

- (21) Application No 8127202
(22) Date of filing 9 Sep 1981
(30) Priority data
(31) 3035929
(32) 24 Sep 1980
(33) Fed. Rep. of Germany (DE)
(43) Application published
31 Mar 1982
(51) INT CL³
G01N 23/08
(52) Domestic classification
G1A A4 C3 C4 CD D10
D12 D3 D6 G17 G7 P11 P1
P5 R2 R7 S10 S6 T20 T2
(56) Documents cited
None
(58) Field of search
G1A
(71) Applicants
GKSS-Forschungszentrum
Geesthacht GmbH.,
Reaktorstraße 7—9, 2054
Geesthacht, Federal
Republic of Germany
(72) Inventors
Walfried Michaelis,
Hans-Ulrich Fanger
(74) Agents
Kilburn & Strode,
30 John Street, London
WC1N 2DD

(54) Device for determining the proportions by volume of a multiple-component mixture

(57) A device for determining the proportions by volume of n-component mixture 3, whose components have differing mean atomic numbers, includes source 1 for irradiating the mixture on a common axis with gamma radiation having n-1

energy peaks at different energies. For each of the energies the absorption coefficients of the components are different. n-1 radiation detectors 4, 5 each measure the intensity of a respective one of the lines after the radiation has passed through the mixture. Calculating unit 11 uses the measured intensities to calculate said proportions by volume.

