Measurement of Concentration of Tungsten Suspensions and Density of Liquid Sodium by Gamma Ray Absorption

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Gamma ray absorption techniques have been applied to measure the density of various media, and the theory of selecting the appropriate energy gamma ray to give optimum sensitivity of density determination for a homogeneous system is outlined. A gauge to determine the concentration of tungsten powder suspended in water in a concentration range of 0 to 8 gm. tungsten/c.c. suspension, has been developed. In this apparatus the mean tungsten concentration, 0.08 gm./c.c., can be measured to an accuracy of approximately ±0.0015gm./c.c. In the case of liquid sodium contained in a stainless steel vessel, preliminary experiments indicate that the density may be measured to an accuracy of ±0.01gm./c.c., and that 0.01gm./c.c. transient changes in density occurring in a few seconds can be resolved.

INTRODUCTION

Gamma ray absorption has been used successfully to measure the density of solids and liquids and the concentration of metal salts or solid suspensions in liquids. The technique has been used to determine the density of uranium pulp and uranyl sulphate solutions (Seymour 1957), and the concentration of heavy metal salts in solution (Thurnau 1957). Bartholomew (1957) has measured solids concentration in fluidised systems, and determined density patterns in transverse sections of tube. Hitchon (1956) has used radiometric techniques to detect variations in concentration of aqueous thoria slurries.

The authors have developed a gauge to determine the concentration of micron-size tungsten particles suspended in water. The tungsten, of mean concentration approximately 0·08 gm. tungsten/c.c. suspension, is pumped around a loop in a pipe of internal diameter one inch. At low pumping speeds, concentrations over a transverse section of pipe may vary over the range of 0 to 8 gm. tungsten/c.c. suspension, the higher concentrations occurring in settled beds of tungsten. It was desired to measure accurately the absolute concentration and the concentration gradient across a transverse section of pipe.

A gauge to measure the density of liquid and boiling sodium contained in a stainless steel vessel of internal diameter 5 in. and wall thickness 0·125 in. has been considered. A preliminary experiment designed to determine the accuracy to which the density may be found and the minimum transient changes which may be resolved in a few seconds, is briefly described.

The measurement of density by gamma rays is based on the principle that gamma rays, when traversing a medium, are partially absorbed by the medium, and the extent of absorption depends on the density of the medium. The theory of gamma ray absorption as applied to density gauges is outlined, and experimental work associated with the tungsten concentration and sodium density gauges is described.

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THEORY

Selection of gamma ray energy

The basic theory of gamma ray density gauges has been described by various authors (Berman 1951; Bogachev 1955; Cameron 1957), and is briefly outlined here.

If a parallel beam of monoenergetic gamma rays traverses a homogeneous medium of density ρ and thickness x, the emergent intensity I is related to the incident intensity I_0 by the equation

$$I = I_0 e^{-\mu_m \rho x}, \quad \ldots\ldots\ldots \quad (1)$$

where μ_m is the mass absorption coefficient of the gamma rays in the medium. μ_m is a function of the energy of the gamma rays and the atomic number of the absorber.

The fractional change of intensity, $\delta I/I$, caused by small changes in density $\delta\rho$ and in thickness δx, is calculated by differentiating equation (1).

$$\frac{\delta I}{I} = -\mu_m . \delta(\rho x), \quad \ldots\ldots\ldots\ldots\ldots \quad (2)$$

whence

$$\delta\rho = -\frac{1}{\mu_m x} \cdot \frac{\delta I}{I}, \text{ for constant } x \quad \ldots\ldots \quad (3)$$

and

$$\delta x = -\frac{1}{\mu_m \rho} \cdot \frac{\delta I}{I}, \text{ for constant } \rho. \quad \ldots\ldots \quad (4)$$

Equation (2) indicates that fractional changes in intensity of radiation are caused by variations in ρx, i.e. weight/unit area of the medium. In most density gauges, x is constant, and fractional changes in intensity of radiation are caused by density variations alone, as in equation (3).

The accuracy to which density may be determined depends on the minimum value of $\delta I/I$ which may be resolved. Uncertainties in measurement of $\delta I/I$ are caused by fluctuations due to the random emission of gamma rays from a radioactive source, and by limitations in the accuracy and stability of the radiation detection equipment.

