[illegible]ECORD CENTER FILE

DECLASSIFIED

HW-34476

Copy #1 - H. M. Parker [redacted]
#2-10 - Files
#11- 300 File
#12- yellow Copy
(Hold for declassification)

[redacted]

This document consists of 17 pages,
[redacted]

[redacted]

[redacted]

DECLASSIFIED

By Authority of W A Strauser
DORE 4-7-55
By Yellow File
AM Eick 3-26-98
P.D. Aman 5-27-98

RECEIVED
JUL 9 1956
300 AREA
CLASSIFIED FILES

DECLASSIFIED

A REACTOR CONTROL SYSTEM UTILIZING BORON TRIFLUORIDE GAS

W. E. Cawley*

INTRODUCTION

A simple reactor control system which utilizes Boron Trifluroide gas as the neutron absorber has been installed on the Hanford 305 test reactor. It is the purpose of this paper to describe this control system as well as to indicate how a control system of this type could be applied to other research reactors such as the Materials Testing Reactor at the National Reactor Testing Station.

The principle of this type of reactor control is quite simple. If BF_3 gas is introduced into a tube located within the neutron flux, some of the neutrons will be absorbed by the Boron (B^{10} isotope) in the gas. The number of neutrons absorbed will be a function of the number of B^{10} atoms present in the flux which is proportional to the pressure exerted upon the gas. Thus by varying the gas pressure, the macroscopic absorption cross section of the gas in the control may be changed.

FUNCTION AND OPERATION OF THE HANFORD 305 TEST REACTOR USING CONVENTIONAL CONTROL AND SHIM ROD

Figure I shows a general view of the 305 Reactor. It is an uncooled, graphite moderated, natural uranium reactor. The normal power level obtained during operation is six watts. The reactor is used to determine the amount of reactivity which will be absorbed by a given amount of material.

The elevated platform in the foreground contains a graphite train which is used to support the test specimens. The platform height may be adjusted by six power-driven screws. This makes it possible to charge the graphite train and test pieces in any one of the available test holes on the face of the unit.

The control rod and the shim rod enter the test reactor from the far side. A vertical safety rod is also provided. Any one of these rods is capable of shutting the reactor down.

It is interesting to note that, since this reactor is not sealed from the atmosphere, any variation in the ambient pressure will have a relatively large effect on the reactivity available. The atmospheric pressure is therefore recorded periodically during the time the reactor is operating so that corrections may be made to compensate for any change.

For routine tests, a graphite train containing material of known nuclear purity is charged into the reactor core. The shim rod is withdrawn to a position such that, after the vertical rod is withdrawn, the control rod will be withdrawn roughly one-hundred inches before the reactor goes critical.

*i.e. Reactor Operations Engineer, General Electric Company, Hanford Atomic Products Operation.

The position of the control and shim rods is then recorded to the nearest one-thousands of an inch and the barometer pressure is measured. After the graphite train bearing the material of known purity is removed from the core of the reactor, the material to be tested is placed in a similar train and charged.

The rods are again positioned carefully. The difference in the final position of the control rod is noted. By means of a rod calibration curve, the difference in the reactivity absorbed by the known and unknown samples may be determined. This reactivity determination may be made with an accuracy of ± .002 inhours.

BORON TRIFLUORIDE CONTROL SYSTEM - 305 TEST REACTOR

The 305 Reactor operates at very low power, and has a negative metal, graphite and temperature coefficient. It was therefore considered to be an ideal reactor for application of a BF_3 control system. Figure 2 is a schematic sketch which shows the components of the system.(1) The control tube A is located within the graphite core. This tube is connected to the bellows B which is mounted outside of the concrete shield. The gas pressure in this completely closed system may be regulated by means of a precision linear actuator which is attached to the bellows.

The control tube was made from a three-eights inch diameter aluminum tube. The small tube connecting the control tube to the bellows is also made of aluminum. When the volume of system, the desired pressure range and the travel of the linear actuator were determined, it was a simple matter to calculate the size of the bellows required.

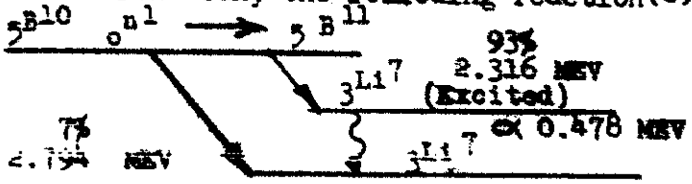
The reactivity effect of the BF_3 control system was determined in much the same manner as the test specimens except that the effect was determined at various pressures so that a calibration curve could be made. It was found that over the narrow pressure range used in this application, the calibration curve is essentially linear. Later recalibrations indicated that no measurable change in control strength of the BF_3 gas had occurred.

