

SATURATION BACKSCATTERING OF BETA PARTICLES IN COMPLEX TARGETS

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Experiments on saturation backscattering of β particles of ^{32}P and $^{90}\text{Sr} + ^{90}\text{Y}$ have been carried out with elements, metallic alloys, salts and their aqueous solutions, and natural and 99.8% heavy water.

Satisfactory correlation was observed between the intensity of the backscattered beam and the equivalent atomic number of the target when Saldick's formula for \bar{Z} and ours was applied. Natural and heavy water backscattered identically however several per cent less than the general trend to elements, metallic alloys and salts. Therefore a small correction for the \bar{Z} values of water, which was also satisfactorily applied to aqueous solutions, has been envisaged.

1 — INTRODUCTION

Saturation backscattering of β particles has been little studied in complex targets and only a few papers were devoted to liquids. The most important experimental work has been done by R. H. MÜLLER and D. C. MÜLLER (1, 2) and by L. DANGUY and R. QUIVY (3, 4, 5), using salts and their aqueous solutions, metallic alloys and organic compounds.

A general review of these and earlier works is given in a recent monography (5) published by L. DANGUY in 1962.

In the papers cited above, the authors applied R. H. MÜLLER's empirical formula for equivalent atomic number (6) which, for a chemical compound A_mB_n , is given by

$$\bar{Z}_{A_mB_n} = \frac{m A Z_A + n B Z_B}{m A + n B} \quad (1)$$

where A and B stand for atomic weights.

A generalised form of it, for homogeneous scatters, is the following:

$$\bar{Z} = \sum_i c_i Z_i \quad ; \quad \sum_i c_i = 1 \quad (2)$$

where c_i is the concentration by weight (weight fraction) of component i .

D. C. and R. H. MÜLLER (1, 2) claimed that backscattered intensity of β beam was a discontinuous function of the above equivalent atomic number, but strictly linear with \bar{Z} in each period of the periodic system. Also, bonded hydrogen exhibits a «negative backscattering» in organic compounds which, however, may be corrected and handled by simple means (2).

On the other hand, L. DANGUY found an almost linear relationship between backscattered beam and $\bar{Z}^{1/2}$ (3, 4) and a linear relation when, instead of this atomic number, the empirical quantity R was used (5)

$$R = \sqrt{\sum_i c_i \frac{Z_i (Z_i + 1)}{A_i}} \quad (3)$$

The author also suggests that R may account for the hydrogen anomaly observed by D. C. MÜLLER (2), through the ratio Z/A , which indeed equals to unity for ^1H whereas it is of the order of 0.5 for deuterium and light elements, and even smaller for heavy ones.

It is well known (5) that measured backscattered intensities are strongly dependent on counting geometry and experimental assembly. Therefore, experiments of different authors may hardly be compared quantitatively. On the other hand, the definition of an equivalent atomic number for complex scatters, or any other quantity related with it, must probably be independent of a particular measuring system and, therefore, such a definition by a consistent theoretical way appears to be one of the most important problems in connection with saturation backscattering, even when we may not go deeply into a treatment of this very complicated phenomenon.

It has been theoretically shown by one of us (7) that a better formula for the equivalent atomic number which would be attached to complex targets, for β scattering, is given by

$$\bar{Z}_{A_mB_n} = \frac{m Z_A^2 + n Z_B^2}{m Z_A + n Z_B} \quad (4)$$

in the case of a chemical compound A_mB_n , or a generalised form of it, namely,

$$\bar{Z} = \frac{\sum_i c_i Z_i^2/A_i}{\sum_i c_i Z_i/A_i} \quad (5)$$

where c_i , as in equation 2, is the concentration by weight of component i .

Formulas 4 and 5 reduce to that of MÜLLER if $(Z/A)_i$ values are all equal for the components of the target. In general, it has been shown (7) that MÜLLER's formula gives rise to higher values of the equivalent atomic number and the difference to our value of \bar{Z} is mostly pronounced when the target contains hydrogen.