The statistical uncertainty in measuring the emission rate from a radioactive source is discussed by Whitehouse (1953, p. 208). If $\bar{N}$ is the mean number of gamma rays counted by a detector in a

time t (i.e. the " true " number of counts), and N is the number counted over one period t, then there is a probability of approximately 68 per cent. that N lies within $\pm\sqrt{\bar{N}}$ of $\bar{N}$. If ε is the efficiency of detection of gamma rays by a radiation detector, the number N of gamma rays detected in a time t, is given by

$$N=\varepsilon It, \quad \ldots\ldots\ldots\ldots (5)$$

and the minimum resolvable value of $\frac{\delta I}{\bar{I}}$ corresponding to the 68 per cent. probability then becomes :

$$\frac{\delta I}{\bar{I}}=\frac{1}{\sqrt{\varepsilon It}}. \quad \ldots\ldots\ldots\ldots (6)$$

For any particular system there is a gamma ray energy which gives optimum sensitivity of density determination. In practice, the activity of the radioactive source is limited, and the selection of the gamma ray energy is a compromise between high absorption and the necessity of having sufficient gamma rays passing through the system for accurate measurement. Assuming that uncertainties in $\delta I/I$ are caused solely by the statistical fluctuations in counting gamma rays from the radioactive source, the gamma ray energy giving optimum sensitivity of density determination is calculated as follows. The minimum resolvable value of $\delta I/I$ is, from (6),

$$\delta I/I=\frac{1}{\sqrt{\varepsilon It}}$$

$$=\frac{1}{\sqrt{\varepsilon I_0 te^{-\mu_m\rho x}}}, \quad \ldots\ldots (7)$$

Substituting (7) in (2), the relative error introduced in density determination corresponding to a minimum resolvable value of $\frac{\delta I}{\bar{I}}$ is

$$\frac{\delta(\rho x)}{\rho x}=\frac{e^{\frac{\mu_m\rho x}{2}}}{\mu_m\rho x\sqrt{\varepsilon I_0 t}} \quad \ldots\ldots\ldots (8)$$

TABLE 1.—GAMMA RAY SOURCES USED FOR DENSITY GAUGES.

Data from A.E.R.E. Isotopes Catalogue (1957).

ISOTOPE	HALF-LIFE	GAMMA ENERGIES (MeV)
Americium 241	470 years	0.060 + X-rays
Thulium 170	127 days	0.084 (3%) 0.052 (8%) and Bremsstrahlung
Selenium 75	127 days	0.121 0.136 0.265 + others 0.280 0.402
Iridium 192	74.4 days	0.296 (19% *) 0.308(18% *) + others 0.316 (48% *) 0.468 (15% *)
Caesium 137	30.0 years	0.662 (82%)
Cobalt 60	5.52 years	1.17 (100%) 1.33 (100%)

*Relative abundance only (Hollander, 1953).

This relative error is a minimum when $\mu_m=\frac{2}{\rho x}$, and so the optimum gamma ray energy for any particular system may be found.

A limited number of radio-isotopes are suitable for use in density gauges and are listed in Table 1. Values of μ_m for well-collimated beams of gamma rays are given by Moteff (1954) and Chapell (1956). For beams not strongly collimated, the values of μ_m may be considerably lower than those given.

Summary

The choice of radioactive isotopes for the tungsten concentration and sodium density gauges was based on calculations from the theory given. Tungsten suspended in water is not a homogeneous system. However, to a first-order approximation, fractional changes in gamma ray intensity are caused by variation in tungsten concentration alone, because :

(i) large variations in tungsten concentration result in small changes in water content per c.c. of suspension,

(ii) the absorption of gamma rays per gram of tungsten is larger than that per gram of water.

Thus, as an approximation, fractional changes in absorption are dependent mainly on changes in tungsten concentration.

The isotopes chosen for use in the tungsten concentration and sodium density gauges are given in Table 2.

TABLE 2.—ISOTOPES USED IN TUNGSTEN AND SODIUM GAUGES.

System	Isotope	Conc. or density range
tungsten in water	iridium 192	0-8gm. tungsten /c.c. of suspension
	thulium 170	0-0.2gm. tungsten /c.c. of suspension
sodium	iridium 192 (or caesium 137)	whole density range (approx. 1-0.75gm./c.c.)

TUNGSTEN CONCENTRATION GAUGE

Discussion

In order to determine tungsten concentrations accurately in the range 0 to 8 gm. tungsten/c.c. of suspension, it was necessary to use two radio-isotopes, thulium 170 and iridium 192. Sensitivity is gained at low tungsten concentrations by using thulium 170 because its gamma rays are strongly absorbed by tungsten, but relatively weakly absorbed by water. Iridium 192 was used for high tungsten concentrations, because thulium 170 gamma rays are not sufficiently penetrating.