There is, of course, a continuous burn-out of the B^{10} isotope while the reactor is operating. However, at the low flux density used in this reactor, it was calculated that the change in control strength due to this cause would be negligible.

If this type of control were to be used in research reactor having a much higher flux, such as the Materials Test Reactor at Arco, or the Brookhaven Graphite Reactor, the problem of B^{10} isotope depletion would become a significant factor as will be discussed in the section on B^{10} burn-out.

NUCLEAR PROPERTIES OF BF_3

When a neutron strikes a Boron atom, the following reaction(2) takes place:



When the Boron is in the form of $B^{10}F_3$, it is expected that the following will occur:

$$B^{10}F_3 + {}_0n^1 \longrightarrow 3F + B^{11*} \longrightarrow (3F) + He^4 + Li^7$$

Assuming the lithium combines chemically with a fluorine atom and the other fluorine atoms remain in the gaseous state; there will then be three mols (LiF, He, F_2) where there was one BF_3 mol originally.

B^{10} BURN-OUT

The rate at which an element is transmuted by irradiation is described by the differential equation:

$$\frac{dn}{dt} = -N \phi \sigma_a$$

integrating, we get: $\int_0^t \frac{dn}{N} = -\int_0^t \quad a \quad dt$

$$\ln N \Big|_0^t = -\phi \sigma_a \int_0^t dt = -\phi \sigma_a (t - t_0)$$

at $t = 0$; $N = N_0$

$$\ln N = \ln N/N_0 = -\sigma_a \phi t$$

Therefore $e^{\ln N/N_0} = -e^{-\phi \sigma_a t}$

Using this relation, it is a simple matter to determine the rate at which the B^{10} isotope would be depleted if a sealed tube containing BF_3 were placed inside, say, the reactor tank of the Materials Test Reactor. The average flux at this position is 2×10^{13} n/cm^2/sec. Figure 3 is a plot of burn-out as a function of time. The calculation assumes there is no reserve BF_3 in the system.

PRESSURE INCREASE DUE TO (n, α) REACTION

For each BF_3 molecule struck, there will be three molecules formed. The LiF is expected to be a solid. It will therefore increase the pressure a negligible amount. The Helium and F_2 will doubtless be gaseous. Thus, effectively, there are two mols of gas formed for each mol destroyed. Figure 3 shows the resulting pressure increase as a function of time. From these two sample curves, it will be noted that if a given pressure is to be maintained in the system by bleeding off the gas as the pressure increases, the control value of the tube decreases not only as a result of B^{10} atoms being burned-out but also because some B^{10} will be removed by bleeding. This factor must be considered in the design of a control system. It indicates that it is probably more desirable to use on-off valves rather than pressure regulators to control the pressure.

HW-34476

CONTROL STRENGTH OF A BF_3 TUBE

If a tube containing BF_3 were placed in the center of the reactor tank of the Materials Testing Reactor, for instance, the reactivity effect would be as shown in Figure 4. It will be noted that the change in reactivity is not a linear function of pressure. The reason for this non-linearity will be made clear by examination of the equation.

$$\Delta K = C \Sigma_a \left(\frac{\phi_{\text{Local}}}{\phi_{\text{Max.}}} \right)^2$$

The macroscopic cross section, Σa, varies directly with pressure increase but the local flux density is reduced as the control strength increases. The curve becomes asymptotic at 100 per cent blackness.

THEORY OF THE B^{10} ISOTOPE MONITOR

The total pressure exerted by the gas will increase as a function of the number of B^{10} atoms which are struck by neutrons. Thus as the "blackness" or control strength is reduced, the pressure increases. To determine the actual control strength, some measurement besides pressure must be made.

Since control strength is directly proportional to B^{10} concentration, it would be most desirable to measure this value directly. Figure 5 is a schematic of the apparatus which may be used to monitor the B^{10} concentration present in BF_3 at any time.

An experiment was performed to determine the feasibility of making this type of measurement. A small capsule of radium-beryllium was used to supply the neutrons in this experiment. Some of the neutrons passed through the paraffin which thermalized them. They then passed into a chamber connected directly to the BF_3 control tube. A neutron counter, located above the BF_3 container, measured the number of neutrons which passed through the gas. As the B^{10} concentration was decreased, the number of neutrons which were allowed to pass through the chamber increased. The sensitivity of this device may be adjusted by varying the strength of the source, changing the volume of the pressure vessel, by varying the thickness of the moderator and by changes in the geometry and shielding.

