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# TWO-PHASE HYDROGEN DENSITY MEASUREMENTS BY THERMAL NEUTRON ATTENUATION

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# TWO-PHASE HYDROGEN DENSITY MEASUREMENTS

# BY THERMAL NEUTRON ATTENUATION[1]

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## SUMMARY

An experimental evaluation of two-phase hydrogen density measurements by thermal neutron attenuation was made, and the dynamic response to transient-flow situations was determined. Transmission measurements of a polyethylene "mockup" of liquid hydrogen were made, and the beam intensity from many geometric and moderator configurations was determined. The data show that good dynamic measurements can be made with the use of compact polonium-beryllium neutron sources in conjunction with an optimum moderator beam source arrangement. The equipment can be mobile and flexible and can be used to measure the full range of hydrogen mixtures from liquid to gas for a range of pipe diameters.

## INTRODUCTION

The development of space propulsion systems using liquid hydrogen requires considerable ground and flight testing. A measurement of the quality of flowing hydrogen is important during chilldown and startup of these systems. The average density is directly related to the quality and provides an indication of the two-phase condition of flowing hydrogen.

Radiation attenuation methods are widely used to measure the density or a related quantity, thickness, in the production of materials. A number of density measurements have been made in connection with boiling-water heat-transfer studies which represent a boiling flow situation similar to that of two-phase hydrogen flow. Representative measurements in connection with a boiling-water heat-transfer study are contained in reference 1. These are based on a common attenuation measurement employing an external $\gamma$-ray source and detector. This type of measurement is desirable because both source and detector are outside the fluid container and can be moved from place to place over the fluid system

While this external $\gamma$-ray attenuation technique is effective with water, it is difficult to apply with liquid hydrogen because of its low attenuation coefficient. Since $\gamma$-ray attenuation coefficients increase with atomic mass, the attenuation in pipes of practical size is large relative to that in hydro-

[1]The information contained herein was presented at the 11th Nuclear Science Symposium, Philadelphia, Pa., Oct. 28-30, 1964. It was also published in the IEEE Trans. on Nuclear Science, Feb. 1965, vol. NS-12, no. 1.

gen . On the other hand, the high-total cross section of hydrogen for neutrons and the relatively low cross section of many container materials make a neutron beam attenuation measurement appear feasible. Some of the advantages and problems of a neutron attenuation measurement are similar to those of neutron radiography (ref. 2).

A primary problem common to this measurement and to neutron radiography is the difficulty in obtaining a sufficiently intense beam of neutrons from a portable source. The intensity problem arises from the necessity of making dynamic density measurements. In the present work, an effort has been made to maximize the neutron beam intensity available from a portable source for use with a particular liquid-hydrogen measurement; and an attempt has been made to generalize the results for other applications. The measuring technique is demonstrated by using a polyethylene "mockup" of liquid hydrogen.

For completeness, another method of fluid density measurement that has been evaluated and that uses neutrons should be discussed. In this method, the hydrogen of unknown density is a part of a small neutron moderator containing a detector and fast neutron source. In this geometry the hydrogen is detected by an increase in the thermal flux in the moderator due to reduced neutron leakage from the moderator. This method has been evaluated in reference 3 for stream quality measurements. The data in reference 3 can be applied on the basis of equivalent cross sections to a liquid-hydrogen quality measurement. These data show that a 10-percent increase in detector count rate would result from increasing the hydrogen density in a 1-inch-diameter pipe from zero to that of the liquid. It is difficult to generalize the results in reference 3 to other pipe diameters, but the method would appear to be more applicable to pipe diameters larger than 1 inch in which larger count-rate changes could be realized.

## METHOD OF MEASUREMENT

The defining equation for transmission T in an attenuation measurement of density is

$$T = \frac{I}{I_o} = e^{-Nt\sigma_t} \qquad (1)$$

where $I_o$ is the beam intensity with no hydrogen present, I is the intensity with hydrogen present in the container, N is the hydrogen atom density, t is the thickness traversed by the neutrons, and $\sigma_t$ is the total microscopic hydrogen cross section, which is primarily an elastic scattering cross section.

Under ideal conditions of good geometry such that a scattered neutron cannot reach the detector, the density of hydrogen can be determined, by using a monoenergetic neutron beam, from a transmission measurement and published values of $\sigma_t$. This type of measurement requires a high-intensity source of neutrons, although some reduction in source intensity requirements can be achieved by using a polyenergetic beam. In addition, something less than good geometry can be employed at the expense of a loss in sensitivity. Under these conditions the measurement requires calibration since the effective cross sections involved are unknown.

