Investigation of the DTPA complex formations of indium and cadmium by a γγ-perturbed angular correlation method

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Introduction:
For qualitative and quantitative speciation of radioelements, the time-integral perturbed angular γγ-correlation (γγ-IPAC) measuring the γγ-IPAC response ($G_2(\infty)$ = time-integral perturbation factor (TIPF)) may provide rich information about the closest chemical environment of a radioactive nuclide [1,2]. The main experimental advantages are:

- ability to study systems in any aggregate state;
- non-invasive character of measurement;
- very small amounts (activity) of the radioactive nuclide required (~300 kBq);
- thus low range of species concentrations (up to 10^{-12} M);
- small volumes of the sample (1 μl – 100 ml);
- wide ranges of temperature and pressure;
- simple equipment due to the development of a γγ-IPAC technique using one HPGe-detector only (1-PAC) and
- simplicity of data interpretation of the TIPF for each chemical state of the radionuclide.

Materials and Methods:
This method was applied to study DTPA complex formation equilibrea of indium and cadmium represented by the radioisotopes ^{111}In and ^{111m}Cd. Measurements have been carried using a 4-detector spectrometer with BaF_2 scintillators.
Nca ^{111}In was produced via the nuclear reaction ^{109}Ag(α,2n)^{111}In. The ^{111m}Cd was prepared from a ^{111}In/^{111m}Cd radionuclide generator.
To the samples containing about ≈100 kBq of the individual radionuclides, DTPA was added in concentration of $3\cdot10^{-5}$ M, with an overall pH ranging from 0.5 to 13.5. The ionic strength was kept constant (μ = 0.30, $HClO_4$, $NaClO_4$, NaOH).

Results:
For pH=0.5 solutions of ^{111}In, the TIPF was found to be 1.00(3), corresponding to the aqua-complex of $[In(H_2O)_6]^{3+}$. At pH > 0.6 the value of $G_2(\infty)$ approached 0.50(3), reflecting the In(DTPA) complex formation. From those data we determined the equilibrium constant for the $[InDTPA]^{2-}$ complex of logβ = 27.2.
In the case of ^{111m}Cd at pH = 0.5-2, the TIPF was found to be 1.00(3). In less acidic solutions of pH > 2.2, $G_2(\infty)$ changes to 0.40(3). The equilibrium constant for $[CdHDTPA]^{2-}$ complex was determined to logβ = 13.6.
It was shown that the time-integral perturbed angular γγ-correlation method is very convenient to study complex formation equilibrea of radioelements.

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References

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