317. *Adsorption of Complex Anions from Uranyl Sulphate Solution by Anion-exchange Resins.*

By T. V. Arden and G. A. Wood.

At pH values below pH 2·5 uranium is adsorbed from sulphate solution by strong-base anion-exchange resins as a complex $UO_2(SO_4)_3^{4-}$. There is evidence that adsorption occurs by the formation of this ion on the resin by the addition of UO_2SO_4 to the resin sulphate, as well as by the normal exchange process. At pH values in excess of 2·5, the uranium loading of the resin increases, owing to the presence of increasing proportions of $U_2O_5(SO_4)_3^{4-}$ on the resin.

Most of the work carried out on uranyl salt solutions has shown that uranium is present largely in cationic forms. McInnes and Longsworth [1] and Longsworth, Best, and Taub [2] showed that the UO_2^{2+} ion is present in acid solutions and is hydrolysed to form $U_2O_5^{2+}$ with increasing pH. Sutton [3] confirmed these results in non-complexing solutions, and showed that further hydrolysis gives $U_3O_8^{2+}$ and $U_3O_8{\cdot}OH^+$. He pointed out that in sulphate solutions his results would be modified by the effects of ion-pair formation. Betts

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[1] McInnes and Longsworth, MDDC 911, Tech. Inf. Div., Oak Ridge, Tennessee.
[2] Longsworth, Best, and Taub, A380, M.R.C. of Canada, 1942.
[3] Sutton, *J.*, 1949, S 286.

and Michels[4] showed by spectrophotometric methods that in the concentration range 0·0—1·2M with respect to sulphate, the only complex present was the uncharged UO_2SO_4. This work has been criticised by Ahrland,[5] who has calculated formation constants for UO_2SO_4, $UO_2(SO_4)_2^{2-}$, and $UO_2(SO_4)_3^{4-}$ from the results of potentiometric and spectrophotometric studies. Nevertheless, his work suggests that, in dilute solutions at least, the predominant complex is UO_2SO_4 and that anionic complexes exist only in low concentrations. The absence of significant concentrations of anions in solution is confirmed by Arden,[6] who showed by conductometric and potentiometric methods that the properties of uranyl sulphate solutions from pH 1·5 to pH 5·0 can be explained almost completely by the postulation of Sutton's condensed cations[3] and the corresponding uncharged sulphates.

All evidence thus points to the virtual absence of anionic complexes from uranyl sulphate solutions. Nevertheless, it has been found that uranium is strongly adsorbed from these solutions by strong-base anion-exchange resins. The purpose of this work, which was commenced at the Chemical Research Laboratory in 1950, has been to study the nature of the anions adsorbed and the adsorption mechanism.

EXPERIMENTAL

The resins used were Amberlite IRA-400, De-Acidite FF, and some samples made at the Chemical Research Laboratory. All these materials are stated to be cross-linked polystyrene, containing quaternary groups of the benzyltrimethylammonium type; and no significant difference between them could be found for this purpose.

The chloride capacities and dry weights recorded were in each case measured on the actual column of resin used for uranium adsorption, by reconverting it into the chloride form at the end of each test, and finally drying and weighing it in this form. By this means, errors due to varying moisture content were prevented, without introducing others by drying the resin samples before use.

All tests were carried out in downward flowing columns, which were run to final equilibrium. In the experiments used for capacity and charge-balance comparisons, the columns were contained in centrifuge tubes closed with filter discs. To avoid errors due to changes in conditions, the resin was not washed at the end of each test, but was freed from excess of solution by centrifuging, the measured ionic content of the resin being corrected for the effect of remaining interstitial liquor by the method of Kraus and Moore.[7] This correction was negligible in cases where the sulphate content of the solution used was less than 0·5M, and in no case exceeded 2%.

Determinations were made first of uranium and sulphate taken up by Amberlite IRA-400 resin in the sulphate form, on passage of a pure uranyl sulphate solution (0·098M) containing a small excess of sulphate through a column of the resin (2·40 g. dry weight). The results are shown in Table 1.

TABLE 1.