Neutrons from radioactive sources or small accelerators are available for a portable neutron beam. The simplicity and compactness of the radioactive source is desirable if sufficient beam intensity is available; therefore, an evaluation of the beam intensity from two such sources is made herein. One source is representative of a small high-specific-activity source and the other of a large low-specific-activity source. In order to take advantage of the large neutron cross section for hydrogen in a density measurement, the average energy of the neutron beam must be less than 10 keV, and thus considerable moderation of the source neutrons is required. Although source moderation effects in homogeneous media are predictable from age diffusion theory (ref. 4) or multigroup calculations, the presence of a reentrant hole in the moderator makes quantitative estimates of beam intensity difficult; on the other hand, relatively few measurements combined with known moderating properties can give some insight into the problem. Large total fluxes are produced in the better moderators such as beryllium (Be), deuterium ($D_2O$), and graphite, but peak local fluxes are higher in hydrogenous moderators. Since available beam intensity depends on a local peak flux feeding a reentrant hole, a hydrogenous moderator is used. In order to achieve the maximum possible flux per source neutron at the center of the moderator, the radius of the moderator must be about 3 migration lengths. For Be($\alpha$,n) neutrons in water this radius is about 7 inches for a maximum central thermal flux. Since a beam of maximum intensity per source neutron is required, it will include many thermal neutrons. It is convenient to measure the thermal energy group, and an estimate of the additional contribution due to higher energy neutrons can be made. A shielding problem arises if neutrons many decades above thermal are used.

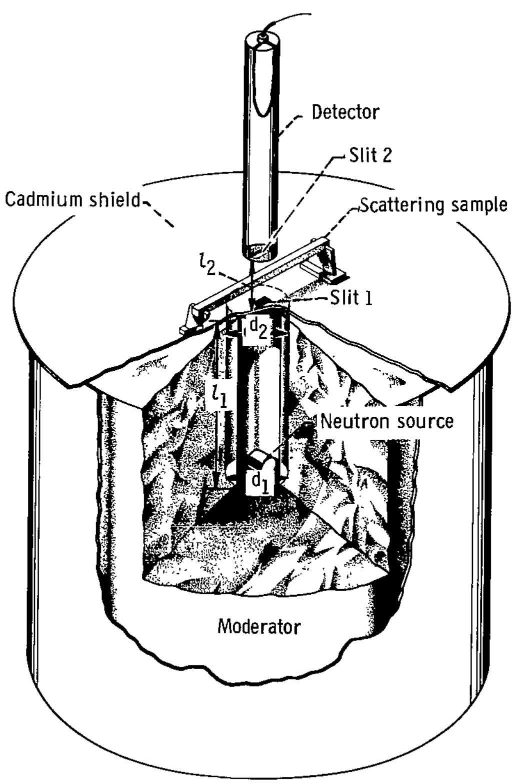

Figure 1. - Neutron beam source.

## Beam Intensity Measurements

The geometrical arrangement of interest is shown in figure 1, in which a reentrant hole is used to obtain a beam from a hydrogenous moderator containing a Be($\alpha$,n) neutron source. A cadmium shield covers the moderator to define a thermal neutron beam. The reentrant hole dimensions are $d_1$, $d_2$, and $l_1$, and the beam defining slits are slits 1 and 2. Three different neutron detectors were used in the measurements, a boron fluoride ($BF_3$) proportional counter, a glass scintillator, and a lithium iodide (LiI) crystal. These detectors are described in detail in connection with the beam intensity data. Standard pulse-counting instruments were used, each consisting of a high-voltage power supply, a linear amplifier, a single-channel analyzer, a scaler, a count-rate meter, and a recorder. Several thermal neutron-beam intensity measurements were made in sources having the geometry of figure 1 with variations in the dimensions $l_1$, $l_2$, $d_1$, and $d_2$ and in slits 1 and 2.

For small values of $l_2$, beam collimation depends on the reentrant hole depth $l_1$ and consequently the total moderator size; it may be possible to reduce $l_1$ and increase the beam intensity even though the central flux is lower in the smaller moderator. Since beam intensity also depends on the hole diameter, this parameter must be evaluated. In addition a heterogeneous mixture of moderator materials may be advantageous.

