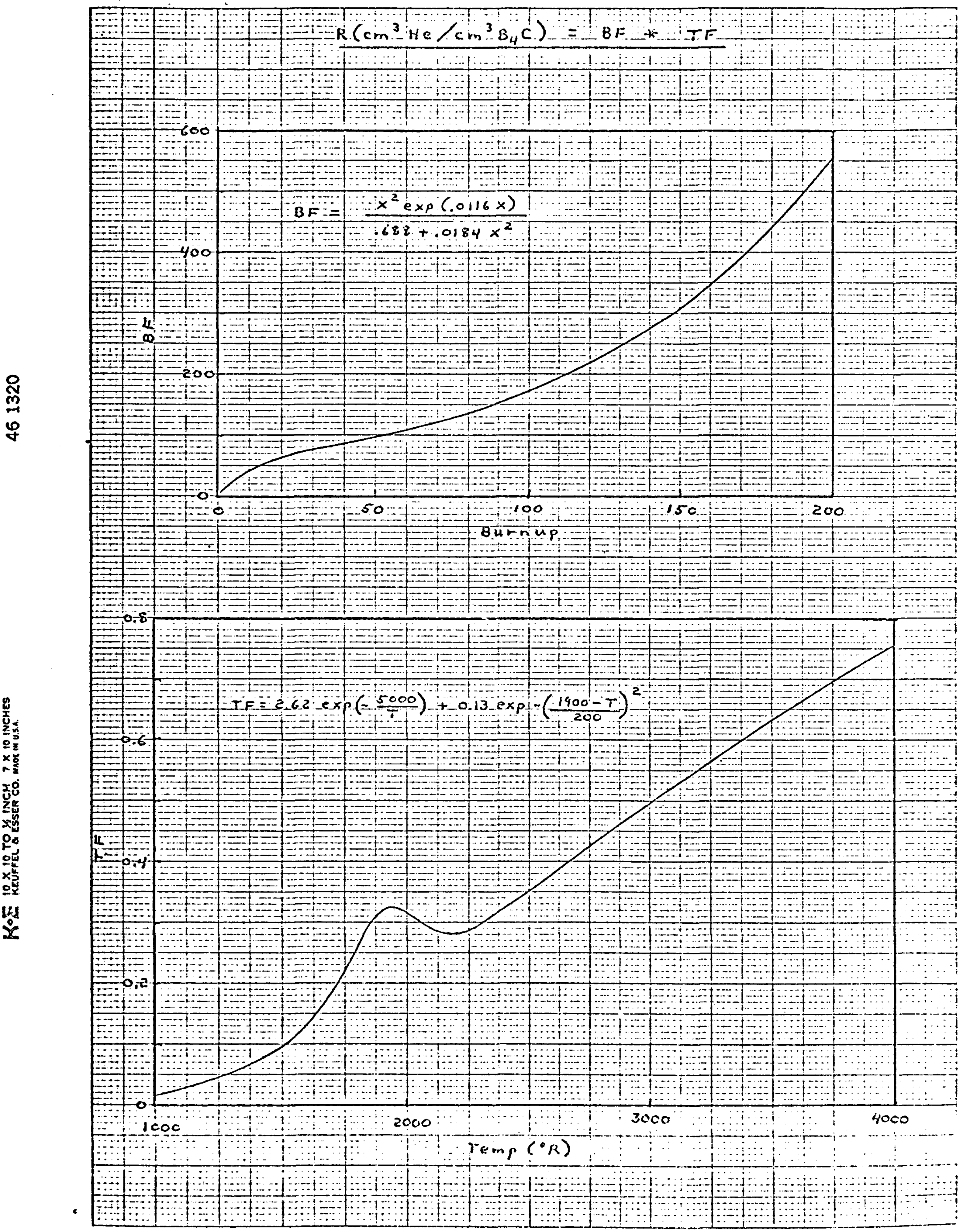

FIGURE 3. Burnup and Temperature Functions of Helium Release Correlation

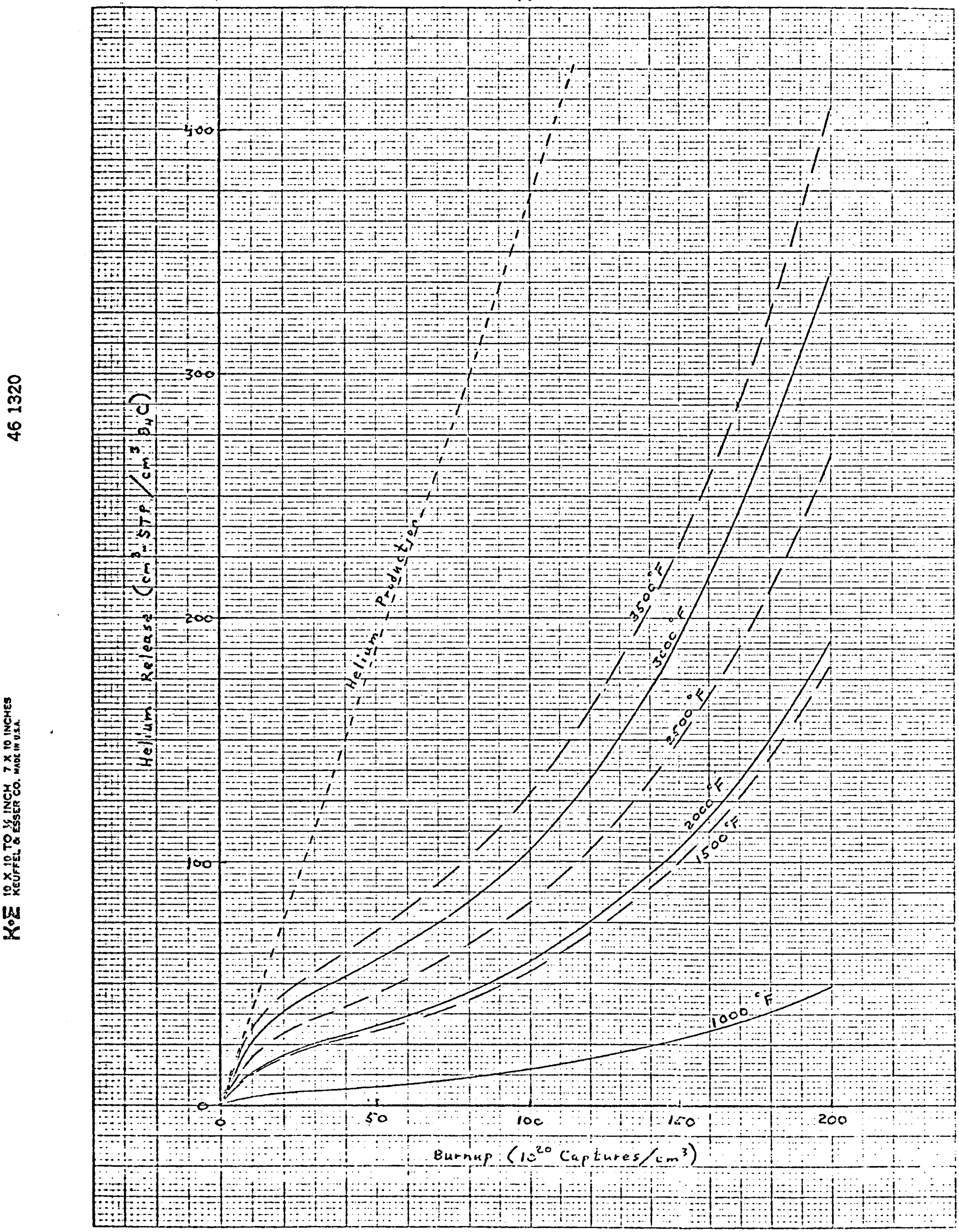

