

RADIOCHEMICAL STUDY
OF THE SORPTION OF TRACE ELEMENTS. VII.*
ADSORPTION OF YTTRIUM
ON POLYETHYLENE AND THE EFFECT
OF LOW CONCENTRATIONS OF IRON

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The paper describes adsorption and desorption of traces of yttrium ($< 10^{-9} - 10^{-5} M$), labelled with ^{88}Y , on polyethylene in relation to pH, composition of the solution, concentration of yttrium and time. The results show that the adsorption increases with pH, beginning from pH 3.5 to 5. At pH > 6 it exhibits a flat peak, the width of which is given mainly by the age of the solution. Indifferent electrolytes suppress sorption of the lowest quantities of yttrium up to pH 8; beyond this value, by contrast, they enhance the sorption. On the basis of the experimental evidence a possible mechanism of the sorption is proposed. By studying the interaction of trace yttrium and iron it has been found that iron affects the sorption of yttrium if the latter co-precipitates with colloidal ferric hydroxide. In a given range of pH the adsorption of yttrium can essentially be governed by sorption of its colloidal carrier.

The adsorption of traces of yttrium from aqueous solutions on hydrophobic organic polymers has already been investigated¹⁻⁶, but the authors have considerably different views on its explanation. Mellish, Payne and Worrall¹ studied the adsorption from specially purified solutions on polystyrene in relation to the surface finish of the adsorbent and time at different values of pH. They interpreted the results as an ion exchange on inorganic inclusions or on some foreign organic groups on the surface of the polymer. Kirby² explained the adherence of yttrium to teflon and other materials after evaporation of the solvent as due to a physical adsorption of undissociated molecules of yttrium compounds. Švehla measured contamination and decontamination of synthetic paints with yttrium as a function of pH and composition of the solution^{3,4}. He assumes that a prerequisite for the sorption of yttrium is the formation of true colloid particles of yttrium hydroxide. The kinetics of the sorption of yttrium on polypropylene from distilled and hard water in relation to pre-saturation of the surface have been studied by Eichholz, Nagel and Hughes⁵. Høgdahl⁶ investigated adsorption and desorption of yttrium from sea water on polyethylene and polypropylene as a function of pH, composition of the solution and time.

The data on the adsorption of yttrium in the cited papers are largely incomplete or refer to conditions considerably different from the usual practice in chemical

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laboratories (composition of the solutions, experimental arrangement). To get a better insight into the regularities of this adsorption under the normal laboratory conditions we have studied more systematically the adsorption of traces of yttrium on polyethylene in relation to pH, composition of the solution, concentration of yttrium and time. Previously we have shown⁷ that the state of yttrium in solutions is considerably affected by traces of iron. Since iron is a most frequent impurity in water as well as chemicals^{8,9} it was of interest to investigate its effect on the adsorption of yttrium on polyethylene. This study was facilitated by our previous work¹⁰ on the sorption of iron on polyethylene.

EXPERIMENTAL

Chemicals and Apparatus

All the stock solutions were prepared using three times distilled water. The stock solution of 0.01M-HCl was prepared by dilution of the commercial concd. hydrochloric acid, A. R. This was found¹¹ to contain ferric cations as an impurity in a concentration of about $3 \cdot 10^{-7}$ M. Solutions of sodium hydroxide (0.1M and 1M) were obtained by dilution of 50% NaOH, A. R. A solution of ammonium hydroxide was obtained by isothermal distillation of ammonia into 0.01M-HCl. The other stock solutions were prepared by dissolution or dilution of A. R. chemicals.

The preparation of ⁸⁸Y (γ -emitter, half life 105 d) was supplied by the Radiochemical Centre, Amersham, in the form of ⁸⁸YCl₃, carrier-free in 0.1M-HCl. Prior to use this solution was diluted with 0.01M hydrochloric acid. In experiments with higher concentrations of yttrium a standard solution of non-active yttrium was added in such a manner that an isotope equilibrium was established in the solution. Some control experiments with iron were carried out with a preparation of ⁵⁹Fe (Zentralinstitut für Kernforschung, Rossendorf ü. Dresden) in the form of FeCl₃ (25 mCi/mg Fe in 0.01M-HCl); the stock solutions were prepared analogously.