Apparatus

A diagram of the apparatus is shown in Figure 2.

A tube of square cross-section was chosen so that, by traversing the tube with a well-collimated beam of gamma rays, the concentration of tungsten could be determined with equal sensitivity in each part of the tube. The walls of the tube were made of " Perspex " to minimize the preferential absorption of the low energy gamma ray component of the thulium 170 spectrum.

The lead source container, collimation block and scintillation head were rigidly mounted on a 1 in. thick steel table. The table was mounted on a system of two wedges, and could be moved vertically by sliding one wedge along the other by rotating a threaded drive rod. The height of the table, relative to the tube, could be determined to better than $\pm 0 \cdot 016$ in. The gamma rays were collimated by the lead block to a circle of aperture approximately 0·063 in. Thus, by traversing the tube with the collimated beam of gamma rays, it was possible to measure the tungsten concentration gradient across the tube.

Sources of 125 millicuries of iridium 192 (cylinder of dimensions 2 mm. diameter by 2 mm. height) and 7 c. of thulium 170 (dimensions 3·2 mm. diameter by 3·2 mm. height) were used in the experiment. The gamma rays were detected by a scintillation counter, with pulse counting timed in each case over 100-second intervals. Counting rates were corrected for dead time (Whitehouse 1953, p. 218). To check the stability of the electronic apparatus, statistical fluctuations in the counting of iridium 192 and thulium 170 gamma rays were analysed by the method described by Cook (1952, p. 286). The results indicate that there is a good probability that the counts can be reproduced with a standard deviation of less than $\pm 0 \cdot 25$ per cent.

Calibration using iridium 192

Attempts were made to calibrate the gauge by suspending tungsten powder in water by mechanical stirring. However, it was found too difficult to produce stirred uniform suspensions within the limits of accuracy necessary. This inability to provide standard concentration suspensions is not a fundamental limitation in the method used to determine the concentrations of suspensions pumped around the loop. In this application it is required to determine the unknown concentration gradients of suspension present in the tube under varying rates of flow.

The gauge was calibrated by measuring the relative absorption of iridium 192 gamma rays by a series of tungsten discs, placed external to the tube of water, and applying a small, experimentally determined, correction for the absorption by the water which the equivalent amount of tungsten in suspension would displace.

Two experiments were carried out to see if the absorption was dependent on the position of the absorber in the collimated beam. In one experiment the tube filled with water was used as absorber, in the other the absorber consisted of five tungsten discs. The relative counting rates with these absorbers separately placed in positions (A) and (B), Figure 2, were compared. The counting rates with each absorber at (A) and (B) agreed within the limits of experimental error ($\pm 0 \cdot 35$ per cent.) indicating that even for such large changes in positioning of the tungsten or water, the relative counting rates are the same.

Each tungsten disc was approximately 0·04 in. thick, and one was spectrographically analysed, showing less than 1 per cent. impurity.

The uniformity of each disc was checked by radiometric methods. The relative counting rate of gamma rays was compared for the sum of 0 to 11 discs, placed external to the tube, in each case for both the tube empty, and filled with water.

The following corrections were made:

(i) The tungsten discs were placed external to the tube filled with water, so allowance was made for the absorption of the water which an equivalent amount of tungsten in suspension would displace. The relative absorption for tube empty and filled with water was determined for various total thicknesses of tungsten discs, and from this the mass absorption coefficient of water calculated and so corrections applied. The mass absorption coefficient varied slightly for different tungsten absorber thicknesses, because of the selective filtering out of the low energy gamma rays compared with the high energy ones.

(ii) In the calibration experiments, the tube used differed from that in the loop experiments. The internal dimension at the centre of the tubes, through which the gamma rays pass, are:

Calibration tube—$1 \cdot 009 \pm 0 \cdot 001$ in.
Tungsten loop tube—$0 \cdot 999 \pm 0 \cdot 001$ in.

