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Publication Date
1991

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April 1991
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THE BETA GAUGE METHOD FOR MASS MEASUREMENT

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This work was supported by the Director, Office of Energy Research, Office of Health and Environmental Research of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098 and through Interagency Agreement No. EPA-IAG-DW89932573-01-0 with the U.S. Environmental Protection Agency.
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INTRODUCTION

The beta gauge method of mass determination depends upon the near exponential decrease in the number of beta particles transmitted by a thin sample as the areal density is increased. The beta particles are emitted as a continuum energy distribution by a radioisotope source and their intensity is measured with a suitable electron counter. The method has the advantages of instrumental simplicity and ease of automation for large scale applications. The dynamic range of sensitivity is well matched to the mass range normally of interest in aerosol monitoring in which thin membrane filters are the substrates. However, since beta attenuation does not involve a direct gravimetric determination of the mass deposited, a detailed understanding of parameters which affect the measurements is necessary in order to ensure optimal instrumental implementation and correct interpretation of results.

DESCRIPTION OF METHOD

Figure 15-1 is a schematic of a basic beta gauge instrument consisting of the radioactive source, detector and sample. The particular instrument illustrated in the figure employs a $^{147}$Pm source and a semiconductor beta particle detector. However, the basic principles of the method are the same for other combinations of source and detector. A measurement consists of determining the counting rate in a continuous beta particle spectrum emitted by the radioisotopic source and transmitted through the sample. Under the proper experimental conditions, the observed rate ($I$) is related to the sample mass through the relationship (Evans 1955):

$$I = I_o e^{-\mu x}$$  \hspace{1cm} (15-1)

where $I_o$ is the counting rate of the incident flux, $\mu$ is the mass absorption coefficient for beta absorption (cm$^2$/gm), and $x$ is the sample thickness (gm/cm$^2$). The mass absorption coefficient is normally determined through a calibration procedure involving the measurement of a series of known standards which bracket the mass range of interest (Jaklevic et al. 1981). The incident flux, $I_o$, can either be derived from the same calibration procedure or, if
the interval between successive measurements is short, the value of $I_0$ can be made to cancel by calculating the ratio between transmitted fluxes measured with and without the particle deposit. The latter case applies for certain beta gauge designs where continuous particle deposition is monitored (Macias and Husar 1970).

**INSTRUMENTAL DESIGN**

The optimal choice of source and detector depends upon many factors. The radioisotope source must have beta particle emission as the dominant mode of decay and exhibit half-life sufficient that large decay corrections or frequent source replacements are unnecessary. The source strength should provide adequate precision in counting statistics within the limitations of the detector rate handling capabilities. Finally, as discussed in more detail below, the energy of the beta spectrum must be chosen to produce a mass absorption coefficient matched to the range of thicknesses to be measured. Table 15-1 lists a selection of radioisotopes appropriate for aerosol beta gauge applications together with relevant parameters for each (Lilienfeld 1975).

The detector must be sensitive to the beta particles (i.e. electrons) in the energy range of interest and capable of operation at a counting rate sufficient to perform measurements in the required time interval. Beta gauges have been operated using a variety of detector types including ionization chambers (Husar 1974), Geiger-Muller tubes (Gleason, Taylor and Tabern 1951; Lilienfeld 1975; Klein, Ranty and Sowa 1984), scintillation spectrometers (Jaklevic, Madden and Wiegand 1983), and semiconductor diode detectors (Macias and Husar 1970; Jaklevic et al. 1981). Of equal importance is the choice of associated pulse processing electronics. Depending upon the type of detector used, it may be necessary to employ a preamplifier, amplifier, and pulse amplitude discriminator. Because of the level of precision required in many measurements, these analogue signal processing components must be selected and tested for long term stability and reliability. For those laboratory applications where extreme limits of precision and accuracy are the objective, temperature stabilization of the operating environment is recommended.

Most recent beta gauge systems employ semiconductor diode detectors and solid state pulse processing electronics because of their simplicity and stability of operation. The following discussion will be oriented toward this type of system although most of the comments also apply to other types of detectors provided the minimum stability and sensitivity requirements are met.
The source-detector geometry must be maintained in a stable mechanical configuration to minimize spurious counting rate variations. Also, it is important that the spacing be as close as possible in order that changes in atmospheric density within the gap are not interpreted as mass variations in the sample (Courtney, Shaw and Dzubay 1982). The lower limit to the spacing is normally determined by the thickness of sample holders and associated handling mechanisms used with automated instrumentation.