We point out that formulas 4 and 5 for \bar{Z} have to be regarded as an approach (7), which may be applied in the energy range from 50-100 keV up to few MeV. The «effective» atomic number of a complex target would be somewhat smaller, indeed in between the value given by eq. 4 or 5 and the one given, approximately, by

$$\zeta = Z_1^{\varepsilon_1} \cdot Z_2^{\varepsilon_2} \dots Z_n^{\varepsilon_n} \quad (6)$$

where, in general,

$$\varepsilon_i = \frac{c_i Z_i/A_i}{\sum_j c_j Z_j/A_j}$$

or, for a compound A_mB_n ,

$$\varepsilon_A = \frac{m Z_A}{m Z_A + n Z_B}$$

and identically for ε_B .

Equation 4 and a modified but equivalent form of equation 5, have empirically been chosen by W. P. JESSE and J. SADAUSKIS (8) when considering backscattering correction, in ionisation chambers, due to the filling gas and, also, by J. SALDICK and A. O. ALLEN for backscattering corrections as well, in experimental determination of the yield of ferrous sulfate radiation dosimeter (9, 10).

One of the aims of this paper is to show that a better correlation with intensities of backscattered β beams was obtained when our formulas 4 and 5 were used instead of those of MÜLLER and DANGUY.

Experimental assembly has been envisaged in order to give precise measurements of saturation backscattering,

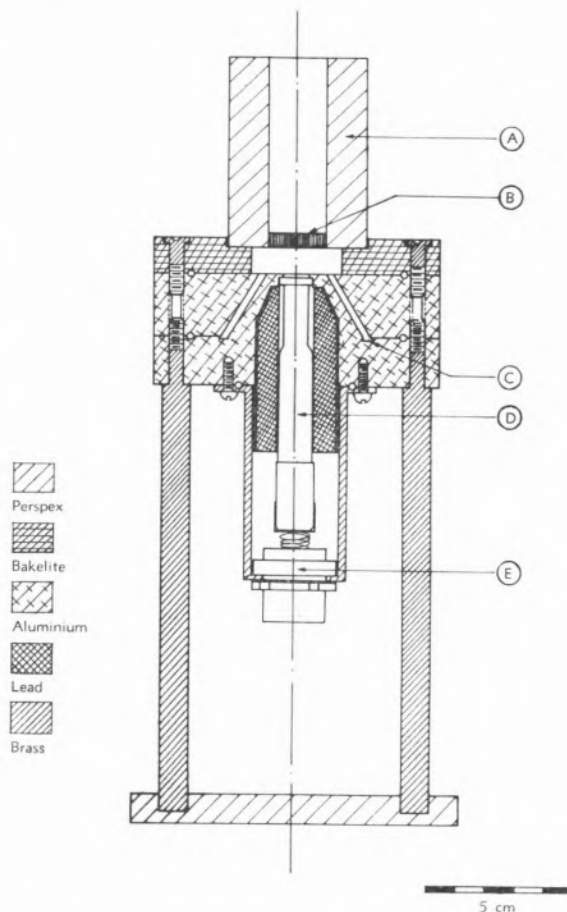


Fig. 1 — Experimental assembly. A — container of the targets; B — solid or liquid target; C — circular groove for β sources; D — G.M. counter; E — electric connection for probe unit and scaler.

which are necessary to make valuable comparisons among the correlations obtained when either MÜLLER's, DANGUY's or our formulas are used.

2 — EXPERIMENTAL

2.1 — EXPERIMENTAL ASSEMBLY

Our experiments on saturation backscattering have been carried out with the system drawn in fig. 1. It has been

designed in order to give precise measurements and, practically, only backscattered β radiation was detected. In a great part of earlier works, the authors used systems either with large backgrounds or in which both incident and backscattered beam were measured which is a marked disadvantage, since backscattered radiation represents only a small part of incident β particles, at least for targets of low atomic number.