All the vessels and pipettes had been carefully washed in a hot mixture of concd. HCl and HNO₃ (1 : 1), then in three times distilled water. pH of the solutions was measured with a glass electrode and a Multoscop pH-meter (Laboratorní potřeby, Prague). Radioactivity was measured with a scintillation counter SČ-4 (U.S.S.R.) and a crystal of NaI(Tl) as detector. The samples (5 ml) in glass ampoules were inserted into a hole in the crystal.

Procedure

The adsorption was studied on the inner surface of 30 ml polyethylene bottles (Plastimat, Liberec); the inner surface area of a bottle, including the stopper was 74 ± 1 cm². Prior to use the bottles were washed in water (three times distilled) or they were cleaned by desorption in 1M-HCl. It had been found that exposure of their surface to the acid or 1M-NaOH had no appreciable effect on the adsorption.

The solutions employed for studying the adsorption were prepared either directly in the sorption bottles or separately in greater amounts in glass or polyethylene flasks. In the latter case the pH was measured and the solution was employed either within 3 hours of its preparation or 7 days later. The solutions contained NaCl (NH₄Cl) in concentrations $\leq 10^{-2}$ M and traces of iron (c. $3 \cdot 10^{-7}$ M). Unless the isotopic carrier for yttrium was added the concentration of yttrium in view of its low abundance was probably lower than 10^{-9} M. For the sake of brevity these solutions will hereafter be referred to as carrier-free. Where the effect of sodium chloride or higher

amounts of trivalent iron on the adsorption was studied these substances were added to the solution either before the addition of radioyttrium or together with it. All solutions were prepared and kept in a dustproof box.

In an experiment 10 ml of a solution was shaken in a closed polyethylene bottle for 15 hours at room temperature. The adsorption was then evaluated from the difference in activity before and after the adsorption. After each experiment the pH of the solution was measured again. Following the experiments with adsorption the empty bottles were employed to study the desorption of yttrium. They were washed with a small amount of water, then 10 ml of the desorbing solution was added to each and the bottles were agitated for 15 hours. The time between the starts of the adsorption and the desorption was 24 hours and the desorption was evaluated indirectly by measuring the radioactivity of the solution after the desorption. The procedure and evaluation of the adsorption and desorption was described previously¹¹.

RESULTS AND DISCUSSION

The results in this paper are largely averages of 3 or more measurements (as many as 20). To illustrate the spread of the data the mean square deviations are plotted at some of the points.

Figs 1 and 2 show the adsorption of the carrier-free yttrium in relation to pH, composition and age of the solutions. The curves in Fig. 1 reveal that up to pH 3.5 the adsorption is negligible, then it rapidly increases up to pH 6.7. At higher pH values the adsorption either no longer changes or decreases. In the region of the increase of the adsorption the nature of the hydroxide employed for the adjustment

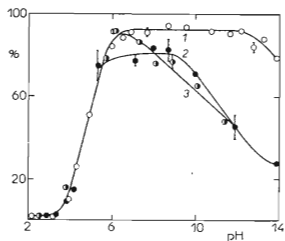


FIG. 1

Adsorption of Carrier-Free Yttrium on Polyethylene in Relation to pH, Age and Composition of the Solution

1 Fresh solutions, pH adjusted with sodium hydroxide; 2 7 days old solutions, pH adjusted with sodium hydroxide; 3 fresh solutions, pH adjusted with ammonium hydroxide.

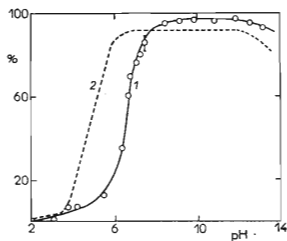


FIG. 2

Adsorption of Carrier-Free Yttrium on Polyethylene in Relation to pH and Composition of the Solution

1 1M-NaCl; 2 ≤ 0.01 M-NaCl (see curve 1 in Fig. 1).

of pH has no effect on the adsorption. At $\text{pH} > 7$ the adsorption is lower in the medium of ammonium chloride than in a solution of sodium chloride. With the aging of the solution the adsorption of yttrium (above pH c. 5.5) decreases. An increase in concentration of the electrolyte suppresses the adsorption in acid solutions, but enhances it in alkaline solutions (Fig. 2).

