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THE DEVELOPMENT OF A PURIFICATION METHOD OF LITHIUM

by

J. PAUWELS and K. F. LAUER

1968



Joint Nuclear Research Center Geel Establishment - Belgium

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European Atomic Energy Community — EURATOM Joint Nuclear Research Center — Geel Establishment (Belgium) Central Bureau for Nuclear Measurements — CBNM Brussels, September 1968 — 22 Pages — 3 Figures — FB 40

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SUMMARY

A purification method of lithium on a cation-exchange resin in 1 M HCl was described. To allow the preparation of lithium containing ≤ 50 ppm alkali and alkali-earth metals, a special purification of the ion-exchanger was needed. A method using batch operations with the tetra lithium salt of EDTA, with 30 % hydrochloric acid, and elution with 1 M hydrochloric acid was described.

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KEYWORDS

REFINING IMPURITIES SEPARATION PROCESSES LITHIUM ION EXCHANGE MATERIALS EDTA HYDROCHLORIC ACID

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1. Introduction

In a previous report, four methods of determination of 0.1 M lithium hydroxide solutions with a final accuracy of 0.02% were described ⁽¹⁾. As these methods, which are not selective, have to be applied to ⁶Li and ⁷Li solutions for the preparation of isotopic blends, and no chemically pure ⁶Li or ⁷Li samples were available, a purification method of lithium was needed.

The purity of the final lithium has to be such, that the residual impurities do not cause any hygroscopy that might disturb gravimetry of the formed lithium salts, and that corrections for systematic errors are practically negligible. Further, the lithium has to be in such a form that its transformation into a pure 0.1 M aqueous hydroxide solution is possible.

The most important impurities found in enriched ${}^{6}Li$ or ${}^{7}Li$ samples were alkali- and alkali-earth metals (up to 0.15% together).

Different separations of lithium from these elements were reported earlier, but they were mostly limited to one or two elements, and few authors give indications about the absolute purity of the separated lithium fraction. Most of these methods are based on extraction $\binom{(2-6)}{}$, precipitation $\binom{(7-12)}{}$, or ion exchange chromatography $\binom{(13-35)}{}$.

Nevertheless, the most recent of them all concern separations on cation exchange resins in the hydrogen form. An important study of the behaviour of the alkali metals in presence of Dowex 50 and Duolite C3 in the system HC1- $H_2O - CH_3OH$ was done by Nelson ⁽³⁵⁾. The described separation method is based on this work.

2. Principle of the Method

In diluted hydrochloric acid the alkali metals are rather strongly absorbed by cation-exchangers like Dowex 50, whereby lithium is the least absorbed one. From the values of the distribution coefficients as determined by Nelson⁽³⁵⁾ for Dowex-X4 it can be seen that quantitative separations must be possible.

To obtain still higher selectivities it is useful to use a resin with a higher DVB-content ("X16").

The described purification was performed on AG 50W-X16 (50/100 mesh), a Dowex 50W-X16 specially purified for analytical purposes. Nevertheless, further purification of this resin was necessary.

This was made by batch operations with E.D.T.A. in LiOH medium, concentrated hydrochloric acid and final elution with 1 M hydrochloric acid.

3. Experimental Details of the Method

3.1. <u>Elements of interest and their determination</u>

3.1.1. Lithium

Lithium was localized in the eluate and determined for the calculation of the distribution coefficients by transformation into and calcination as sulfate.

In the case of the K_d -determinations 5 ml fractions containing 7 to 15 mg of lithium sulfate were analysed. Under these conditions a precision of 1-2% can be obtained.

3.1.2. Sodium, potassium, beryllium, magnesium and calcium During the determination of distribution coefficients and purification factors, these elements were determined relatively by tracing them with ²²Na, ⁴²K, ⁷Be, ²⁸Mg and ⁴⁵Ca. The first four isotopes were measured by integral γ-counting

with a NaI(T1) Well-type scintillation counter, 45 Ca by relative β -counting. During the separation test Li/Mg, magnesium was determined photometrically with titan yellow.