THEORETICAL CONSIDERATIONS

Studies have shown that the functional dependence of beta particle transmission expressed in Eq. 15-1 is not valid for precise experimental measurements (Jaklevic et al. 1981; Macias and Husar 1970; Heintzenberg and Winkler 1984). This result is not unexpected since the exponential behavior is not a reflection of fundamental mechanisms associated with beta particle attenuation in matter. Electrons with kinetic energies less than 1 MeV lose energy primarily through elastic collisions with atomic electrons present in the sample. As a consequence, an electron with a well-defined initial energy will slow down through a series of discrete energy losses as it traverses the sample. An incident electron beam with well-defined initial energy and direction will experience a gradual decrease in the average energy accompanied by a spreading in the energy and angular distributions.

The radioisotopic beta particle emission process results in a continuum of electron energies. Figure 15-2 is an idealized representation of a measured beta spectrum from a typical source. The energy distribution extends from a minimum energy determined by the source window thickness to a maximum endpoint energy, $E_{\text{max}}$, which is the total energy available for the radioactive decay process. An electronic discriminator level, $E_{\text{disc}}$, has been indicated above a low energy electronic noise tail. As the mass between source and detector is increased, the counting rate observed above this discriminator level in the beta gauge detector represents those electrons in the original continuum spectrum that have not been totally stopped in the sample and are still in a direction of incidence toward the detector. This rate reflects a complex energy-loss process which depends on several variables including the average energy of the electrons in the beta spectrum and the amount of material traversed. When averaged over all of these effects, the observed dependence of counting rate on the thickness transmitted is approximately exponential. Repeated measurements in a carefully defined experimental geometry using aluminum absorbers have established an
approximate relationship between the mass attenuation coefficient in Eq. 15-1 and the beta spectrum endpoint energy (Gleason, Taylor and Tabern 1951):

$$\mu (\text{cm}^2/\text{gm}) = 0.017 E_{\text{max}}^{-1.43}$$  \hspace{1cm} (15-2)

However, there is considerable variation among investigators regarding the values of the coefficients reflecting the empirical nature of these parameters (Sem and Borgos 1975).

Since the beta particle energy loss process involves scattering from atomic electrons rather than nuclei, there is the possibility of an additional dependence of the absorption coefficient upon the average atomic number of the samples. Various authors have studied this effect and an empirical relationship has been derived (Klein, Rancy and Sowa 1984):

$$\mu (\text{cm}^2/\text{gm}) = 0.16 (Z/A)^{4/3} E_{\text{max}}^{-1.37}$$  \hspace{1cm} (15-3)

However, the validity of this relationship seems to depend upon the specific geometry of the beta gauge. A less dramatic dependence of attenuation on Z/A has been observed by Jaklevic et al. (1981) and attributed to the relative importance of the particle angular distribution in the particular source-detector arrangement employed in that study. Regardless of the details of functional dependence, it should be noted that variations associated with this effect are normally tolerable since the range of Z/A is small for all elements with the exception of hydrogen.

The practical consequences of these theoretical observations are that, although estimates of beta gauge sensitivity can be obtained using generalized expressions, precise mass measurements require that each specific instrument is calibrated using known gravimetric standards. It is also necessary to limit the dynamic range over which one attempts to apply a given calibration using a strictly exponential approximation. To the extent that the deposited mass is normally a small fraction of the tare weight of the filter medium, this is not a severe limitation in aerosol applications.