The interpretation of these results can proceed from the findings on the state of the traces of yttrium in the solutions employed, obtained by studying their dialysis, centrifugation and electrophoresis⁷, and from the present-day ideas on the mechanism of sorption of trace elements on hydrophobic organic polymers¹⁰. In acid solutions ($\text{pH} < 3.5$) the trace yttrium occurs in the form of Y^{3+} cations; these are sorbed very little because the concentration of protons in the solution is considerable and the ion-exchange activity of foreign groups on the surface of polyethylene is negligible. Beginning from pH 3 the surface of polyethylene assumes a negative charge,* which is probably due to sorption of OH^- ions (judging by the analogy to teflon^{12,13}). Its negative charge increases with pH and the concentration of protons decreases. These facts enhance the adsorption of Y^{3+} ions, proceeding by an ion exchange in the electric double layer on the surface of polyethylene. However, the increase in adsorption can also be explained in a different way. It was found⁷ that at $\text{pH} > 3.5$ the carrier-free yttrium is adsorbed by an ion exchange on the colloid impurities in the solution, thus giving rise to positively charged pseudocolloids. It is possible that these particles get sorbed on polyethylene, or that the inorganic impurities contained in polyethylene or occluded by it operate as centres of the ion exchange. The latter explanation is in a better accordance with some findings on the inhomogeneity of the sorption¹.

Either interpretation is supported by the negative effect of indifferent electrolyte on the adsorption in the pH region from 3.5 to 6.5. For it can be expected that sodium cations in a sufficient concentration will suppress the ion exchange both in the electric double layer on polyethylene and on the colloid impurities. Suppression of the formation of pseudocolloids of yttrium by the action of sodium chloride has been demonstrated experimentally⁷.

In solutions of pH 6 to 9.5 co-precipitation can occur of the carrier-free yttrium with colloidal ferric hydroxide, present in a concentration of about $3 \cdot 10^{-7} \text{M}$ (see⁷). In this region of pH it is probable that great part of yttrium is bound to polyethylene together with its colloidal carrier by physical adsorption, since physical adsorption of ferric hydroxide alone to polyethylene is considerable in this pH range¹⁰. It cannot be ruled out, however, that other mechanisms of sorption are operative simultaneously; it is the ion exchange up to pH 8 (*cf.* the decrease in the adsorption effected by the electrolyte) and, at higher values of pH , chemisorption

* *Note added in proof:* Using the streaming-potential technique, we have recently found that the negative charge on polyethylene increases from pH c. 2.8 to pH 8.

or physical sorption of the products of yttrium hydrolysis. The latter mechanism probably prevails at $\text{pH} > 10$, where the adsorption of ferric hydroxide is low¹⁰ and the solution contains mainly the hydrolysed species of yttrium partially sorbed to the colloidal impurities⁷. This interpretation is supported by the increase in adsorption above $\text{pH} 8$ caused by the addition of an indifferent electrolyte, which is characteristic of physical sorption or chemisorption and is usually ascribed to a "salting out" or coagulation effect of the electrolyte^{10,14,15}. A similar increase was observed in studying the adsorption of traces of ferric hydroxide on polyethylene¹⁰.

What is difficult is how to explain the dependence of the adsorption of yttrium at $\text{pH} > 7$ on the chemical nature of the hydroxide employed for the adjustment of pH . As far as we know no data suggesting the formation of an ammonium complex of yttrium have been reported. In this connexion it can only be stated that ammonium ions have been found to decrease the adsorption of cerium, yttrium and strontium on ferric hydroxide¹⁶ and, if present in a larger amount, to impede the separation of yttrium hydroxide¹⁷.