Effluent (ml.)	50	100	150	200	250	300	350	400	450	500
SO_4 adsorbed (mmoles)	0·497	0·495	0·442	0·268	0·188	0·136	0·108	0·091	0·075	0·062
UO_2 adsorbed (mmoles)	0·491	0·488	0·442	0·270	0·169	0·136	0·107	0·082	0·077	0·060
$SO_4 : UO_2$ ratio	1·01	1·02	1·00	0·99	1·11	1·00	1·01	1·11	0·97	1·03

Determinations of the total uranium uptake of several different types of quaternary ammonium resins were carried out by using 0·005—0·01M-uranyl sulphate solutions, at pH 1·7—2·0. This pH range was chosen since it has been shown by Longsworth *et al.*[1] that, above pH 2·0, there is a measurable degree of hydrolysis of the UO_2^{2+} ion to give the condensed $U_2O_5^{2+}$ ion. Typical results are given in Table 2. The significance of the term *n* is considered on p. 1599.

The effect of sulphate concentration on uranium adsorption was measured by the centrifuge technique, with a sample of De-Acidite FF having a capacity of 3·70 mequiv./g. The solutions used were in each case 0·005M with respect to UO_2SO_4, with varying concentrations of sulphate added as the magnesium salt. Two series of tests were carried out, at pH 1·8 and 2·5. The ratio of chloride desorbed to uranium adsorbed, and the ratio of sulphate to uranium adsorbed,

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[4] Betts and Michels, *J.*, 1949, S 276.

[5] Ahrland, *Acta Chem. Scand.*, 1951, **5**, 1151.

[6] Arden, *J.*, 1949, S 60; Thesis, London, 1953.

[7] Kraus and Moore, *J. Amer. Chem. Soc.*, 1953, **75**, 1497.

TABLE 2.

	Capacity Cl^- (mequiv./g.)	UO_2 (mmoles/g.)	Mol. ratio $Cl : UO_2$	*n*
De-Acidite FF	3·58	0·878	4·08	3·06
Amberlite IRA-400 ...	3·60	0·915	3·94	2·97
De-Acidite FF	3·60	0·913	3·95	2·95
Amberlite IRA-400 ...	3·71	0·918	4·04	3·03
De-Acidite FF	3·87	0·968	4·00	3·00
AMX 35 (CRL)	4·1	0·963	4·26	3·21
AMX 36 (CRL)	4·8	1·073	4·44	3·33

FIG. 1.

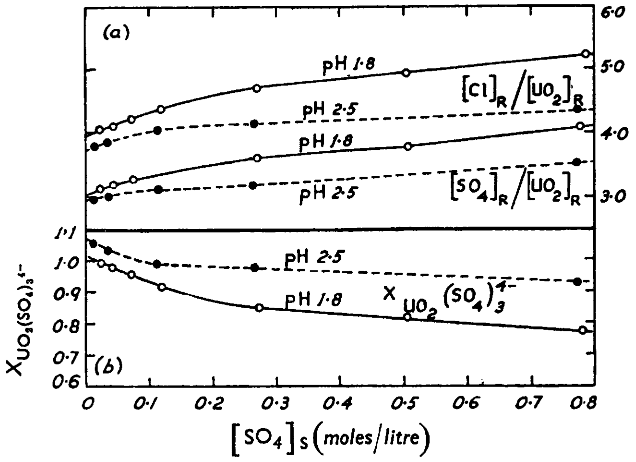
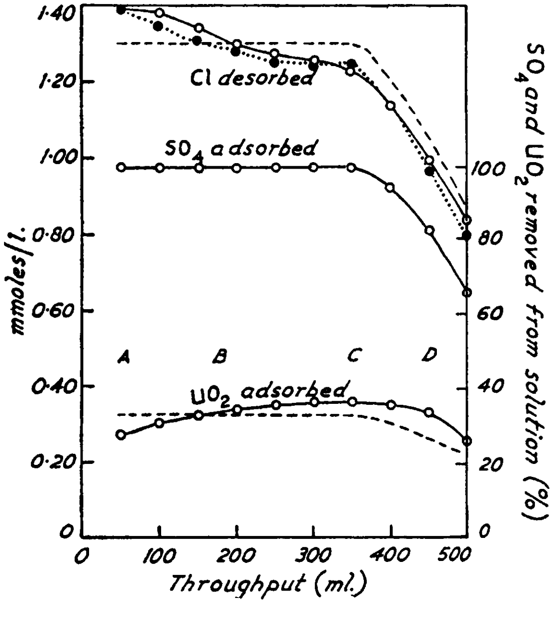


FIG. 2.