METHOD OF CONTROLLING BF_3 PRESSURE

Extreme care must be exercised in the selection of pressure control equipment. BF_3 hardens both synthetic and natural rubber and attacks all plastics except those with a fluorine base such as teflon. Several metals such as brass should be avoided if there is a possibility of the gas ever containing moisture. If two or three-way solenoid valves are to be used, they must give a dead-tight shut-off, otherwise the boron concentration will vary with time.

HW-34476

CHEMICAL PROPERTIES OF BF_3

Dry Boron Trifluoride(3) does not attack aluminum, steel, brass, or any other of the usual materials used for construction of pressure vessels and valves. When boron trifluoride is passed into water, it is hydrolyzed and several acidic compounds may be formed. The formation of specific compounds is a function of the kinetics involved, for the first reaction products may, in time, be solvated. Known reactions with available equilibrium constants appear in Table I.

The solubility(4) of boron trifluoride in water is of the same magnitude as that of ammonia in water. At 0°C under an external pressure of 762 mm., one milliliter of water absorbs 1057 milliliters of boron trifluoride. At room temperature, the ratio of absorption is 1:700. This very high solubility suggests solvation of the gaseous solute.

INDUCED RADIOACTIVITY

A typical sample of commercial BF_3 contains the following constituents:

BF_3	96.2 %
SiF_4	2.39%
SO_2	0.4 %
AIR	0.42%

Calculations to determine the induced radioactivity to be expected in commercial BF_3 after one, two and three months exposure in a reactor were made. The calculations assumed that the sample was contained in a pressure vessel with one cubic foot of volume at 360 psig pressure. The maximum calculated activity at a distance of one foot from the container was 1.4×10^{-3} MR/Hr due to gamma. There are, no doubt, small traces of other impurities in the gas which will increase the activity.

METHOD OF DISPOSAL OF THE IRRADIATED GAS

It would not be desirable to allow the spent gas to escape freely into the atmosphere for two reasons:

1. After irradiation, it would be slightly radioactive.
2. Even diluted concentrations of BF_3 in the atmosphere will cause irritation of the skin and throat. There has been no noticeable physical damage to workers employed in the chemical processing plants which produce BF_3, however.

Figure 6 is a schematic drawing of a method of disposing of the gas after irradiation by absorbing it in water.

The gas passes through a bubbler, past a check valve and is absorbed by the recirculating water at the throat of the aspirator. The affinity of water for BF_3 is such that a large quantity of BF_3 may be absorbed by a small volume of water. The final mixture, of course, is acidic. Mild steel coupons exposed to this mixture were attacked quite severely. The stainless steel components were unharmed.

HW-34476

CHOICE OF CONTROL SYSTEM FOR A PARTICULAR APPLICATION

It is obvious that a gaseous control system would be much less expensive than a system using control rods. Also, very fine reactivity control may be obtained by use of the gas control if the system is carefully designed.

The danger of leakage of the gas may be minimized by periodically pressure testing the control system while the reactor is not operating. However, the possibility of loss of pressure is always present.

Because of this possibility, a fast acting, dependable safety system would be required if this type of control were to be applied to a reactor which is to operate at a significant power level and which is not inherently fail-safe.

In the event that the characteristics of the reactor are such that a failure of the normal control system would be disastrous; the gaseous type of control system would not appear to be applicable.

REFERENCES

1. M. V. Davis and W. A. Clark, "Gas Filled Pile Control," HWIR-379.
2. Mayr, G., Bruner, H.D., and Brucer, M., "Boron Detection in Tissues Using the (n,α) Reaction," Nucleonics, Vol. II, No. 10 (Oct. 1953) pp. 21-22.
3. Hudswell, F., Nairn, J.S., Wilkinson, "Corrosion Experiments With Gaseous BF_3," AERE C/R 468, June 1950, (Unclassified).
4. Booth, H.S., and Martin, D.R., "Boron Trifluoride and Its Derivatives," New York, John Wiley and Sons, Inc. (1949), p. 250.

BIBLIOGRAPHY

1. Horst, R.L., "Corrosion Forum," Chemical Engineering, Vol. 59, No. 5, (May, 1952), p. 308.
2. Wilke-Dorfurt, E., and Balz, G., Z. anorg. allgem. Chem., 159, 197 (1926).
3. Sowa, F.J., Kroeger, J.W., and Nieuwland, J.A., J. Am. Chem. Soc., 57, 454 (1935).
4. Partington, J.R., "A Textbook of Inorganic Chemistry," 6th Ed., Macmillan and Co., Ltd., London (1950), p. 657.
5. Clinton, M., A.P.I. Toxical Rev., March 1948, pp. 3.
6. Klinkenberg, L.J., and Ketelaar, J.A.A., Rec. trav. chim., 54, 959 (1935).
7. Klinkenberg, L.J., Chem Weekblad, 34, 23 (1937).
8. Klinkenberg, L.J., Rec. trav. chim., 56, 36, (1937).
9. Kirk, R.E., and Othmer, D.F., "Encyclopedia of Chemical Technology," The Interscience Encyclopedia, Inc., New York (1951), Vol. 6, pp. 678-88
10. Kroeger, J.W., Sowa, F.J., and Nieuwland, J.A., J. Am. Chem. Soc., 59, 965, (1937).
11. Lewis, G.N., and Seaborg, G.T., Ibid., 61, 1886 (1939).
12. Long, L.H., and Dollimore, D., J. Chem. Soc., 1951, 1608.