A more complete understanding of precision and accuracy requires a detailed error analysis. If one assumes that the value of $\mu$ has been carefully determined and that the counting interval is known with complete certainty,
then the precision of the mass measurement is determined by experimental variations in the determination of \( I_0 \) and \( I \). Using Eq. 15-1, the root mean square error \( \sigma^2(x) \) in the calculated concentration, \( x \), can be derived as (Cooper 1975):

\[
\sigma^2(x) = \frac{1}{\mu^2} \left( \frac{\sigma^2(I_0)}{I_0^2} + \frac{\sigma^2(I)}{I^2} \right)
\]

For the case where the errors associated with \( I \) and \( I_0 \) are the result of Poisson counting statistics only, i.e. \( \sigma(I) = (I)^{1/2} \), and if the counting intervals of the two measurements are equal, the precision \( \sigma(x) \) for a difference measurement varies as the inverse of the mass absorption coefficient and the inverse square root of measurement interval as might be expected. The inverse dependence of precision on mass absorption coefficient supports the intuitive observation that lower energy beta spectra will provide a more sensitive indicator of small mass changes in the sample. On the other hand, if the energy is too low the exponential approximation is no longer valid since an increasing fraction of beta particles is totally stopped in the sample. Referring to Table 15-1, a useful rule of thumb would be to select a beta spectral average energy corresponding to a range which is several times the maximum thickness to be measured. For this reason, most beta gauges designed for aerosol monitoring employ either \(^{14}\text{C}\) or \(^{147}\text{Pm}\) as reasonable choices for use with substrates in the range of 10-100 mg/cm\(^2\) and deposits of 20-500 microgm/cm\(^2\) (Klein, Ranty and Sowa 1984).

The derivation of Eq. 15-4 assumed that the value of the mass absorption coefficient was known from a previous calibration procedure. However, the value of the mass absorption coefficient is normally calculated from transmission measurements performed on a series of mass standards which bracket the anticipated range of operation of the instrument. A more complete error analysis including uncertainties in the fitting procedure is discussed by Jaklevic et al. (1981). A general conclusion of this analysis is that, although the absolute accuracy of a mass measurements is affected by such calibration errors, the precision is dominated by the variability in the determinations of \( I \) and \( I_0 \). In principle, the precision of a given mass determination can be improved by increasing the counting interval to reduce the relative error associated with Poisson counting statistics. However, one
eventually reaches a limit where the variability in the measured counting rate is dominated by other effects which are not easily controlled. Sources of systematic errors can include fluctuations in atmospheric density, changes in laboratory relative humidity which can affect the substrate mass for hydroscopic media, instabilities in the mechanical design and variability in the placement of the sample in the instrument (Courtney, Shaw and Dzubay 1982). A major source of instrumental instability is the result of long and short term drift in the detector and analogue pulse processing electronics. Although modern solid state circuits can have temperature coefficients in the 1 part in $10^4$ per degree Celsius, the demands placed on precision beta gauge measurements can approach this level even in temperature controlled environments. Because of the difficulty in controlling such sources of errors, it should be a requirement in all beta gauge measurement protocols that recalibration of the instrument be performed on a frequent basis and that replicate samples be repeatedly analyzed at the same time as the unknowns in order to monitor instrumental stability.

**SYSTEMATIC EFFECTS**

The near exponential behavior of the beta absorption process and the variations discussed above can result in several potential measurement artifacts which should be understood. Principle among these are particle size effects, substrate inhomogeneity and atomic number dependence.

Particle size effects result from the fact that the beta gauge transmission measurements represent an average of the absorption experienced by an aggregate of particles deposited on the filter substrate. In the limit where this deposition is a homogeneous layer of small particles whose average diameter is much less than the layer thickness, the interpretation of the results in terms of exponential absorption by a uniform deposit is valid. On the other hand, one can imagine a deposit of equivalent mass but consisting of only a few very massive particles. In an extreme manifestation of this latter case, there could exist total absorption within a given particle. The transmission measurement in this limiting case would then reflect the fractional area covered by the particles and interpretation in terms of an exponential absorption by a uniform deposit would be invalid.

A detailed discussion of this problem for the general case exponential absorption is given by Cooper (1976). A similar treatment using a simplified model applied to the case of aerosol particles is given by Jaklevic et al.
Figure 15-3 is a plot of the discrepancy attributed to this phenomenon for the case of a 100 microgm/cm$^2$ deposit of unit density particles. The ratio of the mass as determined from beta gauge measurement to the true gravimetric mass is plotted as a function of the linear dimensions of particles. The discrepancy between the two increases for large particles at a rate which is greater for the source with the larger absorption coefficient.