–○—○– Observed.

— — — — Cl and UO_2, calc. from observed SO_4.

··●····●·· Cl, calc. from observed SO_4 and UO_2.

are given in Fig. 1*a*, and the calculated proportion of resin capacity occupied by a postulated ion $UO_2(SO_4)_3^{4-}$ is plotted in Fig. 1*b*.

This test gave the final equilibrium results. The adsorption process was studied by passing through a column of IRA-400 (chloride form, 2·4 g.) a solution 0·00946M with respect to UO_2 and 0·00972M with respect to SO_4. The reduction in concentration of uranium and sulphate in the emerging fractions, together with the concentration of chloride produced, are plotted in Fig. 2 as continuous curves.

The variations of uranium and sulphate loadings with pH were also measured by the centrifuge technique, using a column containing De-Acidite FF having a chloride capacity of 3·62 mequiv./g., and solutions 0·005M with respect to UO_2SO_4, together with sufficient sulphuric acid or sodium hydroxide to adjust the pH to a series of values from 0·5 to 5·0, above which precipitation of uranyl hydroxide occurred. The uranium loadings are plotted in Fig. 3, and the significance of sulphate adsorption is considered in the discussion of Tables 4 and 5.

A similar series of tests was carried out on the adsorption of sulphate and bisulphate from unbuffered solutions of sulphuric acid, containing no uranium, adjusted to have the same free sulphate content as the corresponding uranium solutions. The sulphate content varied from 0·0044M at pH 2·5 to 0·248M at pH 0·58. These results are plotted in Fig. 4.

FIG. 3.

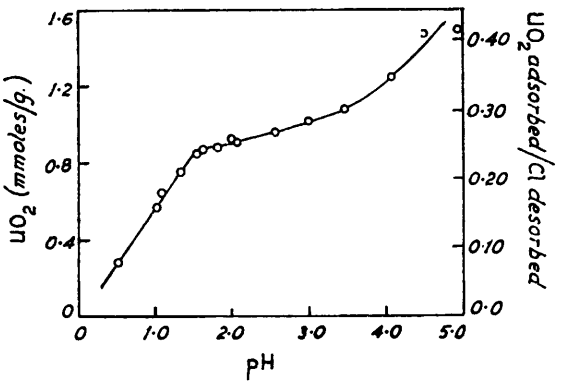
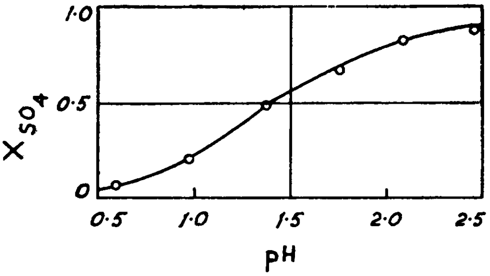


FIG. 4. Ratio, SO_4/HSO_4 on resin.



DISCUSSION

The results given in Table 1 show that, during the adsorption of uranium on a sulphate-form resin, equimolecular amounts of uranium and sulphate are taken up at all stages, suggesting that the complex on the resin is of the type $UO_2(SO_4)_n{}^{2-2n}$, where $n \gg 2$. If it is assumed that the resin is saturated with one ionic type, then the penultimate column of Table 2 gives the charge on this ion, *viz.*, $(2 - 2n)$, from which is calculated the $SO_4 : UO_2$ ratio, n. In tests with 65 different resin samples, this value showed a maximum variation of 2·95—3·06, except with resin samples of exceptionally high chloride capacity, in which the complete adsorption of uranium was apparently prevented by steric hindrance between the large complex anions, giving high values of n. The two lowest members of the series are $UO_2(SO_4)_2{}^{2-}$ and $UO_2(SO_4)_3{}^{4-}$, the measured value of n suggesting that the latter is adsorbed, although the same value would be given if the resin contained at equilibrium an equal mixture of $UO_2(SO_4)_2{}^{2-}$ and $SO_4{}^{2-}$.