The fact that there is no difference in adsorption up to pH c. 5.5 between old and freshly prepared solutions indicates that during the aging of the solutions no important changes of the adsorbable forms of yttrium occur. This is in good agreement with the previously deduced conclusion that an equilibrium is rapidly established between the different forms of yttrium in such solutions⁷. In the range $\text{pH} 6$ to 9 the adsorption exhibits a moderate decrease with the age of the solution. A similar, though even slower, decrease in adsorption was observed with iron¹⁰. A possible explanation is the loss of the adsorbable forms of the trace element by their adsorption on the wall of the vessel where the solution was stored. Another cause may be some alteration of the particles of yttrium itself or of its colloidal carrier as a result of slowly proceeding chemical reactions or recrystallization. A considerable decrease in sorption as against the fresh solutions is observed at $\text{pH} > 10$, *i.e.* in solutions where yttrium gives rise to rapidly sedimenting negatively charged pseudocolloids⁷. From this fact it can be deduced that these pseudocolloids adsorb very little on polyethylene.

This dependence of adsorption on pH is at variance with the results reported by Švehla³, who found a sharp maximum of the sorption of yttrium on epoxide paints at $\text{pH} 7.6$. The discrepancy can be accounted for not only by the different nature of the adsorbent but also by a different method of the study of adsorption. Hogdahl⁶ investigated the adsorption of yttrium on polypropylene and his results somewhat deviate from ours. The adsorption did not increase until the pH had exceeded the value of approx. 5 and the slope of this increase (up to $\text{pH} \sim 10$) was smaller. This fact is probably due to the existence of complex ions of yttrium in sea water from which the adsorption was studied.

In order to ascertain whether the results obtained after 15 hours' adsorption are close to the equilibrium values we studied the kinetics of the adsorption at several values of pH . Fig. 3 shows that the adsorption is very rapid and is practically complete

within two hours. This fact accords with the majority of findings reported in earlier papers. The rapid course of adsorption on hydrophobic organic polymers was explained by the assumption that the sorbing particles do not have to diffuse through the surface layer adhering to the adsorbent¹⁰. In this connexion it is noteworthy that the adsorption of yttrium in sea water on polypropylene was found to be slow⁶. This slow kinetics may be caused by the sluggish re-establishment of equilibrium between the individual forms of the existence of yttrium in sea water during the adsorption. The stepwise increase in the adsorption of yttrium on polypropylene with time⁵ is also rather inconsistent with our results.

The dependence of the adsorption of yttrium on its concentration is shown in Fig. 4. It reveals that at higher concentrations the increase in sorption shifts to higher pH values and the sorption changes irregularly above pH 8. The shift of the start and the increase of the sorption may be due to heterogeneity of the sorption centres on the surface of polyethylene or to changes of the state of yttrium in the solution. If yttrium is sorbed on polyethylene by an ion exchange it is the most active (the most acidic) groups that get occupied first. When these are occupied completely (at higher concentrations of yttrium ions) further sorption can occur only on the less acidic groups, which release protons at higher pH values. This explains the relative decrease in the sorption with the increasing concentration of yttrium and the shift of the sorption to higher pH values. A similar shift was observed in studying the sorption of traces

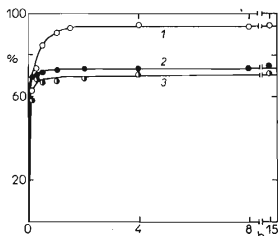


FIG. 3

Adsorption of Carrier-Free Yttrium on Polyethylene in Relation to Time

1 pH 7.5, fresh solution, pH adjusted with sodium hydroxide; 2 pH 9.8, fresh solution, pH adjusted with ammonium hydroxide; 3 pH 5.7, 7 days old solution, pH adjusted with sodium hydroxide.