About 25 microcuries of ^{32}P or $^{90}\text{Sr} + ^{90}\text{Y}$ were deposited uniformly distributed in a circular groove (*C* in fig. 1). β particles were focused by two conical aluminium pieces on a small ring area of the target (*B*). Angle of the cones was 120° .

Backscattered beam was measured by a Geiger Müller counter (*D*) type Philips 18 513, coaxial with cones and target, with a mica window of $2\text{--}2.5\text{ mg.cm}^{-2}$. The counter was shielded with lead. The counting system also included a probe unit type 1014-A fixing a quenching time of 300 microseconds, and a scaler EKCO type n.° 529.

The container (*A*) was made of perspex, with a 1.4 mg.cm^{-2} styrafoil window, strong enough to hold liquid and solid targets with several times the saturation thickness without observable deformation. The inner diameter was 2 cm in the experiments reported in this paper. However, some observations have been made with target diameter of 4 cm and no difference in the intensity of backscattered beam was detected.

Counting times ranged from 2 up to 10 minutes, corresponding to the integration of 20,000 counts. An exception was made for natural and heavy water, for which counting times were 20 minutes (about 40,000 counts) in order to obtain greater precision.

Measurements have been repeated several times, on different days, and mean values were taken. Experimental errors are of the order of 1%.

Due to strong focusing which has been envisaged, as well as the use of material of low atomic number and the shielding of the G. M. counter, backgrounds have appreciably been reduced.

We think, therefore, that our system enables us the experimental test of the best formula for equivalent atomic number which would be attached to saturation backscattering with complex targets.

2.2 — TARGETS

Experiments have been carried out with the following targets:

- Elements — *Al, Fe, Cu, Zn, Ag, Sn, I, Pb*.
- Metallic alloys. (The numbers represent percentage by weight of the lighter element).
Aluminium and zinc — 95.6, 92.7, 84.0, 84.3, 63.0, 42.1, 21.2.
Zinc and tin — 80.1, 60.1, 38.9, 19.8.
Tin and lead — 78.8, 60.0, 47.3, 38.6, 24.7, 20.2, 9.8.
- Salts — *KI, KBr, KCl, NaCl, BaCl₂*.
- Aqueous solutions with natural water. (The numbers represent percentage by weight of the salt).
KI — 0.58, 1.22, 2.47, 4.11, 8.15, 15.46, 28.67, 42.21, 59.06.
KBr — 2.45, 5.95, 11.47, 21.29, 37.19.

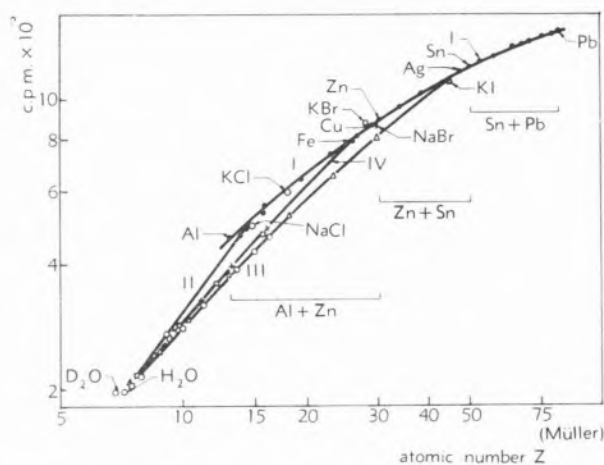


Fig. 2 — Correlation between intensity of backscattered beam and Muller's atomic number, with a source of $^{90}\text{Sr} + ^{90}\text{Y}$.

- elements, and metallic alloys (curve I).
- ⊕ natural and 99.8% heavy water.
- NaCl (curve II), △ KI (curve III), □ KBr (curve IV), × NaBr,
- BaCl₂, ● KCl and their aqueous solutions.

KCl — 19.28, 22.38.

NaBr — 0.93, 1.56, 2.54, 4.21, 6.82, 10.95, 17.36, 26.51, 39.15.