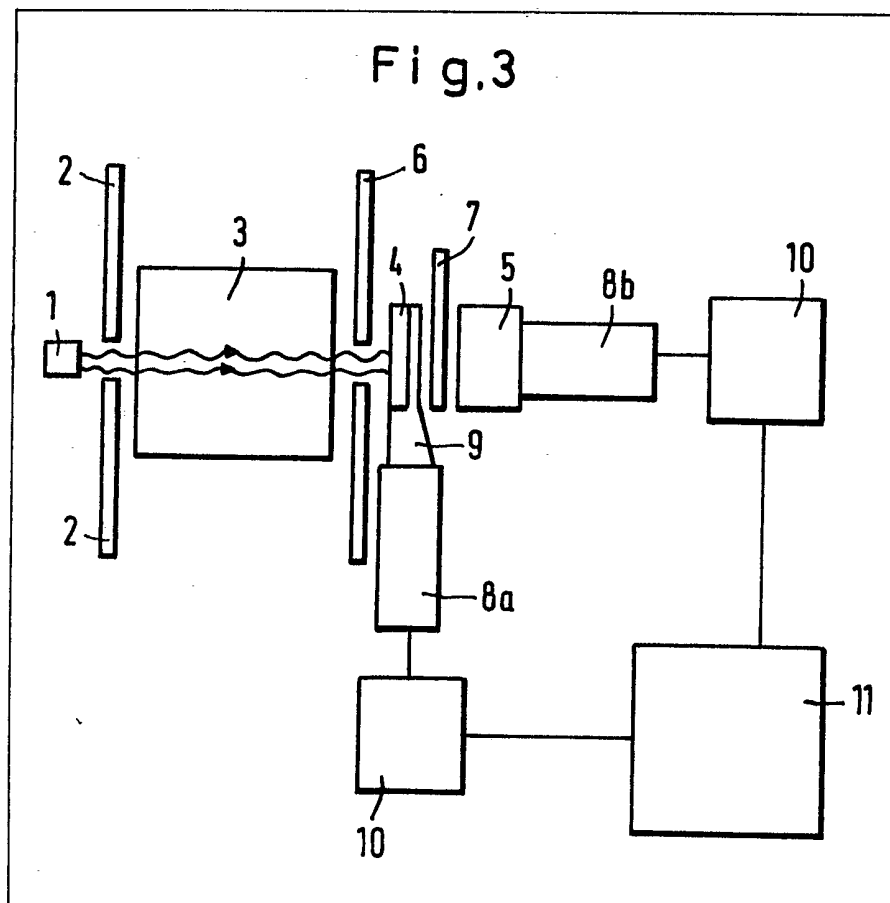


Fig 9.1

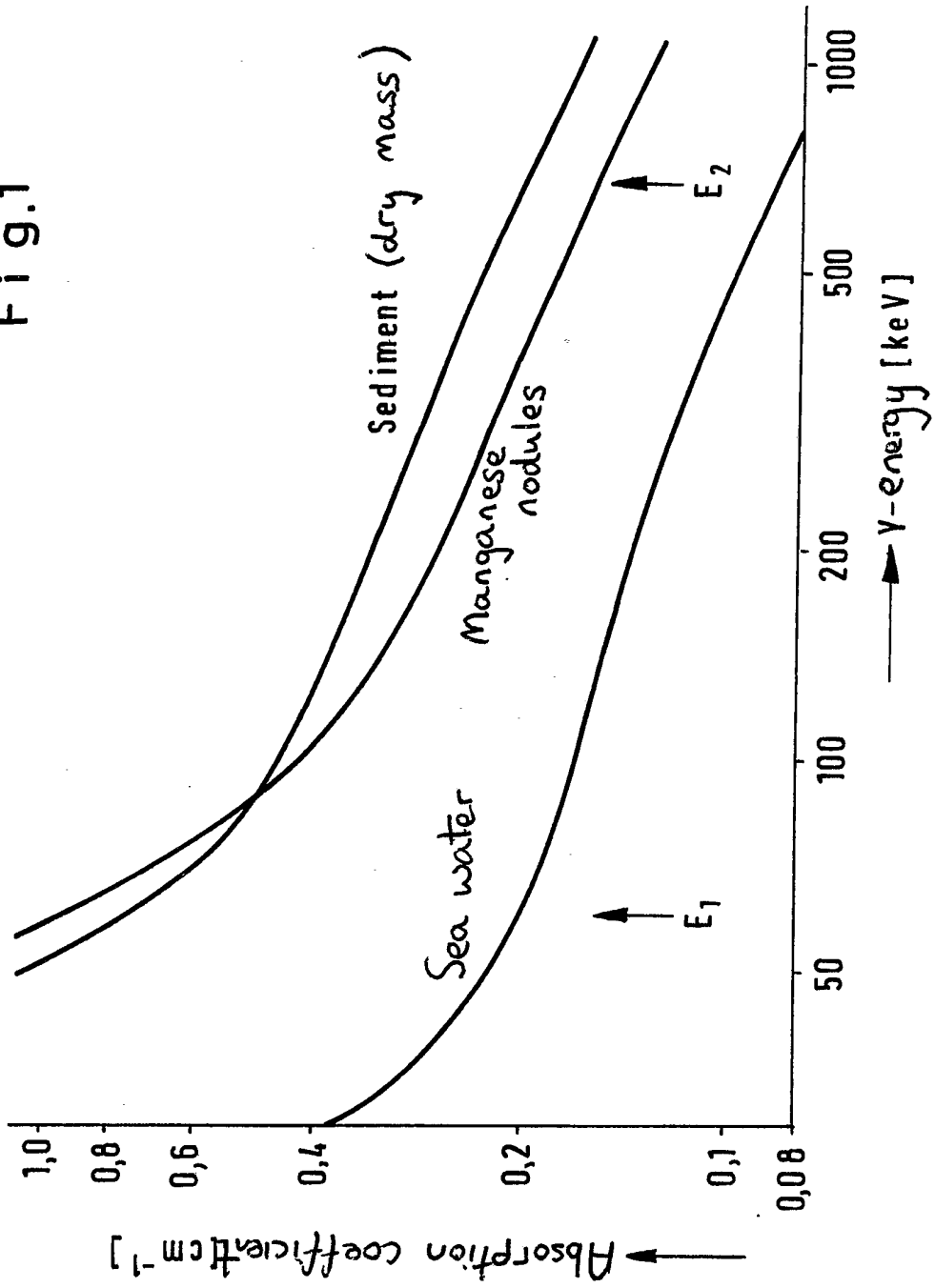


Fig.2

- I Water
 II 7,5 Vol.% quartz sand + water
 III 8,0 Vol.% manganese nodules + water ^{137}Cs