DECLASSIFIED

HW-34476

NOMENCLATURE

σ_a - Absorption Cross Section in Barns.

Σ_a - Macroscopic Cross Section.

ϕ - Flux Density.

C - Experimentally Determined Constant.

ΔK - Change In Reactivity.

P - Pressure At Any Time.

P_o - Pressure At $t = o$.

N - Number Of Atoms Of An Element Present At Any Time.

N_o - Number Of Atoms Of An Element Present At $t = o$.

DECLASSIFIED

DECLASSIFIED

TABLE I

REACTIONS IN THE SYSTEM BORON TRIFLUORIDE-WATER

1. $BF_3 + H_2O \rightleftharpoons BF_3 . H_2O + HBF_3OH$
2. $HBF_3OH + H_2O \rightleftharpoons HF + HBF(OH)_2$ $\quad K_{equil} = 1.1 \times 10^{-2}$ at 25 C
3. $HBF_3OH + HF \overset{slow}{\rightleftharpoons} HBF_4 + H_2O$
4. $HBF_4 + 3H_2O \rightleftharpoons H_3BO_3 + 4HF$
5. $HBF_4 + H_2O \rightleftharpoons HF + HBF_3OH$ $\quad K_{equil} = 2.3 \times 10^{-3}$ at 25 C
6. $BF^3 + 2H_2O \leftarrow BF_3 . 2H_2O \rightleftharpoons [H_3\ O+]\ BF_3OH \rightleftharpoons HF + HBF_2(OH)\ 2$
7. $3(BF_3 . 2H_2O) \rightleftharpoons HBF_2(OH)_2 + HF . 2BF_3 . 4H_2O$
8. $6HBF_2(OH)_2 \rightleftharpoons 4BF_3 + B_2O_3 + 9H_2O$
9. $HBF_2(OH)_2 + 2H_2O \rightleftharpoons 2HF + H_3BO_3$
10. $4BF_3 + 3H_2O \rightleftharpoons 3HBF_4 + H_3BO_3$
11. $H_3BO_3 + HF \overset{fast}{\rightleftharpoons} HBF\ (OH)_3$
12. $HBF(OH)_3 + 3HF \overset{slow}{\rightleftharpoons} HBF_4 + 3H_2O$
13. $H_3BO_3 + 3HF \rightleftharpoons HBF_3OH + 2H_2O$
14. $2BF_3 + 3H_2O + H_3BO_3 \rightleftharpoons 3HBF_2(OH)_2$

For the over-all reaction of boron trifluoride and water, the equilibrium constant k(liters/mole-min.) = $6.4 \times 10^{-2} + 7.35\ (H+)$, at 25 C.