In the first series of measurements a 1-inch-diameter by 10-inch-long cadmium shielded $B^{10}F_3$ neutron detector was used. The data are shown in table I. In all cases, the Po($\alpha$,n)Be (polonium-beryllium) source was held at the surface of the water at the bottom of the hole, and it was found that any water over the source decreased the count rate. For the Pu($\alpha$,n)Be (plutonium-beryllium) source, it was found that 1 inch of water above the source in the hole resulted in the highest count rate at all hole depths. Covering the source with beryllium should reduce the number of neutrons in the beam coming directly from the Be($\alpha$,n) source partially by the Be(n,$\alpha$n) reaction and without significantly reducing the thermal neutron leakage from the hole bottom. Some reduction in uncollided neutrons can be accomplished by making the larger reentrant holes conical in shape.

Any reduction in source energy neutrons in the beam due to the beryllium shell or the shaped hole would not be proportional to the change in the cadmium-covered count rates shown in table I because counting efficiency changes with beam neutron energy spectrum and, more important, because the cadmium-covered detector primarily measures leakage from the entire surface of the moderator. All cadmium ratios of the counting data in table I are low for this reason. While a good boron shield was not used because of the difficulty in changing its shape, measurements of cadmium-covered count rates off the axis of the reentrant holes indicate a possible increase in cadmium ratios of the order of a factor of 10 by using a thick boron shield.

The 15-inch-diameter-moderator data include some measurements using a 1- by 1/4- by 1/8-inch $Li^6$ glass scintillator. The count rates shown are the integral of the thermal neutron peak counts. Both this detector and the $Li^6I$ detector used with the $9\frac{3}{4}$-inch-diameter moderator are essentially black to thermal neutrons, and the detection efficiency should be 90 percent or better. In these measurements $l_2$ is varied for the two reentrant hole and moderator configurations. The data show a rapid change in intensity at distances less than about 4 inches from the moderator which indicates that some of the beam originates from the sides of the reentrant hole.

Some additional beam intensity measurements were made by using a 19-inch-diameter paraffin moderator. These data are plotted in figure 2 (p.6). The count rates shown were obtained with the $Li^6I$ crystal, and slit 1 had a 3/4-inch diameter for the five configurations shown. These data can be compared with the $9\frac{3}{4}$-inch-moderator data in table I and show a small increase in intensity with decreased moderator size and reentrant hole depth.

A comparison of the count rates obtained in figure 2 shows that the larger hole results in higher count rates at distances less than 4 inches from the

TABLE I. - BEAM INTENSITY MEASUREMENTS

[Source intensity, $10^7$ neutrons/sec.]