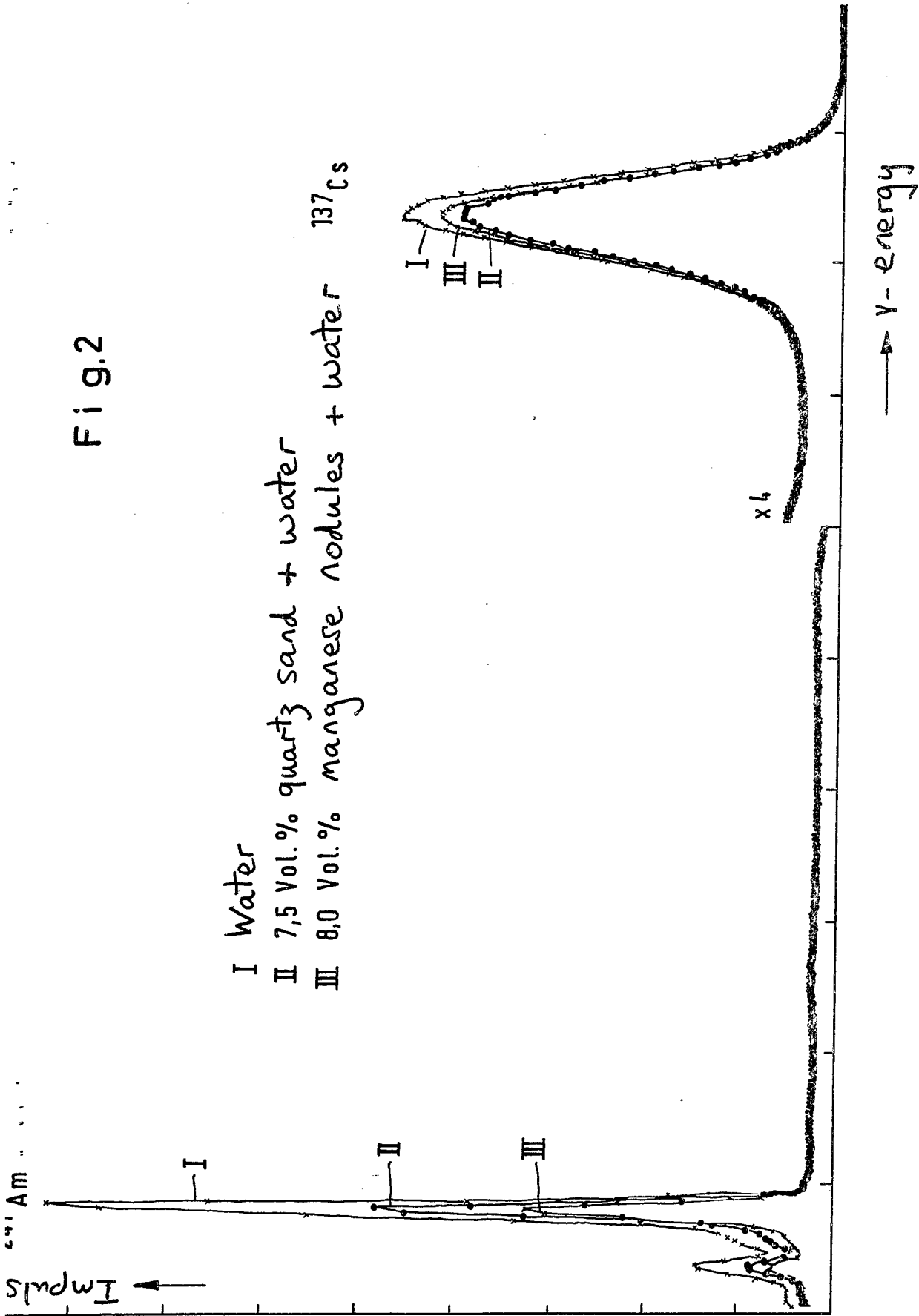
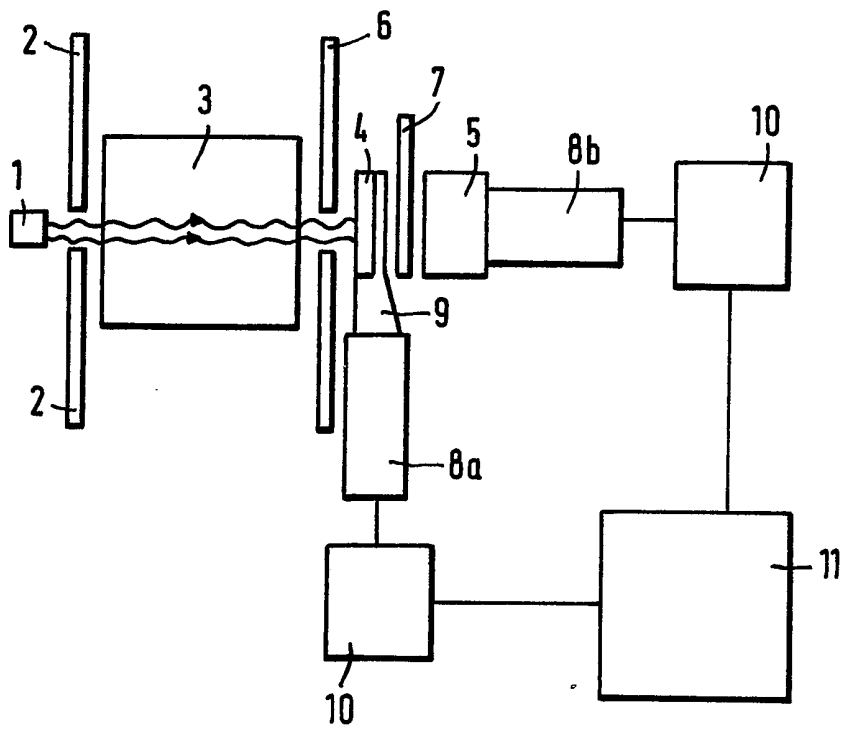