Two possible mechanisms exist for the adsorption of the uranyl trisulphate ion. The first is the formation of the complex in solution, followed by its exchange with sulphate on the resin :

$$UO_2{}^{2+} + 3SO_4{}^{2-} \rightleftharpoons UO_2(SO_4)_3{}^{4-} \quad \ldots\ldots\ldots \quad (1)$$

$$2R_2SO_4 + UO_2(SO_4)_3{}^{4-} \rightleftharpoons R_4UO_2(SO_4)_3 + 2SO_4{}^{2-} \quad \ldots\ldots \quad (2)$$

$$K_1 = [UO_2(SO_4)_3{}^{4-}]_S/[UO_2{}^{2+}]_S[SO_4{}^{2-}]_S{}^3$$

$$K_2 = [UO_2(SO_4)_3{}^{4-}]_R[SO_4{}^{2-}]_S{}^2/[SO_4{}^{2-}]_R{}^2[UO_2(SO_4)_3{}^{4-}]_S$$

Whence $[UO_2(SO_4)_3{}^{4-}]_R/[SO_4{}^{2-}]_R{}^2 = K_1K_2[UO_2{}^{2+}]_S[SO_4{}^{2-}]_S$

By this process, therefore, at constant uranium concentration in solution, the uranium uptake on the resin would decrease with decreasing sulphate concentration in solution. No such decrease can be observed experimentally; and it must be deduced that if the mechanism of equations (1) and (2) operates, the product K_1K_2 is so large that $[SO_4{}^{2-}]_R{}^2$ is reduced to an undetectable level, irrespective of the value of $[SO_4{}^{2-}]_S$. A similar reasoning can be applied to the possible lower complex $UO_2(SO_4)_2{}^{2-}$; and it follows that there is effectively a complete absence of competition for active groups on the resin, which must, under the conditions of these tests, always become saturated with the uranium complex.

The second possible mechanism is by the addition reaction (3):

$$2R_2SO_4 + UO_2SO_4 \rightleftharpoons R_4UO_2(SO_4)_3 \qquad (3)$$

Undissociated UO_2SO_4, being uncharged, would be able to enter the resin freely, without hindrance by the Donnan membrane effect. Equation (3) implies lack of competition for the resin sites, and complete saturation of the resin by the uranium complex irrespective of sulphate concentration. By either of these mechanisms, therefore, the limiting values for $[SO_4]_R/[UO_2]_R$ and $[Cl]_R/[UO_2]_R$ represent the sulphate content and charge of the complex ion. At pH 1·8, these limiting values are 3·0 and 4·0, and the only uranium complex present in measurable quantities is therefore $UO_2(SO_4)_3^{4-}$. The results at pH 2·5 are discussed below.

Since it has been shown that the presence of sulphate in solution has no effect on uranium adsorption, the mole-fraction of the complex on the resin, $X_{UO_2(SO_4)_3^{4-}}$, should remain constant at 1·0, irrespective of $[SO_4]_S$, whereas Fig. 1*b* shows that this value decreases with increasing measured $[SO_4]_S$. This decrease is due to competition from HSO_4^- ions which at pH 1·8 account for approximately half the free sulphate in solution ($K_{HSO_4} = 2 \times 10^{-2}$ at 18°). Table 3 gives a calculation of the charges accounted for, by assuming that the bisulphate and uranyl trisulphate anions are the only ones present on the resin. The total agrees well with the measured chloride capacity of 3·78 mequiv./g. for this resin sample.

TABLE 3. *Charge balance at* pH 1·8.