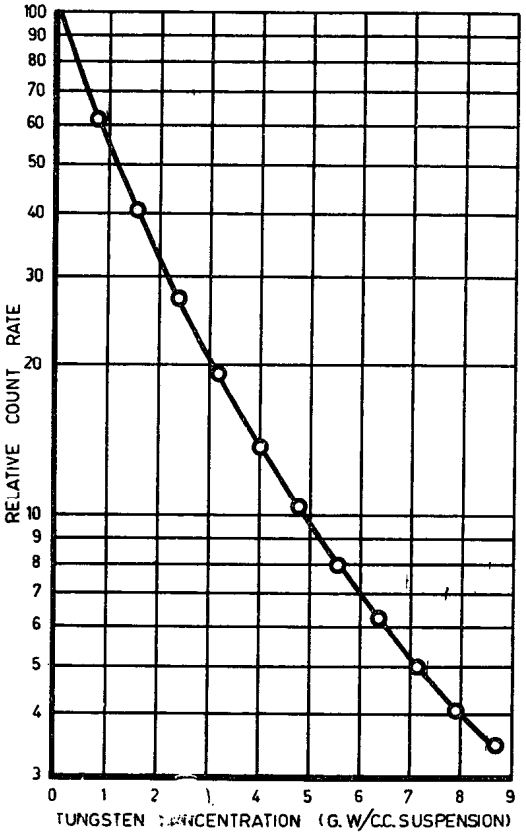

FIGURE 1:—Corrected calibration curve for absorption of iridium 192 gamma rays in tungsten suspensions.

Concentrations of equivalent amounts of tungsten in suspension as in discs were calculated for an absorber thickness of 0·999 in. No correction was made for the slight difference in absorber

thickness of water in the two tubes. The error due to neglecting this is less than 0·3 per cent. of the relative intensity.

Figure 1 shows the relative absorption of iridium 192 gamma rays by different concentrations of tungsten suspension. The standard deviation is approximately ±0·6 per cent. at the highest tungsten concentration.

Calibration using thulium 170

To calibrate the gauge for suspensions of low tungsten concentration, the relative absorption of thulium 170 gamma rays was found for sodium tungstate solutions of various concentrations. Corrections were applied to these values to allow for the absorption by the different water contents per c.c. of suspension and solution, and for absorption by the sodium and oxygen of the tungstate. Thus, relative absorption by equivalent tungsten concentration suspensions was determined. These experimentally found corrections are small because of the high absorption of thulium 170 gamma rays by tungsten, compared with that by low atomic number elements such as hydrogen, oxygen and sodium.

A stock solution of A.R. sodium tungstate in water was analysed for tungsten concentration, and its mean density experimentally determined. Solutions of various concentrations were made by diluting a known volume of stock solution with water and making up to a given volume, in each case measuring the volume of water added. In this way, both the water and tungsten content per c.c. of suspension were accurately measured for each solution. The relative absorption of gamma rays by each of these solutions was determined.

Corrections for different weights per c.c. of water in solution and suspension with equal tungsten concentrations were determined by a method similar to correction (i) for iridium 192. The mass absorption coefficient of water for thulium 170 gamma rays was found to vary slightly with tungsten concentration.

The low atomic number elements sodium, oxygen and nitrogen have approximately the same mass absorption coefficient for thulium 170 gamma rays. Allowance for the absorption by sodium and oxygen in the tungstate solution was made by finding the absorption of thulium 170 gamma rays by sodium nitrate solutions of different concentrations. The water content per c.c. of solution varied for different sodium nitrate concentrations, and corrections were also made for this, using the value of μ_m for water already determined. The mass absorption coefficient for water was found to be slightly higher than that of sodium nitrate. However, because the corrections for water, sodium, and oxygen were small, to a good approximation the same value of μ_m could be taken for water, sodium, and oxygen corrections over the whole tungsten concentration range. The largest correction applied to the relative intensity was 3 per cent., and the error in relative counting rate introduced by assuming this approximation is less than ±0·35 per cent.

Allowance was also made for the difference of internal dimension of calibration and tungsten loop tubes. The absorption of gamma rays by tungsten is dependent on changes in the product of absorber thickness and concentration of tungsten. Thus each concentration was multiplied by the ratio 1·010, which is the ratio of the internal dimensions of calibration tube and tungsten loop tube.