NaCl — 21.34, 25.31, 26.22.

BaCl₂ — 1.95, 5.91, 7.74, 11.33, 17.83, 21.89, 25.56.

- Bidistilled natural water;
99.8% heavy water.

Solid targets were in form of cylindrical pastilles, with a diameter very closed to that of container *A* (2 cm).

Pastilles of dry salts and of iodine were made by compression and all of them showed a good mechanical resistance.

Thickness of both solid and liquid targets were several times the saturation thickness. ⁽¹⁾

2.3 — RESULTS

A few examples of our measurements on saturation backscattering with a source of ⁹⁰Sr + ⁹⁰Y in radioactive equilibrium, are given in table I.

Equivalent atomic number, according to MÜLLER's equations 1 and 2 and our formulas 4 and 5 are presented in columns 4 and 5, respectively.

In order to discuss the correlation between the intensity of backscattered beam and the equivalent atomic numbers, we divide the targets which have been used in the following groups:

a) Elements, metallic alloys and salts

For elements, the values of *Z* are, of course, the same in the limiting case of both MÜLLER's and our formulas.

For metallic alloys, equivalent atomic numbers are approximately equal by both formulas, since values of *Z/A*, given in table I, column 2, are rather close together for the elements of each binary alloy.

The sensibility of our experimental assembly is not enough to detect any eventual difference so that we cannot decide between both formulas for equivalent atomic number, by experimental means.

The same happens with salts which have been used in our experiments.

b) Natural and 99.8 % heavy water

For hydrogen and deuterium, the values of *Z/A* are, respectively, 0.992 and 0.496. Accordingly, MÜLLER's equivalent atomic numbers for natural and 99.8 % heavy water ⁽²⁾ are different, being equal to 7.22 and 6.80, respectively. Our value of \bar{Z} is equal to 6.60 in both cases.

According to the sensibility of our apparatus in this region of *Z*, a difference of about 200 counts per minute should be observed between the intensities of the beams backscattered by *H*₂*O* and *D*₂*O* if MÜLLER's formula was adequated. No such a difference was found. On

the contrary, the same mean value 1970 ± 15 c.p.m. was obtained.

This result supports our formula for equivalent atomic number, which does not distinguish isotopic forms by saturation backscattering experiments.

c) Aqueous solutions of salts

Aqueous solutions of salts in natural water exhibit an interesting feature, if we plot backscattering intensity against MÜLLER's atomic number. This can be seen in fig. 2 where log-log scales have been used. As figure shows, for metallic alloys, elements and salts a smooth curve may be drawn which satisfactorily fits

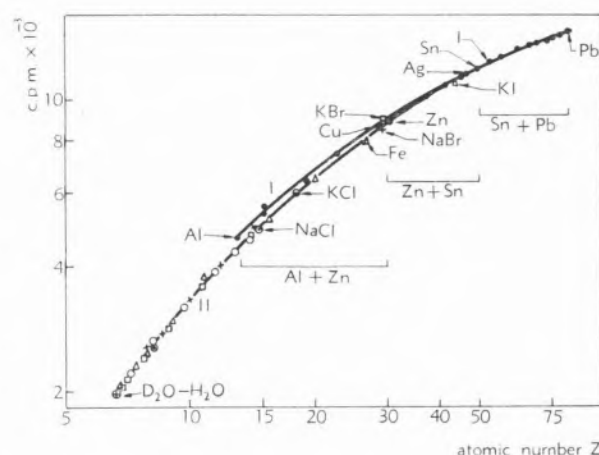


Fig. 3 — Correlation between intensity of backscattered beam and our equivalent atomic number with a source of ⁹⁰Sr + ⁹⁰Y. Symbols as in fig. 2.

Curve I: elements, metallic alloys and salts.

Curve II: KI and its aqueous solutions.

experimental points. On the other hand, each salt and its aqueous solutions follow a specific smooth curve, of course starting at *H*₂*O*. In fig. 2 only the curves corresponding to *NaCl*, *KI* and *KBr* had been drawn. The source was ⁹⁰Sr + ⁹⁰Y, but the same happens with ³²P.