| Moderator |  | Neutron source |  |  | Neutron detector | Reentrant hole dimension, in. |  |  |  |  |  | Detector count rate, counts/sec |  |  |  |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Type | Dimensions | Type | Radiation, Ci | Dimensions |  | $l_1$ | $l_2$ | $d_1$ | $d_2$ | Slit 1 | Slit 2 | Total | Cadmium | Thermal | Thermal, sq cm of detector |
| Water tank | 6-ft diam. by 5-ft high | PoBe | 10 | 0.7-in. diam. by 0.7-in. high | $BF_3$ proportional counter | $16\frac{1}{2}$ | $1\frac{1}{4}$ | $1\frac{1}{2}$ | $1\frac{1}{2}$ | $\frac{3}{4}$ by $\frac{1}{4}$ | $\frac{3}{4}$ by $\frac{1}{4}$ | 29.0 | 4.5 | 24.5 | 20.2 |
|  |  |  |  |  |  | 13 | ↓ | ↓ | ↓ | ↓ | ↓ | 47.0 | 9.6 | 37.4 | 30.9 |
|  |  |  |  |  |  | $8\frac{1}{4}$ | ↓ | ↓ | ↓ | ↓ | ↓ | 93.0 | 24 | 69 | 57.0 |
|  |  |  |  |  |  | 5 | ↓ | ↓ | ↓ | ↓ | ↓ | 125 | 42 | 83 | 68.6 |
|  |  |  |  |  |  | 3 | ↓ | ↓ | ↓ | ↓ | ↓ | 140 | 67 | 73 | 60.4 |
|  |  |  |  |  |  | 2 | ↓ | ↓ | ↓ | ↓ | ↓ | 151 | 84 | 69 | 57.0 |
|  |  | PuBe | 5 | 1.31-in. diam by 2.72-in. high | $BF_3$ proportional counter | $10\frac{3}{4}$ | $1\frac{1}{4}$ | $1\frac{1}{2}$ | $1\frac{1}{2}$ | $\frac{3}{4}$ by $\frac{1}{4}$ | $\frac{3}{4}$ by $\frac{1}{4}$ | 38 | 6.9 | 31.1 | 25.7 |
|  |  |  |  |  |  | $7\frac{1}{2}$ | ↓ | ↓ | ↓ | ↓ | ↓ | 44 | 10 | 34 | 28.1 |
|  |  |  |  |  |  | 5 | ↓ | ↓ | ↓ | ↓ | ↓ | 64 | 18 | 46 | 38.0 |
| Paraffin cylinder | 15-in. diam. by 15-in. high | PoBe | 10 | $1\frac{3}{4}$-in. diam. Be sphere | $BF_3$ proportional counter | $7\frac{1}{2}$ | $1\frac{1}{4}$ | $\frac{3}{4}$ | $\frac{3}{4}$ | $\frac{3}{4}$ by $\frac{1}{4}$ | $\frac{3}{4}$ by $\frac{1}{4}$ | 52 | 8.7 | 43.3 | 35.8 |
|  |  |  |  |  |  | ↓ | ↓ | $1\frac{3}{8}$ | $1\frac{3}{8}$ | ↓ | ↓ | 107 | 24 | 83 | 68.6 |
|  |  |  |  |  |  | ↓ | ↓ | $2\frac{1}{2}$ | $2\frac{1}{2}$ | ↓ | ↓ | 116 | 33 | 83 | 68.6 |
|  |  |  |  |  |  | ↓ | ↓ | $1\frac{3}{4}$ | $1\frac{3}{4}$ | ↓ | ↓ | 133 | 29 | 104 | 86.0 |
|  |  |  |  |  |  | ↓ | ↓ | $1\frac{3}{4}$ | $\frac{3}{4}$ | ↓ | ↓ | 135 | 24 | 111 | 91.8 |
|  |  |  |  | $3\frac{1}{2}$-in. diam. Be sphere | $Li^6$ glass scintillator | $7\frac{1}{2}$ | $2\frac{7}{8}$ | $3\frac{1}{2}$ | $\frac{1}{4}$ by $1\frac{3}{8}$ | $\frac{1}{4}$ by $1\frac{3}{8}$ | 1 by $2\frac{1}{4}$ | 2690 | 1160 | 1530 | 105 |
|  |  |  |  |  |  | ↓ | $4\frac{1}{4}$ | ↓ | ↓ | ↓ | ↓ | 1730 | 840 | 890 | 61.3 |
|  |  |  |  |  |  | ↓ | 6 | ↓ | ↓ | ↓ | ↓ | 1180 | 670 | 510 | 35.2 |
| Paraffin cylinder | $9\frac{3}{4}$-in. diam. by $9\frac{3}{4}$-in. high | PoBe | 10 | $1\frac{3}{4}$-in. diam. Be sphere | $Li^6I$ crystal | $4\frac{3}{4}$ | $1\frac{3}{8}$ | $1\frac{3}{4}$ | $\frac{3}{4}$ diam. | $\frac{3}{4}$ diam. | $1\frac{1}{2}$ diam. | 5700 | 1850 | 3850 | 338 |
|  |  |  |  |  |  | ↓ | $2\frac{5}{8}$ | ↓ | ↓ | ↓ | ↓ | 3260 | 1320 | 1940 | 170 |
|  |  |  |  |  |  | ↓ | $4\frac{3}{8}$ | ↓ | ↓ | ↓ | ↓ | 1830 | 870 | 960 | 84 |
|  |  |  |  |  |  | ↓ | $6\frac{1}{2}$ | ↓ | ↓ | ↓ | ↓ | 1140 | 628 | 512 | 45 |