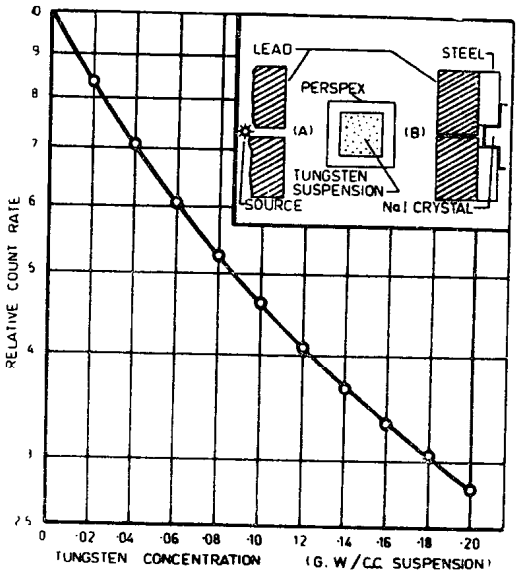

FIGURE 2:—Corrected calibration curve for absorption of thulium 170 gamma rays in tungsten suspensions.

Figure 2 shows the corrected values of relative absorption of thulium 170 gamma rays versus concentration of tungsten in suspension.

At concentrations of approximately 0·08 gm. tungsten/c.c. suspension, the total error in relative counting rate caused by statistical fluctuations and uncertainties in corrections is less than ±1 per cent. A ±1 per cent. change in relative count rate is approximately equivalent to a concentration change of ±0·0015 gm. tungsten/c.c. suspension at this tungsten concentration (i.e. 0·08 gm./c.c.) and this indicates the accuracy to which the concentrations may be determined. Each tungsten concentration across the square tube may be determined to this accuracy if allowance if made for slight changes in the internal dimension across the square tube.

SODIUM DENSITY GAUGE

A preliminary experiment was carried out to assess the approximate sensitivity of the sodium density gauge by finding the fractional changes of intensity of iridium 192 gamma rays caused by a known density change. Water and kerosene were used to simulate sodium at two different densities because the values of μ_m for iridium 192 gamma rays in these liquids are, to a first order approximation, equal.

A 2 mm. diameter beam of gamma rays from an 800 mc. iridium 192 source passed through a diameter of the stainless steel vessel containing liquid, and was detected, in two separate experiments, by geiger and scintillation counters. The results of the experiments are given in Table 3.

TABLE 3.—FRACTIONAL CHANGES $\delta I/I$ OF TRANSMITTED RADIATION FOR KNOWN DENSITY VARIATIONS.

Radiation detector	Liquid	Density (gm./c.c.)	Transmitted radiation I	$\delta I/I$	
				Experimental [(c)-(d)/mean (c) & (d)]	Calculated [from equation (9)]
Scintillation counter	Kerosene	0.82 (a)	Counts/sec. 8000±80 (c)	0.177±0.014	0.19
	Water (at 60°C)	0.98 (b)	6700±67 (d)		
Geiger counter	Kerosene	0.81 (a)	Counts in 100 sec. 9779±99 (c)	0.224±0.013	0.23
	Water	1.00 (b)	7812±88 (d)		

The calculated values $\delta I/I$ in Table 3 were found as follows: The energies of the more abundant gamma rays from iridium 192 are grouped about 0·31 (85 per cent.) and 0·47 (15 per cent.) MeV (Table 1), and the mass absorption coefficients in sodium of these energy groups of gamma rays are 0·100 and 0·088 sq. cm./gm. respectively (Moteff 1954). Values of μ_m for carbon and oxygen are taken to be the same as for sodium, and for hydrogen twice that for sodium.

Equation (1) now becomes

$$I=I_{01}\, e^{-\mu_{m_1}\rho x}+I_{02}\, e^{-\mu_{m_2}\rho x} \quad \ldots\ (9)$$

where the subscripts 1 and 2 refer to gamma rays of energy 0·31 and 0·47 MeV respectively. The values of $\delta I/I$ for known density changes [(a)–(b), in Table 3] are calculated from this equation.

The calculated and experimental results in Table 3 agree well, and this indicates that the theory outlined may be applied to assess the approximate sensitivity of the sodium density gauge.

The sensitivity of the sodium density gauge may be estimated as follows: The long term stability of the scintillation detector and associated electronic apparatus is approximately 1 per cent. Thus $\delta I/I \simeq 0{\cdot}01$, because uncertainties in counting due to random emission of gamma rays may be made small compared to this. Thus from equation (3) the sodium density may be found to better than $\pm 0{\cdot}01$ gm./c.c. For transient density changes over a few seconds, $\delta I/I$ will be determined by statistical fluctuations alone, and thus will depend on the counting rate of the detector. For a counting rate of 10^6/sec., transients of approximately 0·01 gm./c.c. can be resolved in a few seconds.

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