Fig.3



SPECIFICATION

Device for determining the proportions by volume of a multiple-component mixture by irradiation with several gamma lines

The invention relates to a device and method for determining the proportions by volume of a multiple-component mixture in which the mixture is irradiated with two or more gamma lines and the intensities of the radiation which pass through the mixture are measured and used to calculate the said proportions. A gamma line is gamma radiation with a substantial energy peak, that is to say that the substantial majority of the radiation has an energy equal to or very close to a single known value. This fact means that the radiation is essentially monochromatic, that is to say it is of substantially only one frequency, but in the field of gamma radiation it is conventional to refer to energy rather than frequency.

In industrial technology there is an increasing need for methods for a contact-free, rapid and continuous determination of the concentration by volume of one or more of the components of a multiple-component mixture. This need is due to, amongst other things, the increasing importance of hydraulic transport of solid materials. Generally the objects to be measured are opaque bodies, that is to say the bodies themselves or, for instance the surrounding conveyor pipe is opaque, so that for contact-free measurement only the use of penetrating gamma radiation and the analysis of the interaction of the gamma quanta with the object to be studied is practical.

A method of this general type is described in German Auslegeschrift No. 2622175 and in the periodical "meerestechnik" (Marine Technology) 10, 1979, No. 6, pages 190—195, which method is essentially based upon the fact that for two substances (p and q) with sufficiently different mean atomic number Z the ratio of the gamma absorption coefficients μ in the region of low gamma energy up to approximately 1.5 MeV is highly energy dependent. Using this fact it is possible for the two unknown component proportions or volume proportions of these components v_p and v_q in a three component mixture to be unambiguously determined from two equations by the measurement of the radiation intensities J with and without the presence of the absorbing bodies at two different gamma radiation energies (E_1, E_2). Since the dimensional parameters during measuring are normally fixed and thus the length of the transmission path L in the irradiated medium is constant, the third component is given as a by-product from the limiting condition that the sum of the three proportions by volume must be 100%. When one is concerned with hydraulic conveying systems the third component is generally water (w), which as a rule takes up the space in the conveyor pipe which is left free by the solid components p and q . In this case it is convenient to select not the absorber-free (vacuum) intensity, that is to say the radiation intensity when no material whatever is present, but the intensity J_w of the gamma radiation for solid-free water as the reference parameter. Thus the two transmission equations for the energies E_1 and E_2 have the form

$$t_1 = \frac{J_1}{J_{w_1}} = e^{-L} \left[v_p \mu_{p_1} + v_q \mu_{q_1} - (v_p + v_q) \mu_{w_1} \right] \quad 35$$

$$t_2 = \frac{J_2}{J_{w_2}} = e^{-L} \left[v_p \mu_{p_2} + v_q \mu_{q_2} - (v_p + v_q) \mu_{w_2} \right]$$

$$\text{with } v_p + v_q + v_w = 1.$$

Solution for v_p and v_q gives

$$v_p = (LN)^{-1} \left[\int_1 \int_2 (\mu_{q_2} - \mu_{w_2}) + \int_2 (\mu_{q_1} - \mu_{w_1}) \right]$$

or

$$v_q = (LN)^{-1} \left[\int_1 (\mu_{p_2} - \mu_{w_2}) - \int_2 (\mu_{p_1} - \mu_{w_1}) \right]$$

where

$$N = (\mu_{p_1} - \mu_{w_1}) (\mu_{q_2} - \mu_{w_2}) - (\mu_{p_2} - \mu_{w_2}) (\mu_{q_1} - \mu_{w_1}).$$

The two gamma lines advantageously pass through the volume to be measured on a common radiation axis and thus can be used to determine these volume proportions exactly. Differences in

physical structure which would lead to errors of inhomogeneity if the transmission of the two lines occurred at different places in the volume to be measured do not therefore cause any problem.

Naturally this process can be applied to mixtures with more than three components. A further gamma line is then necessary for each additional component. In the calculation a further transmission equation is produced for each additional component.

The errors in determining the volume concentrations depend upon the precision of the measurement of the gamma intensities. The influence of the relative errors in the measured gamma intensities is independent of the volume concentration and inversely proportional to the length of the transmission path L.

Since the proportions of solid material are often only in the region of a few percent, it is desirable that for sufficiently accurate measurements the relative errors should like in the region of, or only slightly above, 0.1%.

In the method disclosed in German Auslegeschrift No. 2622175 the intensities of the two gamma lines are determined separately by pulse height analysis of the pulses emitted in a conventional scintillation counter which responds to the two lines together. The counter must show an adequate energy resolution so that reciprocal interference remains low. The most suitable substance for use as a scintillator material is sodium iodide (NaI) doped with thallium.

The critical disadvantage of conventional spectroscopy with the NaI referred to is the relatively long fluorescent time constant of this scintillator of 0.25 μ s. Of necessity this results in a pulse length in the region of microseconds which at high counting rates leads to pulse pile-up and displacement of the zero line and thus finally to inaccuracies in determining the intensities. A practical upper limit for the counting rate is approximately 50,000 pulses/s, if the errors given above are not to be markedly exceeded by systematic errors. For statistical reasons this counting rate in turn implies minimum measuring times purely arithmetically of approximately 40 s and in practice generally in the region of 50 s. Therefore the process can only be described as quasi-continuous. Faster scintillation detectors do exist but they do not permit adequate energy discrimination.

An object of the present invention, therefore, is to provide a device of the type referred to which avoids the known problems and in particular makes shorter measuring times possible.