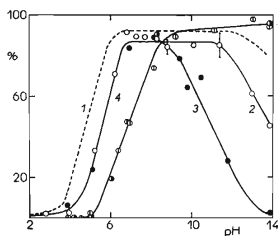


FIG. 4

Adsorption of Traces of Yttrium on Polyethylene in Relation to pH, Age of the Solution and Concentration of Yttrium

1 $< 10^{-9}$ M-Y, fresh solutions (see curve 1 in Fig. 1); 2 $2 \cdot 10^{-6}$ M-Y, fresh solutions; 3 $2 \cdot 10^{-6}$ M-Y, 7 days old solutions; 4 10^{-5} M-Y, fresh (○) and 7 days old (●) solutions. pH adjusted with sodium hydroxide.

of iron and gold on glass^{11,18}. The dialytic behaviour of the solutions and their electrophoresis have revealed that the state of yttrium changes with its increasing concentration⁷. At $\text{pH} < 8$ the portion of the pseudocolloidal forms of yttrium (formed by its sorption on the impurities or its co-precipitation with traces of ferric hydroxide) decreases. It is probable that the discussed shift of sorption is caused, partially at least, by a change of its mechanism, which is directly dependent on the state of yttrium. However, the mechanism of the sorption of yttrium from its $2 \cdot 10^{-6}\text{M}$ and 10^{-5}M solutions of $\text{pH} 4-8$ is difficult to describe since not even the state of yttrium in these solutions is known exactly⁷. We have only found by test experiments that sorption of carbonate complexes of yttrium is not operative since in this range of pH carbonates appreciably influence neither the state nor the sorption properties of yttrium. If the sorption proceeded mainly by an ion exchange of yttrium ions the considerable sorption of yttrium in the pH range 4 to 8 would testify to a rather great ion-exchange capacity of polyethylene, which has not yet been observed¹⁰.

Starting from $\text{pH} 8$ there were formed, in a $2 \cdot 10^{-6}\text{M}$ solution of yttrium, colloidal particles of yttrium hydroxide, which strongly prevail⁷ above $\text{pH} 10$. In accordance with this fact the adsorption of yttrium in this range of pH can be interpreted as a physical adsorption of true colloid particles of its hydroxide. The electrostatic adsorption could hardly occur since the colloid particles are either charge-free or their net charge is negative (as is that of polyethylene). The high value of sorption of yttrium from its 10^{-5}M solutions indicates a considerable capacity of polyethylene for the physical adsorption of colloids. From Fig. 4 it follows that the adsorption

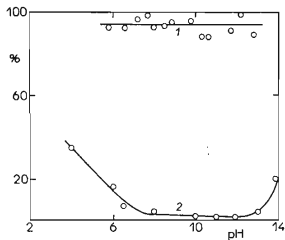


FIG. 5

Desorption of Carrier-Free Yttrium from Polyethylene in Relation to pH of the Solution in the Preceding Adsorption

1 Desorption by 1M-HCl ; 2 desorption by a solution of the same pH and composition as in the adsorption.

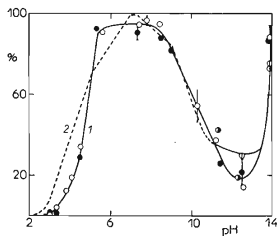


FIG. 6

Adsorption of Carrier-Free Yttrium on Polyethylene in the Presence of $3 \cdot 10^{-5}\text{M}$ Iron

1 Adsorption of yttrium from fresh (\circ) and 7 days old (\bullet) solutions; 2 adsorption of $3 \cdot 10^{-5}\text{M}$ iron (ref.¹⁰) from fresh (\circ) and 7 days old (\bullet) solutions.

of the colloidal yttrium hydroxide is a function of pH, concentration of yttrium in the solution and age of the solution. The experiments we have performed do not allow us to attempt a more detailed analysis and interpretation of these relations. Nevertheless, it is probable that with an increase in pH and age of the solution the structure of the solid phase or the particle size of the hydroxide changes as a consequence of "ripening", recrystallization or coagulation. These changes depend on the concentration of yttrium hydroxide and manifest themselves in the adsorption.