Solution		Resin (mmole/g.)				Charge (mequiv./g.)		
UO_2 (mole/l.)	SO_4 (mole/l.)	UO_2 (measured)	Combined SO_4 (3 × UO_2)	Total SO_4 (measured)	Free SO_4 (diff.)	HSO_4	$UO_2(SO_4)_3^{4-}$ (4 × UO_2)	Total
0·0048	0·780	0·723	2·17	2·92	0·75	0·75	2·99	3·67
,,	0·509	0·766	2·30	2·88	0·58	0·58	3·07	3·65
,,	0·270	0·801	2·41	2·88	0·47	0·47	3·21	3·68
,,	0·119	0·861	2·58	2·92	0·34	0·34	3·44	3·78
,,	0·071	0·899	2·70	2·92	0·22	0·22	3·60	3·82
,,	0·044	0·924	2·77	2·92	0·15	0·15	3·69	3·84
,,	0·023	0·942	2·83	2·93	0·10	0·10	3·77	3·87

Similar calculations were carried out, by assuming that (*a*) $UO_2(SO_4)_3^{4-}$ and SO_4^{2-}, or (*b*) $UO_2(SO_4)_2^{2-}$ and SO_4^{2-} were the only ions present. In each case the total charge increased with increasing values of $[SO_4]_S$, from 3·93 to 4·49. These differences are much greater than the experimental error, and it is concluded that no combinations of these ions can account for the observed results. The only other possibility, that of $UO_2(SO_4)_2^{2-}$ and HSO_4^-, gives very low values.

The adsorption of uranium as a complex sulphate requires that removal of uranium from a stoicheiometric uranyl sulphate solution by a chloride-form resin cannot be complete. This is shown to be true in Fig. 2, which also gives evidence of the mechanism of adsorption. The values calculated for uranium and chloride exchange, based on the observed sulphate figures, with the assumption that the trisulphate anion is adsorbed throughout, are shown as broken lines.

The observed values are sufficiently close to the calculated figures to confirm that the overall process is that of the adsorption of the trisulphate anion; but in the early part of the run, in the region *AB*, the uranium adsorption is slightly lower than the theoretical value, the chloride desorption being higher. It follows that, in this region, sulphate is being partly adsorbed in place of the uranium complex, and is thereby displacing two chloride ions for each sulphate adsorbed, instead of 1·33. This phenomenon would occur if the adsorption of uranium on a given site required the prior conversion of this site into the sulphate form, followed by the addition of UO_2SO_4 (equation 3). The same effect would be given by a normal ion-exchange process if the rate of the sulphate–chloride exchange were higher than the rate of the uranium anion–chloride exchange. The two processes can be distinguished by the next part of the curve. In the region *BC* uranium adsorption exceeds the calculated value, while chloride emission is reduced. This effect, which means that a greater degree of exchange is apparently taking place without any corresponding emission of ions from the resin, can only be explained by the addition of uranyl sulphate molecules on to resin sites already in the sulphate form, this process being superimposed on the adsorption of the trisulphate molecule by either of the mechanisms discussed above. The fact that in the region *CD* uranium adsorption continues unchanged, although sulphate has broken through and chloride exchange is decreasing rapidly, gives further confirmation of this process.

The dotted curve in Fig. 2 gives the calculated chloride emission value based on the assumption that uranium is adsorbed as $UO_2(SO_4)_3{}^{4-}$ in the region *AB*, the excess of sulphate being present as $SO_4{}^{2-}$. In the remainder of the curve it is assumed that uncharged uranyl sulphate and the trisulphate are adsorbed together, the ratios of the two being simply calculated from the measured uranium and sulphate values. Summation of the charges on the anions present gives the calculated curve, which is in good agreement with the observed value.

All the results so far considered have been of measurements at pH 1·7—2·0. Fig. 3 shows the effect of varying pH; and it is clear that the complete adsorption of $UO_2(SO_4)_3{}^{4-}$ which has been postulated occurs only in the region previously chosen. The decrease in loading below pH 2·0 can be explained by the increasing effect of competition from bisulphate ions. It is shown in Fig. 4 that the reduction in sulphate adsorption below pH 2·5 is similar to the fall in uranium loading, supporting the theory that uranium is adsorbed on sites in the sulphate, but not the bisulphate form.

TABLE 4.