3.1.3. Trace analysis in ion-exchangers and purified lithiums Ion-exchangers were calcined and purified lithiums dried as lithium chloride. The residues were analysed for metallic impurities by the emission spectrographic service of the C.B.N.M.

3.2. <u>Determination of distribution coefficients</u> (K_d)

Distribution coëfficients for lithium, sodium, potassium, beryllium, magnesium and calcium were determined for commercial grade Dowex 50W-X16 by batch equilibration tests on 1-2 grams oven dried resin and 10 ml of hydrochloric acid. K_d-values were calculated from relative measurements of identical fractions of blanks and samples using the formula:

$$K_{d} = \frac{M_{t} - M_{1}}{M_{1}} \cdot \frac{ml}{g}$$

in which:

M_t = the total amount of the ion
M₁ = the amount present in the liquid
ml = the number of milliliters of hydrochloric acid.
g = the number of grams of oven-dried resin.

A summary of the obtained results is given in figure 1. Separation factors $\beta = K_{d_{M}} / K_{d_{Li}}$ are summarized in figure 2. From this figure it can be seen that an efficient separation $(\beta > 2)$ can be expected between 0.5 and 2 M hydrochloric acid. To avoid contamination of the separated lithium by trace impurities contained in the ion-exchanger, it is important that the K_{d} -values of the elements to be separated are large, but a too large K_d -value for lithium leads to broadening of the lithium peak in the effluent. 1 M hydrochloric acid was chosen as eluant. The K_d - and β -values derived from figures 1 and 2 for this medium were summarized in Table I.

3.3. Determination of purification factors (P.F.)

3.3.1. Method of separation

Separations were performed on 3 cm diameter quartz columns filled with a quantity of resin equivalent to 80 grams of ovendried Dowex 50W-X16 (\approx 400 meq.). Before the separation begins the resin is washed with bidistilled water, until chloride ions are fully removed.

On the other hand 0.5 g of lithium and 100 to 500 /ug of a traced alkali- or alkali earth metal are absorbed on 16 gram ovendried Dowex 50W-X16 (≈ 80 meq.) by mixing during 30 minutes. After settling of the resin and decantation of the solution, the wet resin is transferred to the top of the column. The elution is performed with (1.00 ± 0.02) M hydrochloric acid.

The effluent is collected in 50 ml polyethylene recipients.

3.3.2. Determination of purification factors

Separations of $\text{Li}/^{22}$ Na, $\text{Li}/^{42}$ K, $\text{Li}/^{7}$ Be, Li/Mg and $\text{Li}/^{45}$ Ca mixtures were made under the conditions described above. Generally 85-90% of the lithium were localized in the fractions 6-9 or 7-10, and the concentration was at a maximum in fraction 8 or 9.

Purification factors (% lithium / % tracer) for the working conditions were determined and summarized in Table II. From these factors it can be derived that under the given conditions the separation is quantitative for all alkali- and alkali earth metals present in lithium at the 0.1% level.

3.4. Purification of AG 50W-X16

3.4.1. Introduction

A first test separation using AG 50W-X16 (50/100 mesh), twice washed with isopiëstically distilled 4 N HCl, was done under the conditions described in 3.3.1. Isopiëstically distilled 1 N HCl was used as eluant. The results are summarized in Table III.

The high calcium content of the so-called purified product could be explained by emmission spectrographic analysis of the AG 50W-X16, which contained up to 1 eq. % of calcium. The residual impurity level in the purified lithium is due to two sources (fig. 3): M₁ depending on the purification factor (P.F.) and M₂ proportional to the impurity content of the ion exchanger and inversely proportional to the distribution coëfficient (K_d) of this impurity. In our case - high P.F. and impurity content of impurified lithium in the 0.1 % range the first factor is negligible, and only the resin purity has an influence on the purity of the purified lithium. For this reason it was important to improve the purity of the ionexchanger used.

Two methods were tested for this purpose:

- batch operations with hydrochloric acid of appropriated concentration
- batch operations with E.D.T.A. at appropriated pH.