⁽¹⁾ Chemical compositions have been determined by Centro de Química de Lisboa de Comissão de Estudos de Energia Nuclear under the direction of Prof. Eng. HERCULANO DE CARVALHO, to whom we are much indebted.

⁽²⁾ This sample of heavy water was kindly lent to us by Prof. Dr. K. JACOBSON.

Such a split of experimental points of aqueous solutions into different and specific curves, may be understood considering that the value $\bar{Z}_{H_2O} = 7.22$, given by MÜLLER, is too high compared to those of elements, metallic alloys and salts themselves.

In fig. 3 we plot the same measurements of backscattering against the equivalent atomic number, \bar{Z} , as given by our formulas.

Much better correlation is obtained, since $\bar{Z}_{H_2O} = 6.60$

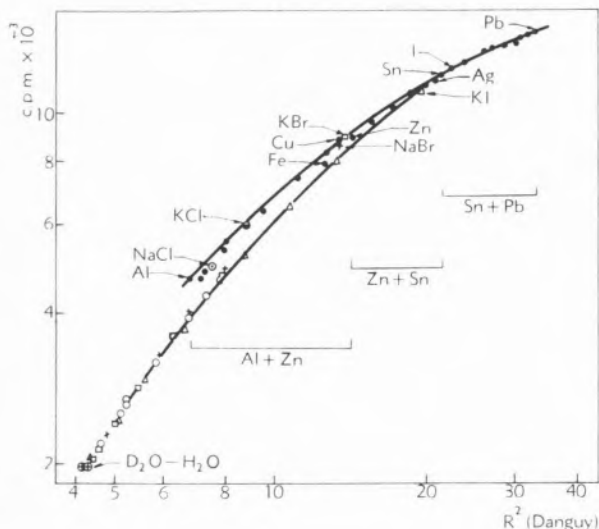


Fig. 4 — Correlation between intensity of backscattered beam and R^2 (Danguy) with a source of $^{90}\text{Sr} + ^{90}\text{Y}$. Symbols as in fig. 2.

Curve I: elements, metallic alloys and salts.

Curve II: KI and its aqueous solutions.

in our case, but experimental points of solutions are still below the mean curve corresponding to elements, metallic alloys and salts, though the difference is now of the order of 5 per cent or less, decreasing with decreasing concentration of water.

The curves corresponding to each salt and its solutions are rather close together and only the curve relative to KI and its solutions has been drawn in the figure, for that reason.

Taking in mind that our formulas 4 and 5 are based on several theoretical approaches, the agreement with experiments may be said to be satisfactory. Indeed, they predict that compounds with different isotopes of the same element should have the same saturation backscattering value, which was experimentally confirmed with natural and 99.8 % heavy water. Further-

more, the correlation intensity of scattered β beam — equivalent atomic number was experimentally verified within a few per cent, for a large number of elements, metallic alloys, salts and their aqueous solutions.

We think, therefore, that our formulas are better than those of MÜLLER.

Correlation with R^2 (DANGUY)

As we have already referred in paragraph 1, L. DANGUY proposed, instead of an equivalent atomic number, the quantity R , defined by equation 3.

In fig. 4 we plot intensity of backscattered β beam against R^2 , in log-log scales, for the case of a source of $^{90}\text{Sr} + ^{90}\text{Y}$. The square of R has been used in order to have, in ordinates, about the same dimensions of the preceding figures.

Fig. 4 shows that experimental points of solutions are, also, below the mean curve fitting elements, metallic alloys and salts, being splitted out into differentiated curves for each salt. Only the mean curve corresponding to KI has been drawn in the figure.

