

ELECTROPHORESIS OF TRACE QUANTITIES OF YTTRIUM*

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Electrophoresis of aqueous solutions of ^{91}Y in trace concentrations (about 10^{-11} – 10^{-12}M) was studied over the pH range 1.6–11.6, and at 25°C, using unscreened electrodes and electrodes screened by a cellophane membrane. It was found that the portion of positively charged forms of ^{91}Y in all (positively and negatively), charged forms of this nuclide exhibited an over-all decrease with increasing pH from very high values in the strongly acid region to very low values in the alkaline region. This dependence showed a deep minimum at pH 3–4 and a maximum at pH 4–5. The portion of cationic forms of ^{91}Y in all charged forms of this nuclide decreased with increasing pH from very high values in strongly acid solutions to a minimum at pH 4–5; over the region pH 5.6–10.4 it was approximately constant and close to 50%. A sudden increase of the portion of negatively charged forms of ^{91}Y with increasing pH, accompanied by a simultaneous abrupt decrease of anions in solutions of pH > 10.4 is undoubtedly connected with the formation of a negatively charged colloid of yttrium hydroxide.

The importance of the electrophoretic and electrolytic method in investigation of trace quantities of radionuclides was pointed out by Starik¹.

EXPERIMENTAL

The chemicals in question, the radionuclide and its solutions, as well as the sorbent, the apparatus, and the procedures used in determining the activity and pH of the solutions have been described earlier². A U-shaped polyethylene tube was used with the following parameters: length 32 cm; inside diameter 15 mm; effective volume 50 ml. Platinum electrodes were immersed into the solution through the tube's upper ends. In some experiments the electrodes were screened from the main volume of the tube by cuvettes made of polyethylene tubings, 15 mm long, of inside diameter 8 mm. Their bottom was a circular window with glued-over cellophane 0.027 mm thick; the pore diameter of the cellophane membrane was of the order of μm , as determined from bubble pressure measurements³. The tube was filled up with the solution of the radionuclide and tempered to $25 \pm 0.1^\circ\text{C}$. The cuvettes were then inserted into the solution. They were filled with the solution of the same composition, same pH, and same temperature, however, without the radionuclide. Thereafter, the current through the tube was adjusted to 2 mA; the potential difference was 110 V. In definite time intervals samples of the solution were taken from the cuvettes and their

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activity was measured. An equilibrium in particle migration was established within two hours since the electrolysis started. In this way the migration of radionuclide forms towards the screened electrodes was investigated. When the migration of radionuclide forms towards the unscreened electrodes was studied, the solution was sampled from the immediate vicinity of the electrodes, in this case immersed directly into the solution. The migration of radionuclide forms was in both cases expressed as a percent ratio of the activity of either positive (A^+) or negative (A^-) forms to the sum of both activities, ($A^+ + A^-$). For particular measurements at a particular pH value the percentage of the corresponding forms was calculated using the relations

$$N_i^+ = 100 A^+ / (A^+ + A^-) (\%), \quad (1)$$

$$N_i^- = 100 A^- / (A^+ + A^-) (\%). \quad (2)$$

Mean values of N^+ and N^- plotted in figures are averages of at least three measurements. All measured N_i^+ and N_i^- fell within $\pm 10\%$ of the mean values.

RESULTS AND DISCUSSION

The percentage of all positively charged forms of ^{91}Y from all (positively and negatively) charged forms of this nuclide showed an over-all decrease with increasing pH (Fig. 1, curve 1) from very high values in strongly acid solutions to relatively small values in alkaline solutions. The dependence $N^+ = f(\text{pH})$ exhibits a deep minimum at pH 3–4, and a maximum at about pH 4–5. It is clear from the published data¹

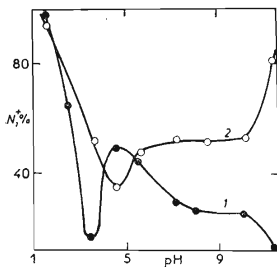


FIG. 1

Dependence of Percentage N^+ of Positively Charged Forms of ^{91}Y from Total Amount of Electrically Charged Forms ^{91}Y in Aqueous Solutions upon pH of the Solutions

Electrodes: 1 unscreened; 2 screened by cellophane membrane.

