A Nuclear Probe in Bioinorganic Chemistry – Sum Peak Intensity Measurements Applicable to the Study of Biological Macromolecules

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Chemical effects of sum peaks in the spectra of γ rays emitted from radioactive nuclides have not been studied except for very few papers [1, 2] of a preliminary nature.

It was found that sum peaks have an intense interrelation to perturbed angular correlation (PAC) in various chemical systems. Sum peak measurements can be carried out without using a complicated coincidence circuit and without applying an intense source of radioactivity. An ordinary γ -ray spectrometer with Ge(Li) detector is the only equipment required by this method.

A sum peak appears when two γ -rays emitted from the cascade decay of a radioactive nucleus enter in the Ge(Li) detector simultaneously. The peak corresponds to the sum of energies of both γ -rays in the energy scale.

The ratio (R) of the intensity of a single peak $(I_{\gamma 1})$ to that of the sum peak (I_{sum}) is expressed as follows:

$$\mathbf{R} = \mathbf{I}_{\gamma 1} / \mathbf{I}_{sum} = \{ \epsilon_{\gamma 2} \cdot \overline{\mathbf{W}(\theta)} \cdot \mathbf{f} \}^{-1}$$
(1)

where $\epsilon_{\gamma 2}$ is the efficiency of the Ge(Li) detector for γ_2 , $\overline{W(\theta)}$ is the average angular correlation coefficient of the source for the given geometry to the detector, and f is a factor equal to $\{W(0^\circ) + W(90^\circ)\}/2$. $\overline{W(\theta)}$ and f depend on the chemical and/or physicochemical states of the source and therefore R varies with the state of the source. This fact is closely related to the phenomena of perturbed angular correlation (PAC), although detailed information on the chemical and/or physicochemical effects of the sum peak is lacking.

In this paper, an application of this method has been made to check the range of biological materials by only measurement of the R value. Among many nuclides, ^{152,154}Eu may be one of the most appropriate nuclides, because some biological influences were detected by the ordinary PAC method using ^{152,154}Eu in our laboratory [3]. Figure 1 shows decay schemes for the radioisotopes. ^{152,154}Eu was added to an aqueous solution (10 mg/ml) of albumin (BSA) which is purified from bovine serum by the Sanko Jun-yaku. On mixing the solution remained transparent showing no change (no precipitate or



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Figure 1. Decay scheme for ^{152,154}Eu.



Figure 2. Measured γ -ray spectrum of a 152,154 Eu sample.

turbidity). An aliquot of the resulting mixture was measured by a 40 ml Ge(Li) detector connected with a 4K PHA (Toshiba USC-1 model 10). A measured γ -ray spectrum is shown in Fig. 2. Among many γ -ray peaks in the figure the peaks due to 1408–122 keV cascade and its sum peak at 1530 keV are the most appropriate ones for this method. It is noticed that background counts are relatively low for the peaks of 1408 and 1530 keV. Therefore the value of R = $I_{1498 \text{ keV}}/I_{1530 \text{ keV}}$ is adopted in the experiment.

The next step was the destruction of albumin by addition of hydrochloric acid to the above mixture of albumin and europium. BSA was destroyed by applying drastic conditions (6 N HCl).

The above procedures are listed in Fig. 3. The measurement of R was done for each step of the procedures in Fig. 3. The results of the measurement are shown in Table I. It is clearly shown that there is a definite difference of R between Steps 1 and 2 showing that europium is bound to albumin. The value of R again increase in the solution of 6 N HCl, corresponding to the destruction of the bond between europium and albumin.

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(Distruction of albumin)

Figure 3. Chemical procedure for an europium-albumin system in solution.

This shows that the sum peak method is of value for studying changes of the bound state between biological macromolecules and metallic ions.

TABLE I. Change of R-value for Various Conditions in the Sum Peak Method.

Step	Sample	R I ₁₄₀₈
1 2 3	EuCl ₃ in dil. HCl Albumin soln. + *Eu Albumin soln. + *EU in 6 N HCl	⁻ sum 19.48 ± 0.11 17.88 ± 0.10 19.38 ± 0.10

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