This result is similar to that obtained by using MÜLLER's values of \bar{Z} (see fig. 2), but the correlation, intensity

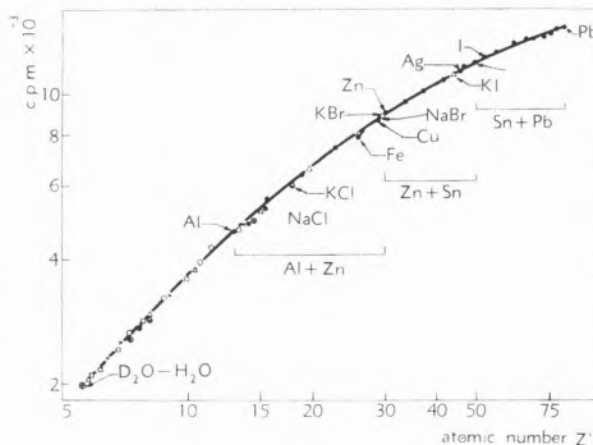


Fig. 5 — Correlation between intensity of backscattered beam and experimental atomic number, Z' , of water and salt solutions. Symbols as in fig. 2. The source was $^{90}\text{Sr} + ^{90}\text{Y}$.

— R^2 , is somewhat better since differences observed are of the order of 15 %, decreasing with decreasing concentration of water.

Values of R^2 for natural and 99.8 % heavy water are 4.22 and 4.19, respectively. Accordingly, the difference

between intensities of scattered beams would be about 20 counts per minute, in our system. This is of the order of experimental errors of measurements of H_2O and D_2O , which gave 1970 ± 15 c.p.m. in both cases. Regarding this specific case of isotopic forms, it might be somewhat hard to make a definite decision between DANGUY's formula and ours, if only experimental results were considered. However, theoretical considerations are in favour of our formulas, as it has been shown by one of us (7).

In general, a comparison between figs. 3 and 4 show that our experimental results are much better fitted when our value of Z is used instead of the DANGUY quantity R^2 .

2.4 — EXPERIMENTAL CORRECTION OF THE EQUIVALENT ATOMIC NUMBER OF WATER AND SALT SOLUTIONS

As fig. 3 suggests, the value 6.60 given by our formula for the equivalent atomic number of water, seems to be somewhat high, when compared with \bar{Z} values of elements, metallic alloys and salts.

This is qualitatively expected since the «effective» atomic number of water would be in between 6.60 and a value roughly predicted by equation 6, which gives 5.28.

As a consequence, \bar{Z} values of aqueous solutions are somewhat high as well, the disagreement decreasing with decreasing concentration of water.

We have tried to find an experimental value of \bar{Z}_{H_2O} , in order to obtain a better correlation between scattered beams and equivalent atomic numbers for water and salt solutions.

Let us consider the formula giving the atomic number, \bar{Z}_s , of a solution, easily deduced from equation 5:

$$\bar{Z}_s = \frac{c_1 \overline{(Z^2/A)}_1 + c_2 \overline{(Z^2/A)}_2}{c_1 \overline{(Z/A)}_1 + c_2 \overline{(Z/A)}_2} \quad (7)$$

Indices 1 and 2 refer to water and salt, respectively. Means values $\overline{(Z^2/A)}$ and $\overline{(Z/A)}$, for a general chemical compound $A_m B_n$, are given by

$$\overline{(Z^2/A)} = \frac{m Z_A^2 + n Z_B^2}{m A + n B} \quad (8)$$

$$\overline{(Z/A)} = \frac{m Z_A + n Z_B}{m A + n B} \quad (9)$$

Their ratio gives our equivalent atomic number, $\bar{Z}_{A_m B_n}$, of the compound (see eq. 4).

Now, if we assume that only the quantities $\overline{(Z^2/A)}_1$ and $\overline{(Z/A)}_1$, for water, are in error, the mean curve corresponding to backscattering with elements, metallic alloys and salts may be taken as a standard one, for comparison.