According to the present invention there is provided a device for determining the proportions by volume of an n-component mixture, the components having differing mean atomic numbers, including means for irradiating the mixture on a common axis with n—1 gamma lines of different energies at which the absorption coefficients of the components are different, a number of radiation detectors equal to the number of gamma lines used, each detector being responsive to substantially only a respective one of the gamma energies and arranged to measure the intensity of its respective gamma line after it has passed through the mixture and calculation means connected to the radiation detectors arranged to calculate the said proportions by volume from the measured radiation intensities.

According to the invention a separate detector is used for each gamma line and essentially measures the intensity of only this line. The difficulties which occur when several gamma lines have to be discriminated in one detector and which lead to considerable reduction in the achievable counting rate are thus avoided. The pulse height analysis which is electronically complicated and lowers the counting rate can be dispensed with. The individually determined counting rates can be directly used in calculations in a simple manner. With this arrangement it is easily possible to use detectors which each have substantial sensitivity only for the gamma line which it is to detect. The residual sensitivity of the detector for the other lines can be compensated for either arithmetically or by suitable arrangement or selection if it significantly alters the measured result. Possible arrangements of the different detectors are arrangement adjacent to one another or arrangement behind one another in the path of the radiation. In addition it is also possible to split the ray by means of radiation splitters, for example in a frequency selective manner by means of crystal lattices or the like. The essential advantage of the prior arrangement adjacent to one another or arrangement behind one another in the path of the gamma lines used on a single axis.

The detectors are preferably arranged one behind the other on the common axis. With the detectors arranged adjacent to one another in the radiation, each detector sees only a part of the cross-section of the radiation. Thus slight variations in intensity caused by inhomogeneities in the volume to be measured may cause inaccuracies. This does not occur when the detectors are arranged one behind the other, since all the detectors are subjected to the same or the entire cross section of the radiation. The different gamma lines can be easily discriminated in the respectively associated detectors in two ways. On the one hand selective detectors can be used which are sensitive only to the energy of one respective gamma line. However, even with identical detectors arranged one behind the other different gamma energies can still be selectively detected. The exemplary table below shows the results using two lines (low energy and high energy) and two detectors (D_1 and D_2):

	D ₁	D ₂
low	95%	5%
high	20%	16%

D₁ and D₂ are two identical detectors, of which D₂ is arranged behind D₁. Both are subjected to the same cross-section of the radiation. The detectors have differing sensitivity to the two lines and absorb in each case 95% of the energy incident on it of the low energy line and 20% of the high energy line. Thus in the detector D₁ 95% of the first line is absorbed (and thus indicated). The second detector can only receive the remaining 5% and indicates 95% of this, i.e. approximately 5%. In the first detector 20% of the higher energy line is absorbed. Thus 80% reaches the second detector and 20% of this 80% is absorbed there, i.e. approximately 16%. Thus it will be seen that the two lines of differing energy are sufficiently discriminated in both detectors, namely with difference factors 95:20 or 5:16. These ratios can be further improved by differing thicknesses of the detectors. Thus in the present example the thickness of the first detector D₁ can be chosen so that it absorbs substantially 100% of the line of lower energy. Then the second detector no longer sees this line. This would give the following result:

	D ₁	D ₂
low	100%	0%
high	21%	16%

In this way the effort required for performing the calculation is reduced.

Thus the selective sensitivity to particular energies can be achieved in identical detectors solely by means of their arrangement. Such an arrangement preferably includes an absorber between each adjacent pair of detectors which permits the passage only of those gamma energies to which the subsequent detectors are responsive. If behind one detector the line which is to be measured by that detector is blocked by a suitable absorber which allows the other lines to pass through substantially unattenuated, then this absorbed line does not influence the subsequent detectors, so that discrimination and subsequent calculation of the values is significantly simplified.