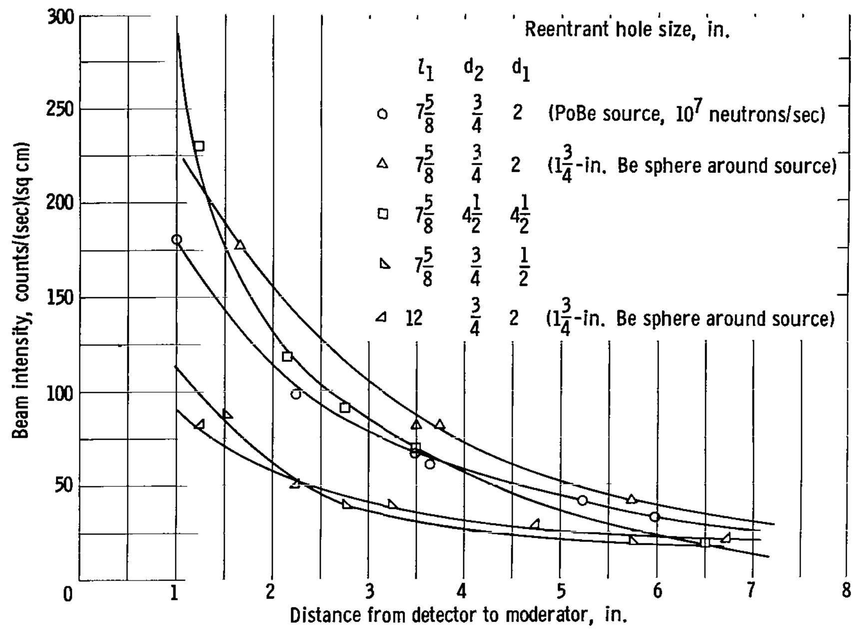

Figure 2. - Beam intensity from paraffin moderator and Be(α, n) neutron source.

moderator. At larger distances in which the beam is better collimated the smaller hole appears better. This is consistent with the results obtained in reference 5, in which the smallest reentrant hole diameters resulted in the greatest intensity for a well-collimated beam.

Polyethylene transmission measurements. - Transmission measurements were made by using two of the moderator configurations shown in table I. The configuration with a $\frac{1}{4}$-by $1\frac{3}{8}$-inch beam defining slit 1 was used with a range of polyethylene thickness and width corresponding in total cross section (refs. 6 and 7) to a range of hydrogen densities in a 1/4- and a 1/2-inch-diameter pipe. In these measurements, the glass scintillator was positioned $2\frac{1}{2}$ inches from the moderator. In the second series of measurements the $9\frac{3}{4}$-inch-diameter moderator was used with the LiI crystal 4 inches from the moderator. The polyethylene thicknesses used were a "mockup" of hydrogen of various densities in 1- and 2-inch-diameter pipes. The data are shown in figure 3. The measured transmissions are higher than the calculated exponential, particularly for the higher densities, because of inscattering in the poor geometry of the measurement. Increasing the detector-to-moderator distance not only improves the geometry and reduces the transmission but also reduces the detector count rate.

Optimization of the attenuation method therefore involves maximizing both count rate and the slope of the transmission curve. The beam sources and detectors used in this study are near optimum for dimensions of slit 1 used. In the case of the 1/4-inch-diameter pipe measurements, slit 1 could be made longer with an improvement in intensity and transmission but with an increase in the length of pipe over which the hydrogen density is averaged. For the larger