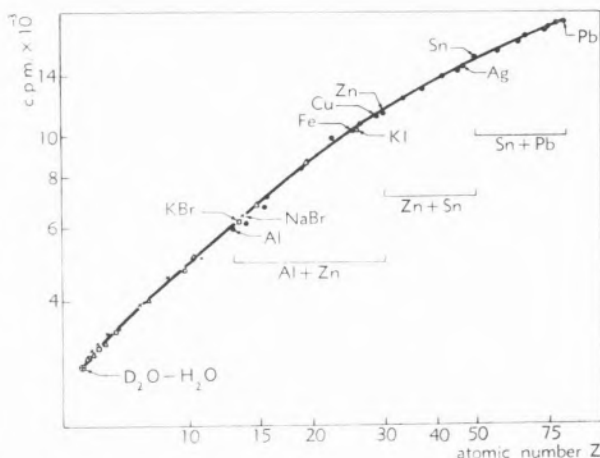


Fig. 6 — Correlation between intensity of backscattered beam and experimental atomic number, \bar{Z}' , of water and salt solutions.

Symbols as in fig. 2. The source was ^{32}P .

Therefore, by using this mean curve, we can graphically obtain the value of «correct» atomic numbers, \bar{Z}'_s , which should be attached to each salt solution, according to the intensity of its backscattered β beam. Having a sufficient number of experimental values \bar{Z}'_s , we identify them with \bar{Z}_s of equation 7, where $\overline{(Z^2/A)}_1$ and $\overline{(Z/A)}_1$, referring to water, are regarded as unknown quantities which we want to determine by the method of least squares.

To do so, our mean curve was somewhat extrapolated to lower values of the atomic number, in order to consider larger number of experimental equations for \bar{Z}'_s .

The mean experimental values are the following:

$$\overline{(Z^2/A)}'_{H_2O} = 3.60 \pm 0.03 \quad ; \quad \overline{(Z/A)}'_{H_2O} = 0.610 \pm 0.03$$

so that the experimental value of the atomic number of water is, accordingly

$$\overline{Z'}_{H_2O} = 5.90 \pm 0.08$$

whereas theoretical values are, respectively, 3.663, 0.555, and 6.60.

By using these experimental values for water, we can recompute now all the «corrected» equivalent atomic numbers, $\overline{Z'_s}$, for salt solutions, according to their

As figures show, good correlations are obtained which experimentally support the correction envisaged for water and salt solutions.

Curves of figs. 5 and 6, have slightly different slopes for the same value of atomic number. The ratio between the intensities backscattered by lead ($Z = 82$) and by water ($Z' = 5.90$) is equal to 6.80 for β particles of ^{32}P , with maximum energy of 1.71 Mev, and equal to 7.32 for $^{90}Sr + ^{90}Y$ whose maximum energies are, respectively, 0.54 and 2.27 Mev.

Table I

SATURATION BACKSCATTERING IN COMPLEX TARGETS WITH A SOURCE OF $^{90}Sr + ^{90}Y$

Target	Values of Z/A	Percentage (*)	Equivalent atomic number			R^2 DANGUY (empirical)	Backscattered beam (c.p.m.)
			MÜLLER (empirical)	Ours (theoretical)	$Z'(\text{ours})$ (experim.)		
$Al + Zn$	$Al = 0.482$	84.3	15.65	15.55		7.90	5332 ± 51
		63.0	19.28	19.06		9.50	6422 ± 31
	$Zn = 0.459$	21.2	26.49	26.31		12.65	8187 ± 39
$Zn + Sn$	$Sn = 0.421$	80.1	33.96	33.73		15.61	9620 ± 60
		60.1	37.94	37.51		17.10	10266 ± 40
$Sn + Pb$	$Pb = 0.396$	60.0	62.80	62.33		26.02	13362 ± 57
		38.6	69.64	69.10		28.45	13659 ± 42
		9.8	78.86	78.73		31.72	14335 ± 84
KI	$I = 0.418$	8.15	10.30	9.03	8.16	5.47	2935 ± 13
	$K = 0.486$	42.21	23.16	20.20	18.93	10.69	6525 ± 24
KBr	$Br = 0.438$	11.47	9.80	8.78	7.96	5.31	2844 ± 14
		37.19	15.59	14.03	13.07	7.77	4740 ± 68
$NaBr$	$Na = 0.478$	6.82	8.75	7.86	7.09	4.85	2513 ± 35
		39.15	16.00	14.34	13.38	7.90	4879 ± 31
$BaCl_2$	$Cl = 0.479$	17.83	13.55	11.61	10.61	6.72	3904 ± 19
	$Ba = 0.408$						
$NaCl$		25.31	9.10	8.42	7.74	5.05	2720 ± 33
KCl		22.38	9.64	8.90	8.16	5.33	2860 ± 14
H_2O	$H = 0.992$	natural	7.22	6.60	5.90	4.22	1970 ± 15
	$D = 0.496$						
D_2O	$O = 0.500$	99.8	6.80	6.60	5.90	4.19	1970 ± 15