The detectors are preferably arranged so as to be responsive to increasing energies of radiation in the direction in which, in use, the radiation passes. In this way the physical features of the detectors and the optionally interposed absorbers are taken into account. Higher energies are more penetrating and can still be detected in the last detector without significant attenuation, whereas lower energies are better detected at the beginning of the detector chain because they are more strongly attenuated in the detectors.

The detector or detectors responsive to the lowest or lower energy lines are preferably scintillation counters whilst the detector responsive to the highest or higher energy lines are preferably Cerenkov counters. These types of detectors are selectively responsive to the respective energy ranges and are characterised by high counting rates.

Further features and details of the invention will be apparent from the following description of one specific construction which is a device which can be used in marine technology for examining a mixture of manganese nodules, sediment and sea water which is given by way of example only with reference to the following drawings in which:—

Figure 1 shows the curves of the energy dependence of the absorption of gamma energy in three different substances;

Figure 2 shows the spectrum of two different gamma lines which are preferably used; and

Figure 3 is a block diagram of a device in accordance with the invention for irradiation of a test volume with two gamma lines.

Figure 1 shows the different absorption coefficients of manganese nodules, sediment and sea water as a function of the energy of the incident gamma radiation used. It can be seen that with differing gamma energies differences in absorption characteristics are clearly present and measurable. Figure 2 shows the energy spectrum of two gamma radiation emissions, namely that from Americium 241 and Caesium 137. The two lines are each shown at three different heights I, II and III after passing through different media, the associated reference numerals indicating the medium as follows:

- I water
- II 7.5% by volume quartz sand in water
- III 8.0% by volume manganese nodules in water.

The X axis of the graph shows the energy of the gamma radiation, whilst the Y axis shows the number of impulses produced by a scintillation counter on which the radiation is incident and thus indicates the intensity of the radiation at different energies, which is to say at different frequencies. As can be seen, the different intensities resulting from the differing absorption coefficients can be easily evaluated and plotted, each substance emitting only one pronounced gamma line.

One embodiment according to the invention will now be described with reference to Figure 3.

A gamma radiation source 1 emits two gamma lines at energies E_1 and E_2 . E_1 is chosen to have a relatively low energy (see Figures 1 and 2) and E_2 has a significantly higher energy. The source 1 may be Americium 241 and Caesium 137 either in discreet lumps or amalgamated in a form of alloy, or any other convenient source of two gamma lines. After collimation in a device 2 the gamma radiation passes through a body 3 to be examined and is absorbed by detectors 4 and 5, optionally after further collimation in a device 6. These detectors should on the one hand be characterised by very small time constants and on the other hand must also be advantageously selected and dimensioned so that the first detector 4 almost completely absorbs the low energy radiation, but allows the high energy radiation to pass through substantially unattenuated. The second detector 5 then in practice responds only to the high energy components of the gamma radiation. An absorber 7 of suitable thickness and atomic number can optionally be provided between the two detectors. The detectors are generally (i.e. in light-emitting systems) connected to photomultipliers 8a and 8b, in the case of the first detector conveniently via a suitably formed optical guide 9. The pulses from the two counting systems are counted in respective electronic counters 10 and the figures thus obtained are passed to a calculating unit 11 which performs calculations with the aid of the transmission equations given above and reference parameters, which may be obtained as described above, and calculates the proportions by volume of the constituents in the body 3.

In the method described the mixture is irradiated with the two gamma lines simultaneously and this is important with mixtures of this type which are inhomogeneous and whose components tend to move relative to one another since if the irradiation and measurement of the two gamma lines were carried out sequentially the irradiated portion of the mixture might have a slightly differing composition at the different times which would lead to errors in the end result. If, however, the mixture is completely homogeneous or entirely static, e.g. solid, the irradiation and measurement of the two gamma lines may be carried out either simultaneously or sequentially and it is to be understood that in this case the gamma radiation source may comprise two separate single line sources which are used sequentially.