3.4.2. Hydrochloric acid method

From fig. 1 it appears that the K_d-values are propitious between 3 and 6 M for calcium, and at high molarities for the other elements of interest. Further it is advantageous to take the ratio HCl : resin as high as possible for each batch operation. This is limited by the necessity to use important quantities of extra pure acid, and by the purity of the acid used.

It was decided to proceed with a 5 : 1 ratio 5 M HCl air dried AG 50W-X16.

Test purifications were carried out with 'suprapur' HCl (Merck A. G. /Germany) and with isopiëstically distilled HCl. 10 gram fractions of resin were mixed for two hours by magnetic stirring with 50 ml HCl in closed teflon containers and decanted after settling. After each batch operation the resin was rinsed with 50 ml bidistilled water. After the last batch operation this rinsing was repeated three times.

The resin was then transferred to a platinum crucible, dried for one night at 100°C and calcined. Impurities were determined by emission spectrography. The results summarized in Table IV show that the calcium contamination is lowered by a factor 20-40. No important lowering of the impurity content of the other alkali metals is obtained.

3.4.3. E.D.T.A. method

Ethylene diamine tetra acetic acid being sparingly soluble in water, and the stability of its calcium complex increasing with increasing pH, it seemed necessary to neutralize EDTA with a base.

 NH_4^{OH} has the inconvenience that eventually contaminating NH_4^{\dagger} ions cannot directly be determined by emission spectrography or activation analysis.

Finally LiOH was preferred: it can easily be determined by emission spectrography, and the Li⁺-ion is the easiest one to rinse from the ion exchange resin with hydrochloric acid.

3.4.3.1. Influence of the pH

Batch operations on 10 g of air-dried resin (approx. 30 meq.) were carried out in presence of different amounts of lithium hydroxide. The ion exchanger was mixed for 2 hours with 2.92 g of EDTA (10 mmol) and 30, 40, 50, 60 and 70 mmol of LiOH in 50 ml of water. As the distribution coefficient of lithium for AG 50W-X16 is very high in neutral- and alkaline medium, one can consider that 30 mmol of LiOH neutralizes the resin, and the rest forms resp. H_4Y , LiH₃Y, Li₂H₂Y, Li₃HY and Li₄Y.

After settling of the resin and decantation of the solution, the resin was rinsed with bidistilled water to eliminate E.D.T.A. Two other batch operations with 30% HCl 'suprapur' were made to eliminate Li, Na, K and Mg, which have very low distribution coefficients in this medium. The K_d value of Ca being high, one can easily appreciate the influence of the E.D.T.A. treatment on the elimination of calcium from the resin (Table V).

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Two batch operations with $\text{Li}_4 Y$, followed by two others with 30 % hydrochloric acid (Merck AG-Suprapur) allow rather good purifications of AG 50W-X16.

Nevertheless, it was necessary to perform a supplementary rinsing of the purified ion-exchanger with 1 M 'suprapur' HCl to eliminate completely the lithium ions. This supplementary elution leaded to a gain of a factor 10 on the sodium elimination too.

A comparison of the impurity levels of the original resin versus the purified resin is given in Table VI.

4. <u>Results and Conclusion</u>

400 gram aliquots of AG 50W-X16 cation exchanger were purified by successive batch operations with:

- 1. 400 mmol E.D.T.A. and 2800 mmol LiOH in a total volume of 2 1.
- 2. 400 mmol E.D.T.A. and 1600 mmol LiOH in a total volume of 2 1.

3. 2 1 30% 'Suprapur' hydrochloric acid.

4. 21 30% 'Suprapur' hydrochloric acid.