HW-34476

HP 352

HW-34476

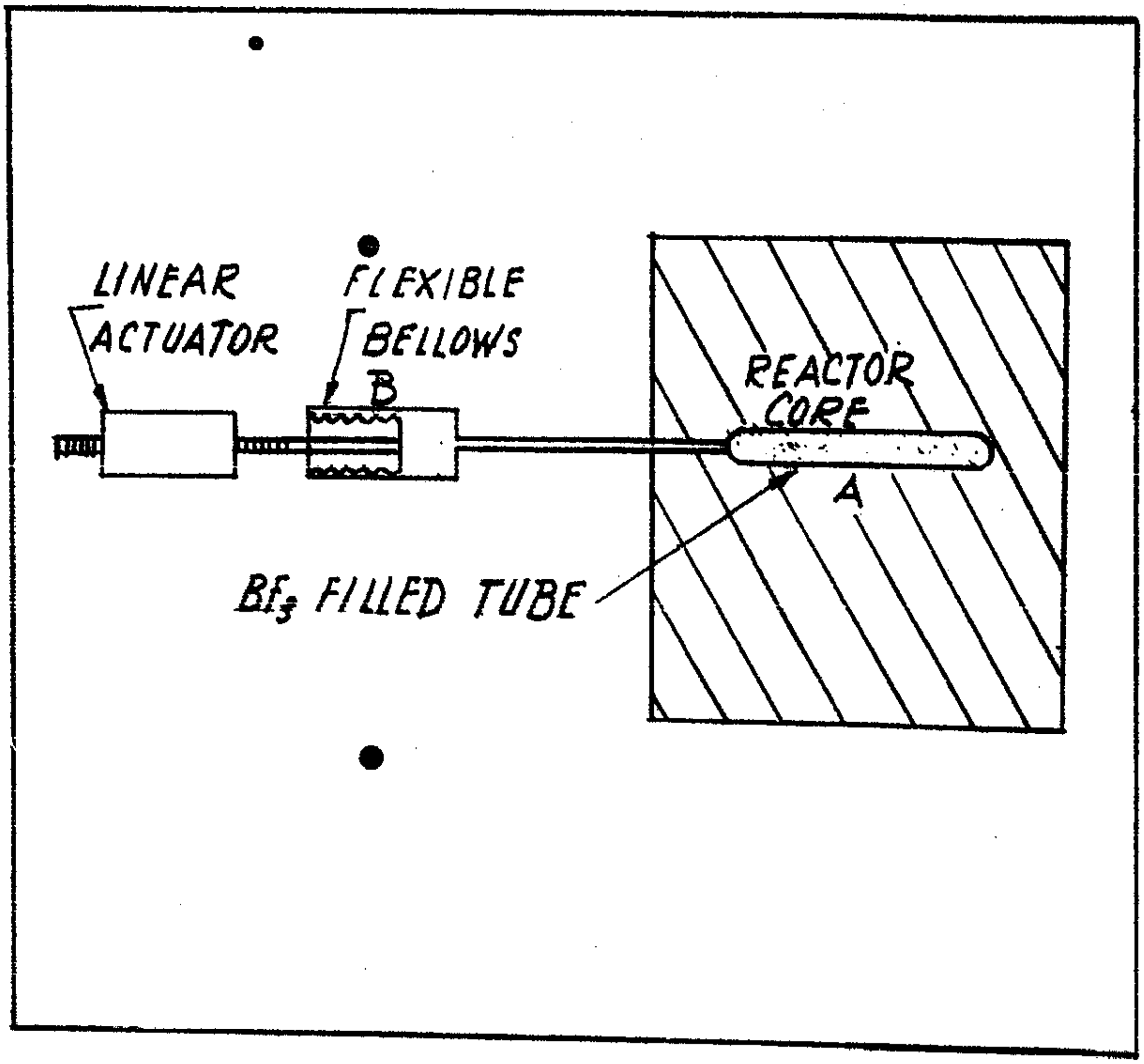


FIGURE 2

HW-34476

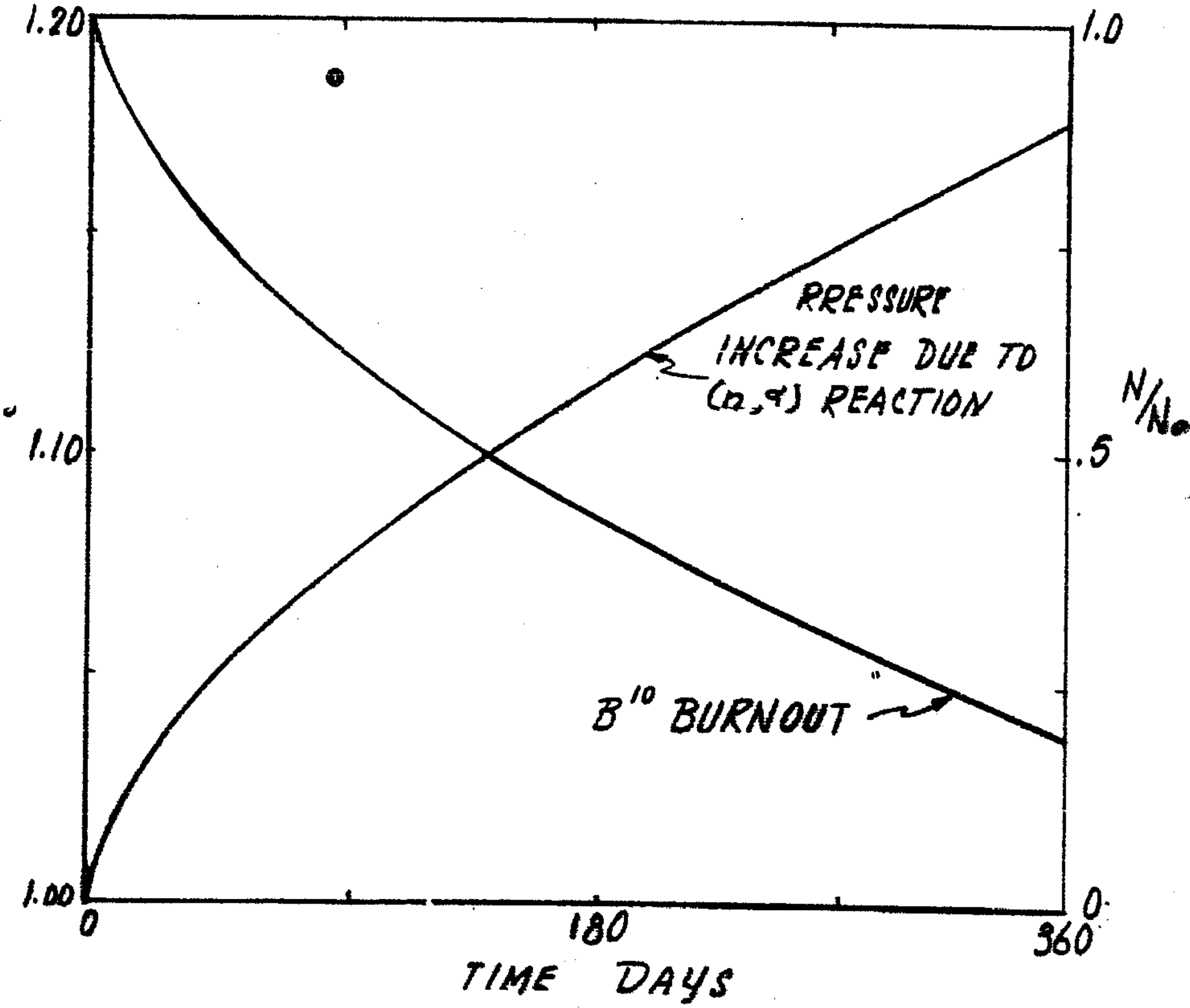