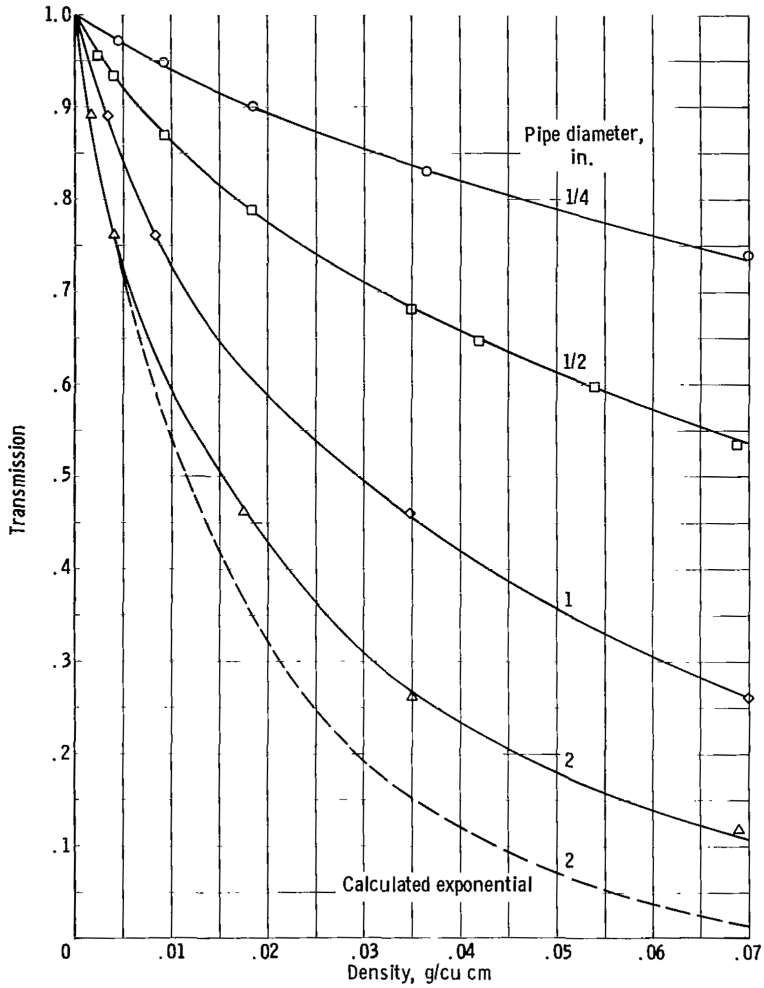


Figure 3. - Thermal neutron beam attenuation by polyethylene.

diameter pipes, the size of slit 1 could be significantly increased over that used in the $9\frac{3}{4}$-inch-diameter moderator.

Dynamic response calculations. - An essential characteristic of a two-phase hydrogen-density measurement is its ability to respond dynamically to a transient-flow situation. The ability to follow transients with a reasonably short time constant is dependent on having an intense neutron source, an effectively optimized moderator arranged about the source to produce a neutron beam, and finally a neutron detector of high efficiency. The dynamic response has been calculated for the "mockup" measurements shown in figure 3 by using the relation $SD = 1/\sqrt{2RCA}$, where SD is the standard deviation of a single observation of the recorder or count-rate meter, RC is the time constant, and A is the detector count rate. The computed standard deviation in count rate was converted to standard deviation in density by using the slope of the experimental transmission curves in figure 3. The calculation for three pipe

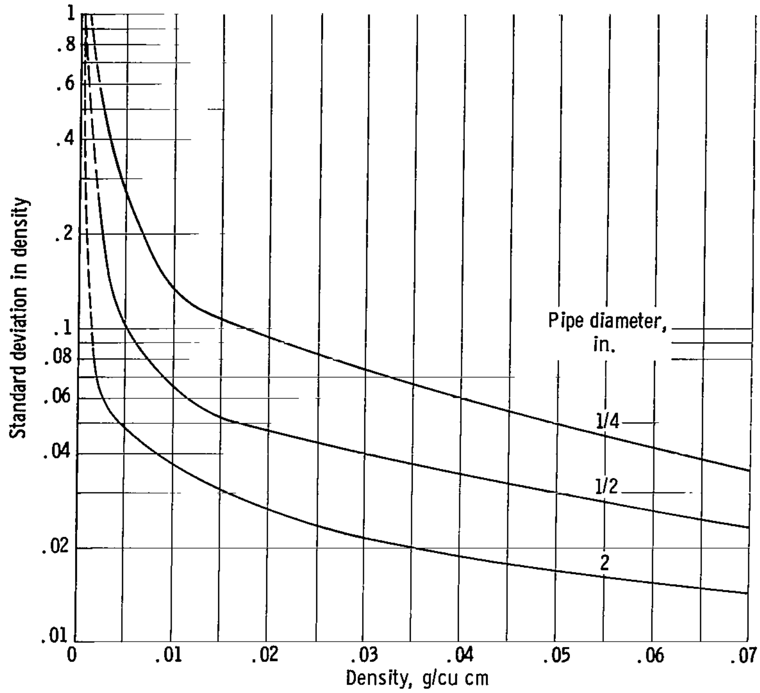


Figure 4. - Standard deviation in density for single observation of recorder. Time constant, 0.5 second; source intensity, $10^8$ neutrons per second.

diameters is shown in figure 4, in which RC is 0.5 second and a $10^3$-neutron-per-second source is used.

## SUMMARY OF RESULTS

Available beam intensities from a hydrogenous moderator containing a beryllium ($\alpha$,n) neutron source have been measured for a number of reentrant hole configurations. The measurements show a lack of a strong dependence of beam intensity on reentrant hole geometry due to close compensation of beam collimation and moderator leakage for poorly collimated beams. A strong dependence of beam intensity on distance from the moderator was observed. A polonium-beryllium source surrounded with beryllium produced a somewhat more intense beam than a plutonium-beryllium source at all distances.

A moderator configuration employing a large conical reentrant hole containing beryllium and a polonium-beryllium source resulted in the highest intensity of a 1/4-inch-wide beam at distances less than 4 inches from the moderator surface. Obtainable detector count rates permit useful dynamic measurements to be made. Expected standard durations for 1/4- to 2-inch pipe diameters and for a 0.5-second time constant are obtained by using experimental transmission and intensity data.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, February 11, 1965.

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