	Resin (mmole/g.)				Charge (mequiv./g.)		
Solution pH	UO_2 (measured)	Combined SO_4 (3 × UO_2)	Total SO_4 (measured)	Free HSO_4 (diff.)	HSO_4	$UO_2(SO_4)_3{}^{4-}$ (4 × UO_2)	Total
0·52	0·276	0·83	3·21	2·38	2·38	1·10	3·49
1·01	0·573	1·72	3·10	1·28	1·28	2·29	3·57
1·32	0·738	2·21	2·90	0·68	0·68	2·95	3·63
1·61	0·840	2·52	2·87	0·35	0·35	3·36	3·71
1·82	0·866	2·59	2·80	0·21	0·21	3·47	3·68
2·01	0·903	2·71	2·67	—ve	—	3·61	3·61

Table 4 gives the total of ion-active groups accounted for, on the assumption that $UO_2(SO_4)_3{}^{4-}$ and $HSO_4{}^-$ are the only ions adsorbed on the resin. Over the pH range 0·5—2·0, the results agree well with the total measured chloride capacity of 3·62 mequiv./g., the maximum difference being 0·13, or 3·6%.

The pH curve for values higher than pH 2·5 cannot be explained on the basis of the quadrivalent complex, since the total resin loadings are higher than the saturation value for this ion. These values could be given by the bivalent $UO_2(SO_4)_2{}^{2-}$ ion, but there is no theoretical reason why increasing pH should favour the adsorption of this ion at constant uranium and sulphate concentration.

A more probable explanation is given if it is assumed that the condensed uranyl ions which occur with increasing pH can form complexes of the type $U_2O_5(SO_4)_3{}^{4-}$, similar to that formed by the simple $UO_2{}^{2+}$ ion. As the effective charge per uranyl group is reduced, the increase in resin loading with pH is explained.

Table 5 gives the ratios of uranium and sulphate uptake to chloride capacity, which would be given by saturation of a resin (capacity 3·62 mequiv./g.) with each of the possible ions which can be formed by combination of sulphate with the various uranyl cations. The observed values at pH 2·0 and 5·0 are included. A number of different mixtures of complex anions can be posulated to account for the observed uranium uptake at high pH, and two typical examples are given.

All bivalent complexes give SO_4 : Cl ratios of 1 : 1, the ratios for quadrivalent complexes being 0·75 : 1. The observed results at pH 5·0 have varied by ±5%, but in no case have they exceeded the 0·75 : 1 ratio. It is therefore improbable that $UO_2(SO_4)_2^{2-}$ is present on the resin under these conditions, and the enhancement of uranium adsorption must be due to an increasing proportion of $U_2O_5(SO_4)_3^{4-}$ with increasing pH.

TABLE 5.

	Ratios			Total mmole/g.		
	UO_2	SO_4	Cl	UO_2	SO_4	Cl
$UO_2(SO_4)_2^{2-}$	0·5	1·0	1·0	1·81	3·62	3·62
$UO_2(SO_4)_3^{4-}$	0·25	0·75	1·0	0·91	2·72	3·62
$U_2O_5(SO_4)_2^{2-}$	1·0	1·0	1·0	3·62	3·62	3·62
$U_2O_5(SO_4)_3^{4-}$	0·5	0·75	1·0	1·81	2·72	3·62
$U_3O_8(SO_4)_2^{2-}$	1·33	1·0	1·0	4·80	3·62	3·62
$U_3O_8(SO_4)_3^{4-}$	0·75	0·75	1·0	2·72	2·72	3·62
Observed at pH 2·0	0·25	0·75	1·0	0·91	2·72	3·62
Observed at pH 5·0	0·41	0·69	1·0	1·50	2·48	3·62
$1UO_2(SO_4)_3^{4-}$: $3{\cdot}8UO_2(SO_4)_2^{2-}$	0·41	0·90	1·0	1·50	3·25	3·62
$1UO_2(SO_4)_3^{4-}$: $1{\cdot}9U_2O_5(SO_4)_3^{4-}$	0·41	0·75	1·0	1·50	2·72	3·62

The observed values could also be explained by the presence of increasing amounts of precipitated uranyl hydroxide on a resin remaining in the $UO_2(SO_4)_3^{4-}$ form throughout. This explanation is considered improbable, since no precipitate could be detected visually in the experiments recorded, whereas it was shown, in tests carried out at a slightly higher pH, that a quantity of precipitate which made no significant difference to the measured uranium loading was clearly visible. The polyuranate anions of the type $U_3O_8(OH)_3^-$ and $U_3O_8(OH)_4^{2-}$, suggested both by Sutton [3] and by Arden,[6] which would also have this effect, were shown by Arden to be significant only above pH 7·0.