FIGURE 3

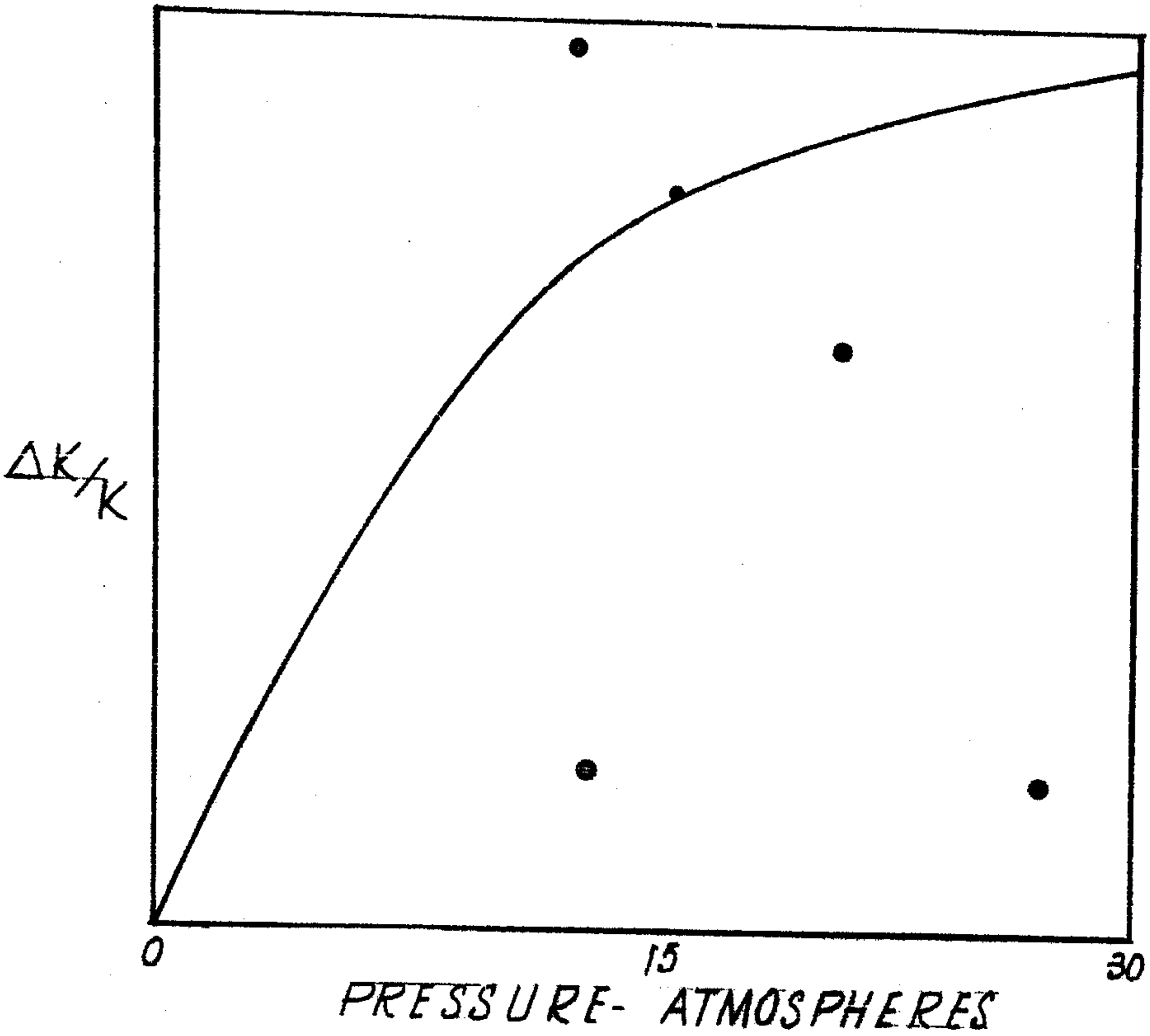


FIGURE 4

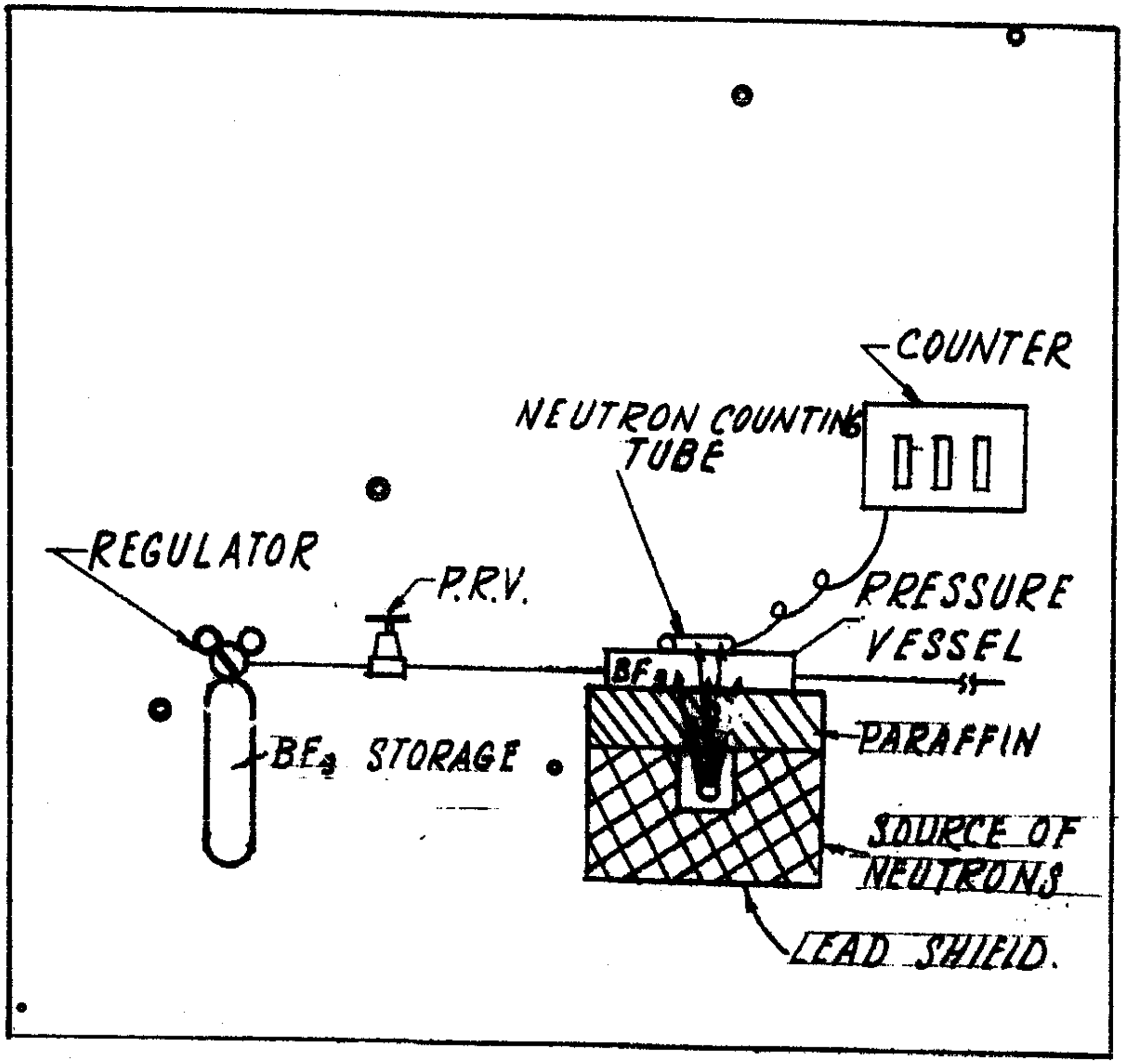

COUNTER
NEUTRON COUNTING TUBE
REGULATOR
P.R.V.
PRESSURE VESSEL
BF_3
BF_3 STORAGE
PARAFFIN
SOURCE OF NEUTRONS
LEAD SHIELD.