FIGURE 4. Helium Release at Various Temperatures

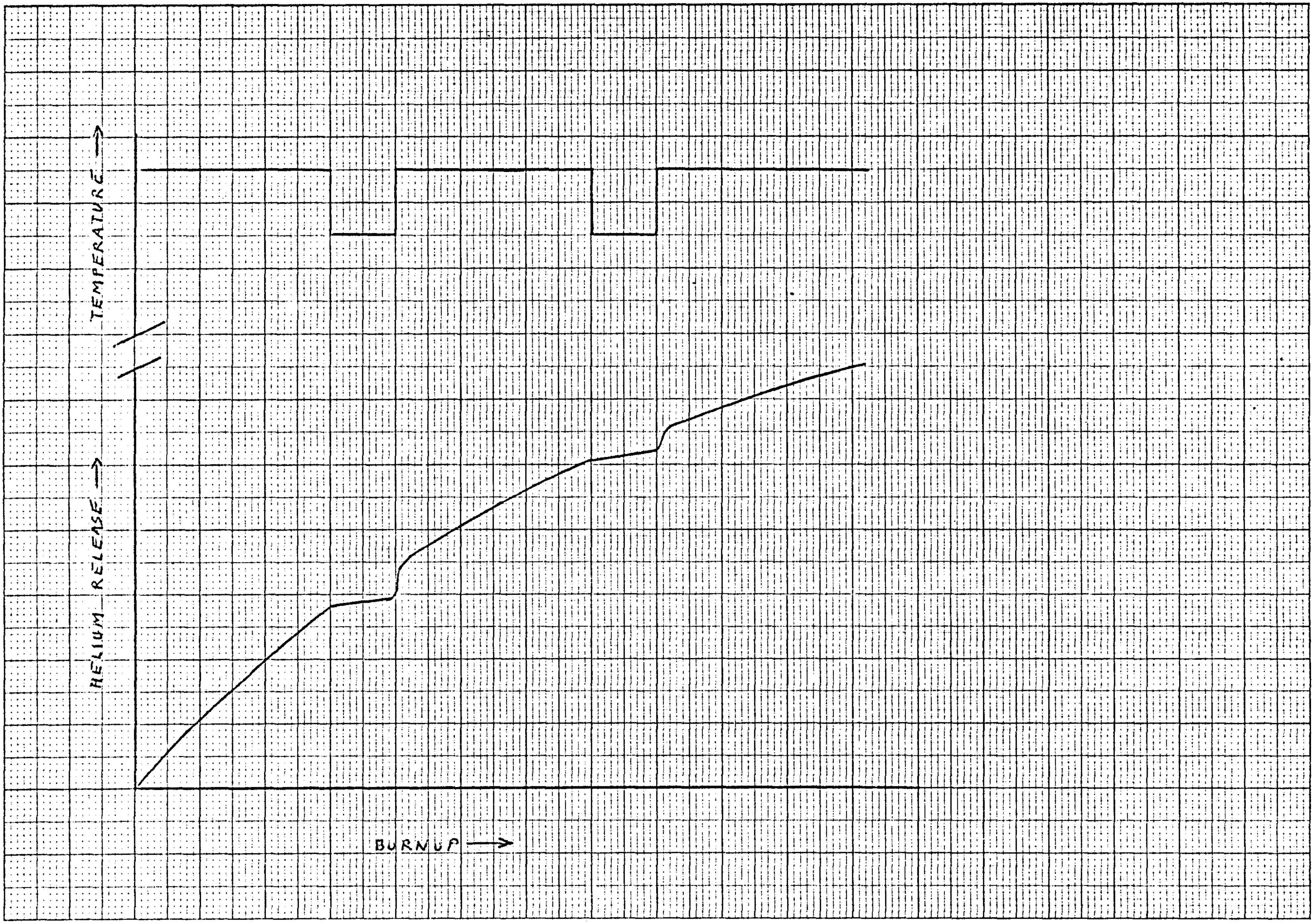

FIGURE 5. Effect of Temperature Change on Helium Release

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FIGURE 5. Effect of Temperature Change on Helium Release