Determination of K_d values of ^{44}Ti and ^{44}Sc in $HCl/H_2C_2O_4$ solution of various concentrations

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Introduction: For preparation of $^{44}Ti/^{44}Sc$ radionuclide generators, several radiochemical criteria are relevant, such as effective separation strategies providing high Sc yields and low Ti breakthrough, high long-term stability, and type of Sc eluates useful for subsequent labelling reactions (i.e. low volume, low pH, high purity etc.). In this study, ion exchange experiments have performed systematically using AG-1x8 (200-400 mesh Cl^--form) and AG-50x8 (200-400 mesh H^+-form) resin in $H_2C_2O_4$/HCl solution in order to evaluate the potential of ion exchange-based generators.

Experimental: K_d values for both Sc and Ti were determined in batch experiments using different concentrations of HCl / oxalic acid mixtures. $^{44}Ti(^{44}Sc)$ and ^{46}Sc were used as isotopic tracers for Ti(IV) and Sc(III) distributions. $^{44}Ti(^{44}Sc)$ was prepared as described earlier [1]. ^{46}Sc was produced via γ-reaction on natural Sc at the HMI Berlin reactor. A stock solution of ^{44}Ti (30 KBq) and ^{46}Sc (1 mg, 20 mCi) was dried and dissolved in 100 µl 0.1 M $H_2C_2O_4$ (solution X).

Aliquots were prepared in Eppendorf 1.5 ml vials with 100 mg of AG-1x8 (200-400 mesh Cl^--form) or AG-50x8 (200-400 mesh H^+-form). To all probes 1 ml of $HCl/H_2C_2O_4$ mixture was added. Than 5 µl of solution X was added to probes 1-9 and they were shaken during 2 days.

Another solution Y – a probe with ^{44}Ti (117 KBq) with the remaining solution of X and ^{46}Sc were dried and dissolved in 0.025 M $H_2C_2O_4$. 5 µl of solution Y was added to probes 10-16 and they were shaken during 2 days. Another solution Z was prepared – a probe with ^{44}Ti (27 KBq) with the rest of solution Y and ^{46}Sc dried and dissolved in 0.005 M $H_2C_2O_4$. 5 µl of solution Z was added to probes 17-26 and they were shaken during 2 days.
K_d was calculated by
$K_d = (4A-10A')/A'$
A – activity of a whole probe with resin
A' – activity of 400 µl sample of a probe after ion-exchange reaction

Results and Discussion:
Results of the K_d values obtained for the two different ion exchange resins and the various mixtures are shown in table 1.

Table 1. Distribution coefficents of Ti(IV) and Sc(III) in HCl / oxalic acid mixtures for cation and anion exchange resins

	N	Concentration of solution, mol/l		K_d			
				AG-50x8		AG-1x8	
		$H_2C_2O_4$	HCl	Ti	Sc	Ti	Sc
X	1	0.1	0	-	-	>1000	184
	2	0.1	0.05	-	-	>1000	41
	3	0.1	0.1	-	-	>1000	14
	4	0.1	0.15	<< 1	12.0	>1000	5.1
	5	0.1	0.20	<< 1	10.7	>1000	1.7
	6	0.1	0.30	<< 1	7.0	370	0.2
	7	0.1	0.50	<< 1	11.2	105	<< 1
	8	0.1	0.75	~0,5	14.0	-	-
	9	0.1	1.0	<< 1	8.1	17	<< 1
Y	10	0.025	0	1.0	201	>1000	954
	11	0.025	0.025	1.0	148	>1000	168
	12	0.025	0.050	0.6	129	>1000	40.9
	13	0.025	0.075	1.8	128	>1000	14.2
	14	0.025	0.125	3.3	124	1050	2.68
	15	0.025	0.175	3.1	120	410	0.3
	16	0.025	0.250	2.9	119	290	<< 1
Z	17	0.005	0	32	7619	>1000	2340
	18	0.005	0.025	30.4	2378	>1000	67.2
	19	0.005	0.0375	34.2	2242	>1000	24.0
	20	0.005	0.05	33.6	2665	>1000	10.9
	21	0.005	0.065	28.2	1872	>1000	4.0
	22	0.005	0.08	33	1715	844	1.27
	23	0.005	0.10	33	1646	688	0.71
	24	0.005	0.125	25.6	1398	457	<< 1
	25	0.005	0.25	-	-	46	<< 1
	26	0.005	0.5	-	-	3.8	<< 1

Best conditions for efficient separations and for the design of generators are to elute AG-1x8 resins with 0.2 M HCl / 0.1 M $H_2C_2O_4$, 0.125 M HCl / 0.025 M $H_2C_2O_4$ or 0.06-0.008 M HCl / 0.005 M $H_2C_2O_4$ mixtures.

Conclusions: Using optimum K_d values for the HCl / oxalic acid mixtures of 0.2 M HCl and 0.1 M $H_2C_2O_4$, model generators will be constructed and evaluated. However, in view of subsequent ^{44}Sc labelling reactions, a post-elution processing of the ^{44}Sc fraction may be required. For example, ^{44}Sc may be on-line absorbed selectively on a cation exchange cartridge. In this case, 0.06-0.008 M HCl / 0.005 M $H_2C_2O_4$ mixtures offer optimum K_d values for Sc(III) adsorbtion.

References:
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