FIGURE 5

HW-34476

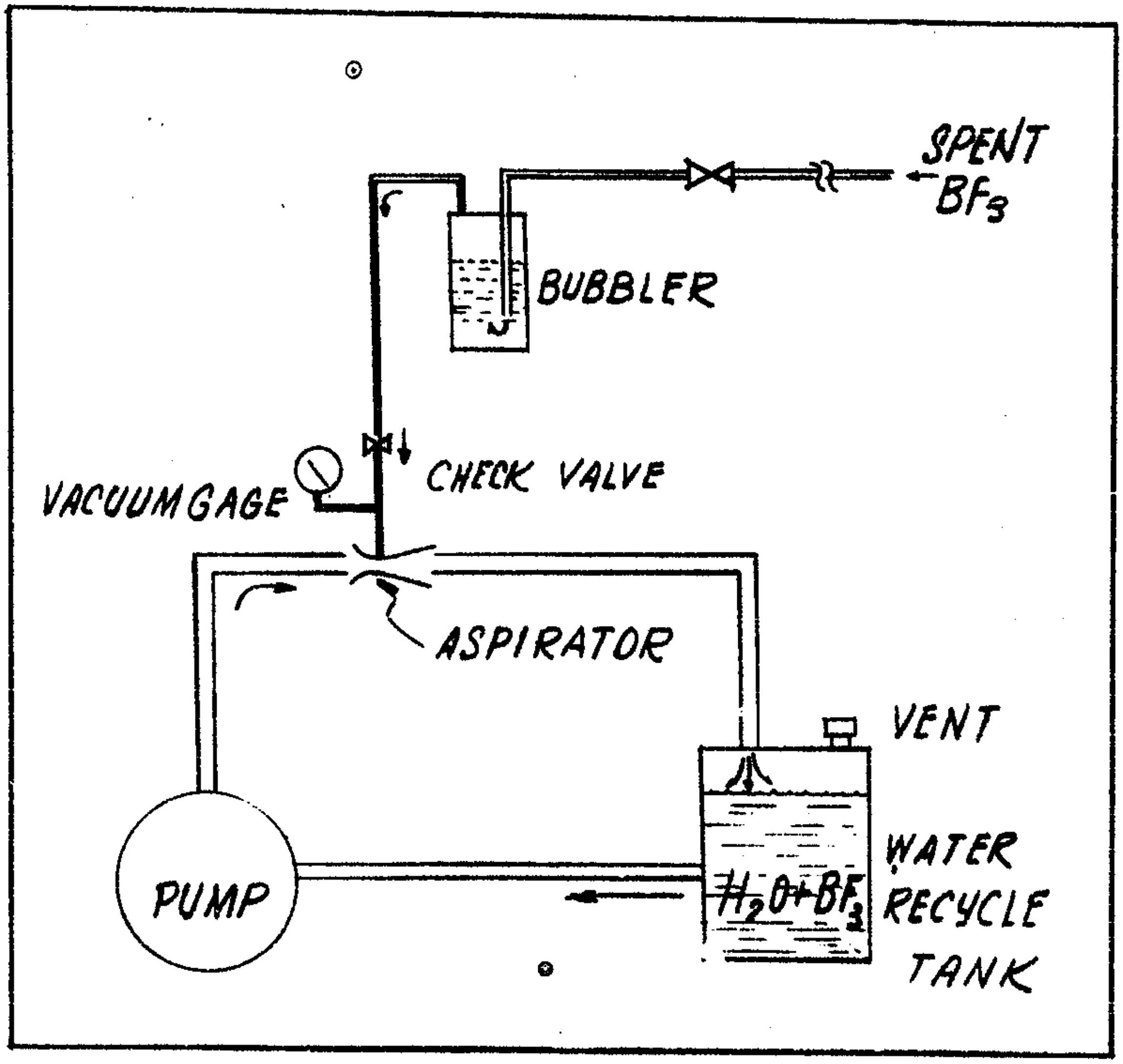


FIGURE 6