These results indicate that one must either limit the size distribution to particles below 10 microns diameter or, if larger particles are to be analyzed, ensure that the average deposit thickness is sufficient that a statistically meaningful number of particles is present. Similarly, for impaction samples, the deposit thickness must be uniform over the measurement area to a degree where the average of the exponential is equal to the exponential of the average.

An effect which can be explained using similar logic has to do with discrepancies caused by filter and source inhomogeneities. Microscopic examination of most membrane filter media shows that the substrate consists of a non-uniform distribution of fibers or floculated material with relatively open spaces in between. Similarly, a radioisotope source is normally fabricated by methods which result in local inhomogeneities in the radioactivity across the face of the source. (Although a point source can in principal alleviate this problem, there is a practical limit to the specific activity that can be concentrated in a small volume). Variations in the apparent mass of the substrate can result from random alignments between respective inhomogeneities causing spurious high or low mass readings depending upon details. The problem is exacerbated by the large mass of the substrate relative to the deposit. If either the source or the substrate could be made to be perfectly spatially uniform this effect would vanish. Since this is in most cases impractical, it is possible to constrain the measurement protocol to position the filter in the instrument identically for both the initial and final weighings. This is most easily implemented in the case of large scale automated systems and is necessary for mass measurements which aspire to achieve the limits of instrumental precision (Courtney, Shaw and Dzubay 1982).

The atomic number dependence of the mass absorption coefficient requires that certain precautions be taken regarding choice of calibration standards and in the interpretation of results from discrete pollution sources. Table 15-2 shows the $Z/A$ values for a list of compounds commonly observed in ambient aerosol sampling and for some selected foil materials which might be used for calibrations. The mass absorption coefficients are calculated using both the $Z/A$ dependence observed by Jaklevic et al. (1981) and those calculated from Eq. 15-4. The mass absorption coefficient for a mixture of compounds would be the weighted sum of the respective coefficients. It is obvious that
an inappropriate choice of calibration foils can affect the accuracy of the measurements if not corrected for in the data
analysis. Similarly, measurements of a suite of samples in which the relative contribution of diverging Z/A
compounds varies widely will need special consideration in the interpretation. Although a complete correction
requires that the sample composition be known, some estimate of the probable error can be obtained by observing
the range of values of M for the compounds listed in Table 15-2 and incorporating an error analysis based on Eq. 15-
1. It should be noted that the errors associated with Z/A variations affect the accuracy of the measurements but not
the precision and, as a consequence, have little effect on the lower limit of sensitivity of the beta gauge method.

RESULTS AND APPLICATIONS

There are in the literature descriptions of a number of beta gauge designs optimized for specific particle
monitoring applications. In general a distinction can be made between those made for continuous monitoring of
discrete sources or ambient aerosols and those designed for precision laboratory analysis of collected particles. In on-
line monitoring applications, it is difficult to control the systematic errors associated with variability in the tare
weight and non-uniformities in the substrate mass. Such systems usually exhibit a root mean square precision in
the order of 25 microgm/cm² (Macias and Husar 1970; Husar 1974). In laboratory based systems where calibrations
are frequently performed, tare weights are individually measured and systematic effects such as sample placement are
reduced, it is possible to achieve precisions of 3 microgm/cm² for individual mass measurements corresponding to
precision of difference measurements of 5 microgm/cm² (Courtney, Shaw and Dzubay 1982; Jaklevic et al. 1981).
However, there are a number of criteria other than precision and individual designs which need to be evaluated in
terms of specific applications. These include speed, convenience, cost, operating environment and automated
operation.

Beta gauge mass measurements have been incorporated into a number of studies. Loo et al. (1978) report the
use of a beta gauge to measure the mass of aerosols collected on cellulose ester filters as part of a large scale
sampling program in St. Louis. They report a precision of 5 microgm/cm² for substrate masses in the order of 20
mg/cm². Stevens et al. (1980) have used beta gauge mass determinations for samples collected on
polytetrafluorethylene substrates with average areal densities of less than 600 microgm/cm². Spengler and Thurston
(1983) describe the use of beta gauge measurements in an extensive indoor air pollution study. Recent applications using continuous mass monitors have been described by Klein, Ranty and Sowa (1984), and Heintzenberg and Winkler (1984).
REFERENCES


FIGURE CAPTIONS

Figure 15-1 Schematic of a beta-gauge suitable for measuring thin aerosol samples.