These considerations of the effect of pH give the reasons for the difference in results at pH 2·5 as shown by the broken curves of Fig. 1. This pH was chosen as being the point at which both HSO_4^- adsorption (Fig. 4) and $U_2O_5(SO_4)_3^{4-}$ adsorption (Fig. 3) were low. The reduced effect of bisulphate results in a reduction in the slope of the broken curves, as compared with the continuous curves measured at pH 1·8. The apparent anomaly in the value for $X_{UO_2(SO_4)_3^{4-}}$ at low sulphate concentrations is also explained. Table 6 gives the charge balance, calculated on the assumption that at pH 2·5 the uranyl anions are present in all tests in the ratio $18UO_2(SO_4)_3^{4-}$: $1U_2O_5(SO_4)_3^{4-}$, the value which corresponds with the observed uranium uptake in the lowest line of the table. Then the mean charge per uranyl group is —3·8 and the mean SO_4 : UO_2 ratio 2·85. The free sulphate is assumed to be present as HSO_4^-. The total charge accounted for is in good agreement in every case with the total chloride capacity of the resin sample used, *viz.*, 3·62 mequiv./g.

TABLE 6. *Charge balance at* pH 2·5.

Solution		Resin (mmoles/g.)				Charge		
UO_2 (mole/l.)	SO_4 (mole/l.)	UO_2 measured	Combined SO_4 (2·85 × UO_2)	Total SO_4 (measured)	Free SO_4	HSO_4	U complex (3·80 × UO_2)	Total
0·048	0·771	0·835	2·38	2·97	0·55	0·55	3·09	3·64
,,	0·266	0·879	2·50	2·80	0·30	0·30	3·25	3·55
,,	0·117	0·895	2·55	2·70	0·24	0·24	3·31	3·55
,,	0·035	0·930	2·65	2·80	0·15	0·15	3·44	3·59
,,	0·011	0·953	2·72	2·82	0·10	0·10	3·53	3·63

It is concluded that the whole of the observed phenomena can be explained by the assumptions that, at pH values below 2·0, only $UO_2(SO_4)_3^{4-}$ and HSO_4^- are present on the resin, neither $UO_2(SO_4)_2^{2-}$ nor SO_4^{2-} being adsorbed in measurable quantities. The increase in loading at higher pH values is explained by the presence of $U_2O_5(SO_4)_3^{4-}$. There is nevertheless no conflict with the work of Ahrland,[5] who postulated an equilibrium in solution between $UO_2(SO_4)_2^{2-}$ and $UO_2(SO_4)_3^{4-}$. It is generally agreed that uranium exists in sulphate solution almost entirely as the uranyl cation, or as undissoicated uranyl sulphate, and that the complex anions exist in very low concentrations. Under these conditions, it is to be expected that the affinity of the quadrivalent complex will be so much higher than that of the bivalent complex, that the equilibrium in solution will be almost completely displaced in the presence of the resin.

Since the completion of this work O'Connor's [8] studies on the same subject have been declassified. Ahrland's equilibrium constants were used as a basis for the calculations of the equilibrium concentrations of the various anions on the resin. Although in general agreement with us that the predominant species present is $UO_2(SO_4)_3^{4-}$, O'Connor has calculated that under conditions similar to ours, up to 10% of the resin sites may be occupied by $UO_2(SO_4)_2^{-2}$. The calculations are based upon the fundamental assumption that the HSO_4^-–SO_4^{2-} equilibrium on the resin, which was studied separately, is unchanged by the presence of the complex anions. It is considered unlikely that this can be true in view of the agreement between O'Connor and ourselves that almost, if not all, the sulphate on the resin is present as complex. O'Connor's further deduction that solutions containing high uranium concentrations with low free sulphate yield high proportions of $UO_2(SO_4)_2^{2-}$ on the resin, is not in conflict with our results. From general considerations of mass action effects in a 2 : 4-valent system it is to be expected that increasing ionic strength would favour the lower complex.

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[8] O'Connor, T.O. ACCO-61 (U.S.A.E.C.), 1954.