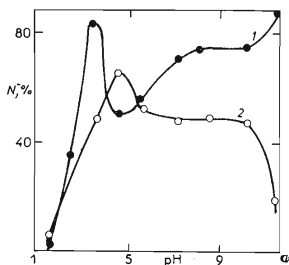


FIG. 2

Dependence of Percentage N^- of Negatively Charged Forms of ^{91}Y from Total Amount of Electrically Charged Forms ^{91}Y in Aqueous Solutions upon pH of the Solutions

Electrodes: 1 unscreened; 2 screened by cellophane membrane.

that the positively charged forms of radionuclides of metals in aqueous solutions (if present in trace amounts) can be either simple and complex cations, or adsorption pseudocolloids. Of these forms, only the cations can penetrate through the celophane pores of the μm order-of-magnitude size. Their portion in all ionic forms of ^{91}Y decreases with increasing pH (Fig. 1, curve 2) from very high values in strongly acid solutions towards a minimum at pH 4–5. This portion is roughly constant over the pH interval 5.6–10.4 and it is close to 50%. At pH > 10.4 again a substantial increase of the percentage of cationic forms relative to all ionic forms occurs, though it has to be kept in mind that the cationic forms represent in this region a relatively minor fraction of all positively charged forms of ^{91}Y (Fig. 1, curve 1).

Because it follows from (1) and (2) that $N^+ + N^- = 100\%$, the dependences $N^- = f(\text{pH})$ in Fig. 2 are mirror images of the analogous dependences $N^+ = f(\text{pH})$ in Fig. 1. Therefore, complementary statements to those concerning the positively charged forms of ^{91}Y can be formulated concerning the negatively charged forms of this nuclide, and *vice versa*.

The decrease in the relative percentage of positively charged forms of ^{91}Y with pH increasing (Fig. 1, curve 1) up to pH 3–4 may be explained by a decrease of cationic and an increase of anionic forms of this nuclide (curves 2 in Figs 1 and 2). The following increase of percentage of positively charged forms of ^{91}Y as pH increases to 4–5 may be explained by a formation of positively charged products of hydrolysis of ^{91}Y compounds which are adsorbed to a considerable extent on solid impurities in the solution (mostly microscopic particles of silicic acid gels eluted from the glass surface of the distillation vessel, transfer ampules *etc.*). In this way, pseudocolloids of the radionuclide are formed. A favourable factor in the formation of adsorbed pseudocolloids is that at about pH 4.1 there is a pronounced adsorption maximum of ^{91}Y over the glass surfaces² from which the active "carrier" of the pseudocolloids forms of the radionuclide originates. The positive sign of the over-all charge of these particles is given by a predominance of positively charged forms of ^{91}Y adsorbed on the surface of the "carrier" over a negative charge of the "carrier" itself in aqueous solutions. With increasing pH the polymerization of hydrolytic forms of ^{91}Y continues. As a result, the positive charge of these polymers, related to one atom of yttrium, decreases. At pH > 4–5, therefore, the negative charge of the non-active "carrier" begins to predominate over the positive charge of the ^{91}Y forms adsorbed on it, and the percentage of negatively charged pseudocolloidal particles increases (Fig. 2, curve 1).

Even in the region pH 4–5 the minimum of the percentage of negatively charged ^{91}Y forms reaches the value 50% (Fig. 2, curve 1). Thus, even in this region the co-existence of negatively and positively charged pseudocolloids is possible. The occurrence of the minimum of cationic forms at pH 4–5 (Fig. 1, curve 2) evidently hangs together with the maximum of the percentage of positively charged forms in the same pH region (Fig. 1, curve 1), as a substantial amount of cations is con-

sumed under these conditions to form positively charged adsorption pseudocolloids. A sharper increase of negatively charged forms of ^{91}Y with a simultaneous sudden decrease of anions at $\text{pH} > 10.3$ (Fig. 2) has to be ascribed to the formation of a true colloid of $^{91}\text{Y}(\text{OH})_3$. It was calculated² that its presence over the pH region pH 9–11 has to be expected. It is well known that the true colloids of multivalent metal hydroxides are usually negatively charged in aqueous solutions^{1,4}.

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