(*) Percentage by weight of the lighter element in the alloy or of the salt in the solution.

weight fractions. In table I they are given in column 6. For elements, metallic alloys and salts, \overline{Z} values coincide, of course, with $\overline{Z'}$.

In figs. 5 and 6, we present against $\overline{Z'}$, in log-log scales, the intensities of backscattered β beams obtained with sources of $^{90}Sr + ^{90}Y$ and ^{32}P , respectively.

This result cannot be explained by different absorptions of β particles of ^{32}P and $^{90}Sr + ^{90}Y$ in the air and in the window of G. M. counter which correspond to an over all thickness of about 3 mg.cm^{-2} . It may mean a small dependence of saturation backscattering with energy of incident β particles but we are not in position

to make a definite statement, since we have to take into account, for example, the possibility of different anisotropies of backscattered beams (13) because we have not used a measuring solid angle of 2π or, as well, the possibility of a small effect of bremsstrahlung radiation.

From experiments of several authors (5, 11, 12) it appears that saturation backscattering factor, i.e. the ratio of the total backscattered and incident β beams, would be independent of the maximum energy of emitters, in a range which extends from 0.15 Mev up to 3 Mev.

3 — CONCLUSION

We concluded that our experiments on saturation backscattering of β particles are much better fitted when we use our formulas 4 and 5, for the equivalent atomic number of complex targets, instead of those of R. H. MÜLLER and L. DANGUY.

This is specially emphasised for targets containing hydrogen as, for example, the cases of water and aqueous solutions of salts which have been observed. MÜLLER's formulas 1 and 2 are definitely inadequate and the DANGUY equation 3, though somewhat better, gives rise to appreciable differences when the results of aqueous solutions are compared to those obtained with elements, metallic alloys and dry salts.

By using our formulas for the equivalent atomic number, differences of a few per cent are still observed in aqueous solutions. Since they may be attributed to water, an experimental correction has been envisaged, assuming the mean curve corresponding to elements, metallic alloys and salts as a standard one. A good correlation between the intensity of backscattered β beam and the atomic number is then obtained.

Our experiments show that natural and 99.8% heavy water backscatter identically, within experimental errors. This result supports again our formulas which give the same value for the equivalent atomic number of compounds with different isotopes.

RESUMO

Estudámos o efeito da retrodifusão de partículas β em condições de saturação, utilizando fontes de ^{32}P e $^{90}\text{Sr} + ^{90}\text{Y}$ e alvos constituídos por elementos, ligas metálicas, sais e respectivas soluções aquosas, H_2O e D_2O a 99,8%. Verificou-se que existe uma correlação entre a intensidade do feixe retrodifundido e o número atómico equivalente proposto por Saldick e teoricamente confirmado por um de nós. A água natural e a pesada retrodifundem análogamente, diferindo, no entanto, ligeiramente do comportamento geral dos elementos, ligas e sais. Como consequência, propomos uma correcção para o valor de \bar{Z} da água, a qual é, igualmente, aplicável às soluções aquosas.

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