

1-PAC: A one-detector gamma-gamma perturbed angular correlation technique for the determination of physical-chemical properties of radioelements: Hydrolysis of radioindium

DV Filosofov*, AF Novgorodov*, NA Korolev*, VG Egorov*, NA Lebedev*, ZZ Akselrod†, J Brockmann‡, F Rösch‡

*Joint Institute of Nuclear Research, LNP, 141980 Dubna, Russia; †Skobeltsyn Institute of Nuclear Physics, Moscow State University, 119899 Moscow, Russia; ‡Institut für Kernchemie, Johannes Gutenberg-Universität Mainz, Germany

Introduction: As shown in [1], the ratio of sum peaks (SP) intensities S compared to intensities of photopeak and roentgen emissions may be used to determine gamma-gamma perturbed angular correlations using one detector only. It is assumed that the relative probability of recording of cascade γ -rays $R_{\gamma_n+\gamma_m}$ in TSP at the individual values of A_{ii} , $\overline{G}_i(\infty)$ and Q_i is equal to TIAC $\overline{W}(\theta, \infty)$, where A_{ii} is the angular correlation coefficients depending on spins and multiplicities of transitions; $P_i(\cos\theta)$ are the Legendre polynomials; Q_i are the solid angle correction factors taking into account the detector and source finite sizes; and $\overline{G}_i(\infty)$ is the TIAC perturbation factor which yields information on hyperfine interactions. The following calculations were applied.

$$R_{\gamma_1+\gamma_2} = N_{K_{\alpha,\beta}+\gamma_1} \cdot \frac{S_{\gamma_1+\gamma_2} \cdot S_{K_{\alpha,\beta}}}{S_{\gamma_2} \cdot S_{\gamma_1+K_{\alpha,\beta}}} \text{ or } R_{\gamma_1+\gamma_2} = N_{K_{\alpha,\beta}+\gamma_2} \cdot \frac{S_{\gamma_1+\gamma_2} \cdot S_{K_{\alpha,\beta}}}{S_{\gamma_1} \cdot S_{\gamma_2+K_{\alpha,\beta}}}$$

with $N_{K_{\alpha,\beta}+\gamma_1} = 0.9176$ and $N_{K_{\alpha,\beta}+\gamma_2} = 0.9507$.

The first observations of the sum peak intensity change due to chemical environment was reported for ^{181}Hf compounds by De Bruin et al [2] and for ^{111}In compounds by Yoshihara et al [3,4]. However, proposed methods had not received practical applications. In the present work it is proposed to utilize the SP in the γ -spectrum from the radionuclide ^{111}In obtained with one detector to define the physico-chemical state of the hydrolysis of $\text{In}^{(III)}$.

Experiments: Measurements for ^{111}In have been carried out with a coaxial HPGe detector of a volume of 200 cm^3 . The radioactivity of the samples amounted to $\approx 40 \text{ kBq}$ resulting in a count rate of ≈ 8000 pulses/s at the selected distance of 37 mm from the surface of the detector. Within the time of 30 min of registration, the events accumulated in TSP were in the order of $\sim 10^5$. For the calculations the decay data of the radionuclides given in [5,6] were used. No-carrier-added ^{111}In was purchased from Amersham. Samples were prepared from high-grade pure chemicals (HNO_3 , NaOH , NaNO_3 , $\text{In}(\text{NO}_3)_3$, deionised water). Carrier concentrations of 10^{-5} M for In were added to the samples. The ionic strength was kept constant ($\mu=0.1$). The volume of the samples was $500 \mu\text{l}$ using polyethylene Eppendorf vials. The measurements were carried out at room temperature. The obtained data are summarized in Fig. 1.

Results: For the first series of measurements the pH was changed from 1 to 13 without adding DTPA. The $R_{\gamma_1+\gamma_2}$ value significantly varies between $\text{pH}=5$ and $\text{pH}=7$, indicating the transition from $[\text{In}(\text{H}_2\text{O})_6]^{3+}$ to the non-soluble form of $\text{In}(\text{OH})_3$. This shift corresponds to the hydrolysis equilibrium of indium [7]. The difference of $R_{\gamma_1+\gamma_2}$ values in the acidic and alkaline media is due to the differences of $\overline{G}_2(\infty)$ for the $[\text{In}(\text{H}_2\text{O})_6]^{3+}$ ("liquid" state, $\overline{G}_2(\infty)=1$) and $\text{In}(\text{OH})_{3(\text{solid})}$ ("solid" state, $\overline{G}_2(\infty)=0.2$).

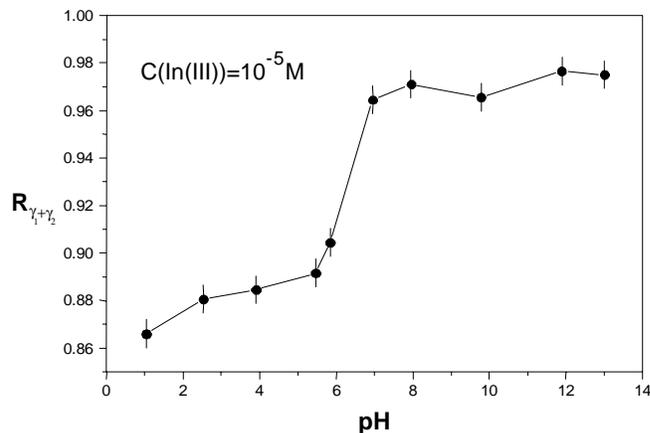


Fig. 1. The $R_{\gamma_1+\gamma_2}(\text{exp})$ dependencies for ^{111}In in aqueous solutions of different pH as measured by means of 1-PAC method.

Conclusion: It was shown that it is possible to measure the perturbation of an angular correlation of a suitable radionuclide depending on its physico-chemical state with just one detector. The 1-PAC method allows to study properties of the condensed matter at submicro-level concentration using a simple and widespread instrumentation. On the basis of defined $R_{\gamma_n+\gamma_m}$ values and of the

dependence of perturbation of an angular correlation on an electronic constitution of matter, parameters of an elements hydrolysis equilibrium can be analysed. Other chemical equilibria, temperatures of phase changes, kinetics of chemical reactions and a number of other properties might be determined as well.

It was estimated in the experiment performed with the large-volume samples ($V_{\text{sample}} \geq V_{\text{detector}}$) that relative changes in $R_{\gamma_n+\gamma_m}$ are just slightly different from that for point-like samples.

This allows to apply the 1-PAC method even for investigation of biological samples and objects "in vitro" and "in vivo".

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