To obtain more data on the character of the adsorbed particles and their bonding to the surface we have studied desorption of the carrier-free yttrium from the surface of polyethylene in relation to the composition of the desorption solution and to pH of the solution in the preceding adsorption. From Fig. 5 it is seen that desorption of yttrium by 1M hydrochloric acid is almost quantitative throughout the pH range studied. The easy desorption by an acid is characteristic of colloidal particles which dissolve in it. Desorption of ions or molecules may not be so ready, especially if they are bound by chemisorption^{19,20}. Consequently, the course of desorption of yttrium effected by hydrochloric acid suggests that chemisorption is not the predominant mechanism of the sorption of yttrium on polyethylene at pH 6 to 13. The second curve in Fig. 5 represents the desorption of yttrium by a solution of the same pH and composition as in the adsorption. This desorption decreases up to pH 7; between pH 8 and 13 it is very low and virtually constant. What is noteworthy is the increase in desorption in strongly alkaline solutions, which, unlike other cases, cannot be explained by dissolution of the adsorbent. A similar increase in desorption from polyethylene has not yet been reported. It is probably caused by the action of sodium hydroxide on the colloidal impurities, which carry part of yttrium and are sorbed in alkaline solutions.

In the range pH 6–9.5 the desorption of yttrium by either solution is practically identical with the desorption of traces of ferric hydroxide from polyethylene¹⁰. This fact and the identity of regularities of the adsorption of yttrium and iron in the above-mentioned range of pH support the interpretation of sorption of the carrier-free yttrium as co-precipitate with colloidal ferric hydroxide. However, it was not possible to prove directly the interaction of the least quantities of the two elements in the adsorption on polyethylene since no data are available on the behaviour of yttrium in the absolute absence of iron. For this reason we tried to find at least an indirect proof of this interaction by studying the effects of larger quantities of ferric hydroxide (c. $3 \cdot 10^{-5}M$) on the sorption of yttrium. The results of this study are given in Fig. 6; for the sake of comparison we have also plotted some earlier data¹⁰ on the sorption of iron alone, supplemented by only a few points in the alkaline region. Comparison of curve 1 in Fig. 6 with curves 1 and 2 in Fig. 1 reveals that the addition of iron does not affect the adsorption of yttrium at pH < 5. Consequently, up to pH 5 no form of the existence of iron in the solution suppresses the adsorption of yttrium. However, if the concentration of iron is two orders of magnitude higher

the adsorption of yttrium is increased in the pH range 5 to 8.5 (particularly in old solutions) and decreased at higher values of pH. Since at $\text{pH} > 5$ considerable co-precipitation of yttrium with ferric hydroxide was found²¹ these changes can be explained by the assumption that the sorption behaviour of yttrium is essentially given by the sorption of its colloidal carrier. The identity in the course of adsorption of yttrium and iron (Fig. 6), which is especially marked in the range pH 6 to 11.5, substantiates this assumption. In this region of pH the electrophoretic and colloid properties of the two elements are also very similar⁷. The deviations in their sorption properties at higher values of pH, which are not very marked, cannot be explained as due to an incomplete co-precipitation since if this were the case the sorption of yttrium should be higher (see curve 1 in Fig. 1). We consider it probable that the colloidal particles of ferric hydroxide in alkaline solutions are heterogeneous and yttrium does not co-precipitate equally with any of them. It is of interest that the adsorption of both yttrium and iron increases above pH 13. This fact may be caused by a change in the properties of ferric hydroxide with the increasing concentration of sodium hydroxide.

CONCLUSIONS

Traces of yttrium are adsorbed on polyethylene at pH 3.5–6 mainly by an ion exchange of yttric ions in the outer part of the electric double layer or on the occluded impurities. At $\text{pH} > 6.5$ there prevails physical adsorption or chemisorption of pseudocolloids of yttrium or of the products of its hydrolysis. At higher concentrations of yttrium and $\text{pH} > 8$ physical adsorption of colloidal yttrium hydroxide occurs. It is corroborated¹⁰ that the capacity of polyethylene for the adsorption of colloidal particles is rather high.

From the data on the interaction of iron and yttrium it can be concluded that iron affects the sorption of yttrium on polyethylene only if present in the form of colloidal ferric hydroxide. No effect of traces of ionic forms of iron has been observed. The above-mentioned conclusion agrees with some qualitative findings on the interaction of traces of other elements, obtained in preliminary experiments^{22,23}. The close similarity of the adsorption of yttrium in the presence of low amounts of ferric hydroxide to the adsorption of this hydroxide at $\text{pH} > 6$ is another proof that under certain conditions in the solution the trace element can lose some of its chemical properties²⁴.

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