Figure 15-2 Idealized beta particle spectrum emitted from a radioisotope source.

Figure 15-3 The discrepancy in beta gauge mass measurements as a function of particle size for the case of two commonly used isotopes.
Figure 15-1  Schematic of a beta-gauge suitable for measuring thin aerosol samples.
Figure 15-2  Idealized beta particle spectrum emitted from a radioisotope source.
Figure 15-3  The discrepancy in beta gauge mass measurements as a function of particle size for the case of two commonly used isotopes.
Table 15-1. Commonly Available Sources Suitable for Beta Attenuation Measurements.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half Life (years)</th>
<th>$E_{\text{max}}$ (MeV)</th>
<th>Range (mg/cm$^2$) in Carbon at $E_{\text{max}}$</th>
<th>Range (mg/cm$^2$) in Carbon at $E_{\text{aver.}} = 0.4 E_{\text{max}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{64}\text{Ni}$</td>
<td>92</td>
<td>0.067</td>
<td>7.7</td>
<td>1.6</td>
</tr>
<tr>
<td>$^{14}\text{C}$</td>
<td>5730</td>
<td>0.156</td>
<td>32</td>
<td>6.6</td>
</tr>
<tr>
<td>$^{147}\text{Pm}$</td>
<td>2.62</td>
<td>0.225</td>
<td>60</td>
<td>13</td>
</tr>
<tr>
<td>$^{85}\text{Kr}$</td>
<td>10.76</td>
<td>0.67</td>
<td>290</td>
<td>77</td>
</tr>
<tr>
<td>$^{36}\text{Cl}$</td>
<td>$3.1 \times 10^4$</td>
<td>0.712</td>
<td>320</td>
<td>84</td>
</tr>
<tr>
<td>$^{204}\text{Tl}$</td>
<td>3.8</td>
<td>0.765</td>
<td>340</td>
<td>94</td>
</tr>
</tbody>
</table>
Table 15-2. Effect of Atomic Number Dependence on the Measured Mass of Several Compounds.

<table>
<thead>
<tr>
<th>Compound</th>
<th>Z/A</th>
<th>$\mu$ (cm²/mg) \textsuperscript{a) }</th>
<th>$\mu$ (cm²/mg) \textsuperscript{b) }</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(\text{NH}_4)_2\text{SO}_4$</td>
<td>0.530</td>
<td>0.153</td>
<td>0.166</td>
</tr>
<tr>
<td>$\text{NH}_4\text{H SO}_4$</td>
<td>0.521</td>
<td>0.152</td>
<td>0.163</td>
</tr>
<tr>
<td>$\text{CaSO}_4\cdot\text{H}_2\text{O}$</td>
<td>0.511</td>
<td>0.152</td>
<td>0.159</td>
</tr>
<tr>
<td>$\text{SiO}_2$</td>
<td>0.499</td>
<td>0.154</td>
<td>0.154</td>
</tr>
<tr>
<td>$\text{CaCO}_3$</td>
<td>0.500</td>
<td>0.154</td>
<td>0.154</td>
</tr>
<tr>
<td>Carbon</td>
<td>0.500</td>
<td>0.154</td>
<td>0.154</td>
</tr>
<tr>
<td>$\text{Fe}_2\text{O}_3$</td>
<td>0.476</td>
<td>0.163</td>
<td>0.144</td>
</tr>
<tr>
<td>$\text{NaCl}$</td>
<td>0.478</td>
<td>0.172</td>
<td>0.145</td>
</tr>
<tr>
<td>$\text{PbSO}_4$</td>
<td>0.429</td>
<td>0.193</td>
<td>0.126</td>
</tr>
<tr>
<td>$\text{PbCl}_2$</td>
<td>0.417</td>
<td>0.204</td>
<td>0.121</td>
</tr>
<tr>
<td>$\text{PbBrCl}$</td>
<td>0.415</td>
<td>0.206</td>
<td>0.120</td>
</tr>
</tbody>
</table>

\textsuperscript{a) } From Jaklevic 1981.

\textsuperscript{b) } Z/A dependence calculated from equation 15-3. Values normalized to 0.154 for carbon to account for instrumental differences.