A suitable substance for the first detector 4 is, for example, CsF, a scintillator with a time constant of $0.005 \mu\text{s}$. It only has a low light yield (3% relative to doped NaI) and thus a poor energy resolution (which is not essential here), but because of its favourable time constant it permits very high counting rates (up to the region of several MHz.). CsF is well suited to discrimination of both gamma energies; a 1 mm thick detector absorbs 94% of a 60 keV radiation, but only 2.5% of a gamma radiation at 1250 keV. The corresponding figures for a 5 mm thickness are 100% and 12% respectively. Since it can be ensured that the second counter 5 responds exclusively to the high energy components, the slight absorption of this radiation in the first detector can be easily corrected for. Other possible materials for the first detector — although they have somewhat less favourable discrimination properties — are, for example, plastic scintillators, preferably with Sn or Pb doping.

A Cerenkov counter, for example, is suitable for the second detector. If lead glass is used, a high density and atomic number (and thus very favourable absorption properties) can be achieved as well as a high refractive index.

When measuring two gamma lines of different energy, e.g. from a ^{60}Co source (1.17 and 1.33 MeV), the high energy component can be detected with a high degree of efficiency if the photomultiplier is selected with regard to low photon yields. This result was confirmed experimentally. The low energy component of the gamma radiation is, however, not registered since the energy can be so selected that the maximum speed of the electrons produced in the counter is below the limiting speed below which no Cerenkov radiation occurs. In this way complete discrimination is achieved.

It is a characteristic feature of the Cerenkov effect that the counter responds very quickly to the gamma quanta (in 10^{-11} s or less). The temporal limitation occurs with a considerably slower multiplier (≈ 1 ns). Thus for the second detector which detects the gamma quanta of the energy E_2 , counting rates in the region of several MHz can be achieved. CsF can also be used as the detector for the high energy radiation. In this case, if necessary, residual low energy radiation can be prevented from triggering signals in the second detector by a suitable absorber 7.

The arrangement described thus permits pulse height analysis to be dispensed with completely. The combination for example of a CsF counter and a Cerenkov counter in a "sandwich" arrangement permits counting rates which were not previously possible and thus measuring times in the region of

seconds or even less. In this way a truly continuous measurement is possible.

The device illustrated uses two gamma lines for determination of three components. By analogy four components, for example, can be determined with three gamma lines.

5 The described detectors which operate by energy selection can be arranged adjacent to each other in the path of the radiation instead of behind one another. In addition, for example, parts of the radiation may be deflected via crystal lattices and directed to different detectors arranged at an angle to the common axis. 5

10 It is also possible to dispense with detectors which only respond to certain energies but completely suppress others. The energy discrimination in the detectors can be achieved solely by the arrangement of the detectors one behind another, even when these are sensitive to all energies, so long as the sensitivity is only energy dependent. This method was described in greater detail above. 10

CLAIMS

15 1. A device for determining the proportions by volume of an n-component mixture, the components having differing mean atomic numbers, including means for irradiating the mixture on a common axis with $n-1$ gamma lines of different energies at which the absorption coefficients of the components are different, a number of radiation detectors equal to the number of gamma lines used, each detector being responsive to substantially only a respective one of the gamma energies and arranged to measure the intensity of its respective gamma line after it has passed through the mixture and calculation means connected to the radiation detectors arranged to calculate the said proportions by volume from the measured radiation intensities. 15 20

2. A device as claimed in Claim 1 in which the detectors are arranged one behind the other on the common axis. 20

25 3. A device as claimed in Claim 2 including an absorber between each adjacent pair of detectors which permits the passage only of those gamma energies to which the subsequent detectors are responsive. 25

4. A device as claimed in Claim 2 or Claim 3 in which the detectors are responsive to increasing energies of radiation in the direction in which, in use, the radiation passes.

5. A device as claimed in Claim 4 in which the detector responsive to the lowest energy is a scintillation counter and the detector responsive to the highest energy is a Cerenkov counter.

30 6. A device for determining the proportions by volume of an n-component mixture substantially as specifically herein described with reference to Figure 3 of the accompanying drawings. 30