After the first three batch operations the ion-exchanger was washed with 11 bidistilled water, and after the third one three times. Then, quartz columns were filled with quantities equivalent to 80 grams of the oven-dried resin, (\approx 400 meq.) and eluted with 3 1 1M. hydrochloric acid and 0.5 1 bidistilled water. The whole operation was made in a glove box to avoid contamination from the atmosphere. On the other hand, quantities of 25 g air-dried unpurified AG 50W-X16 (\approx 80 meq.) were mixed for 30 minutes with approximately 75 mmol LiOH solution in 20-25 ml. After settling of the lithium-charged resin, the supernatant

solutions were decanted, and the wet resins transferred to the top of the different columns.

Elutions were performed with (1.00 ± 0.02) M HCl 'suprapur', and eluates collected in polyethylene recipients of 50 ml. 1 ml aliquots were analysed by transformation to and weighing as lithium sulfate.

Generally, 80 - 90% of the starting lithium were recovered in fractions 7-9 or 8-10 of the eluate, with maximum concentration in fractions 8 or 9.

2 g ⁷Li and 1.8 g ⁶Li were purified following this scheme, and analysed by emission spectrography (Table VII).

As can be seen from Table VII the total alkali- and alkaliearth level can be reduced to approximately 50 /ug/g Li, which is low enough to allow accurate transformations of lithium into sulfate, without measurable hydroscopy (<0.01%). It is important that this can be done without measurable contamination using 'suprapur' sulfuric acid. This allows one to prepare lithium sulfate containing as few as 3-5/ug/g Li₂SO₄ alkali- and alkali-earth metals, which can be used for the preparation of spectrographic standards. The silicium found is probably due to the utilization of quartz columns. The use of teflon columns will probably eliminate this, but the quantity found is unimportant for volumetric as well as for gravimetric analyses with precisions of 0.02 % ⁽¹⁾.

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TABLE I

Element	Distrib. coëff. K _d	Sep. factor $\beta_{M/Li}$
Ti	2 45	1
	2.45	
Na	6.4	2.6
к	22.3	9.1
Be	9.6	3.6
Mg	14.0	5.7
Ca	230	94

 K_d and p-values for Dowex 50W-X16 in 1 M HCl

TABLE II

Purification factors for Li/Na, K, Be, Mg, Ca for the working conditions

Separation	% of Lithium in the 4 maxim	% of tracer num fractions	P.F.
Li/ ²² Na	86 %	0.3 %	280
Li/ ⁴² K	89 %	< 0.9 %	> 100
Li/ ⁷ Be	90 %	0.003 %	30.000
Li/Mg	90 %	< 1.1 %	> 80
Li/ ⁴⁵ Ca	85 %	< 0.04 %	> 2150

TABLE III

Results of a test separation using AG 50W-X16, twice washed with isopiestically distilled 4 N HCl

Element	ppm before separation	Expected (based on PF)	ppm dtd by Em. Spectrogr. after separation
Na	260	< 1	
K	90	< 1	
Mg	720	< 10	
Ca	120	< 1	

TABLE IV

Elimination of Ca from AG 50W-X16 by batch operations with 5 M HCl

Number of	ppm in relation to oven-dried resin		
batch operations	HCl 'suprapur'	HCl 'isop.dist.'	
0	≈1000	≈ 1000	
1	512	880	
2	192	400	
3	168	188	
4	76	90	
5	52	-	
6	30	17	

TABLE V

Influence of the neutralization of E.D.T.A. on the elimination of calcium from AG 50W-X16

Neutr. of E.D.T.A.	ug Ca/g oven dried resin
H ₄ Y	212
LiH ₃ Y	96
Li ₂ H ₂ Y	38
Li ₃ HY	9
Li ₄ Y	5

TABLE VI

Comparison of the impurity levels of the original resin versus the purified resin

Element	Original resin (/ug/gram of over	Purified resin n dried resin)	
Na	10	< 0.05	
К	8	< 0.1	
Rb	< 8	<< 0.25	
Cs	< 8	<< 1	
Mg	4	~ 0.5	
Ca	≈1000	0,15 - 0,3	
Ba	. 4	< 0.15	
Sr	4	< 0.1	
Si	1	2.5	
Fe	< 4	0.3	
Al	- 4	≈ 1	
Cu	4	≈0.5	

TABLE VII

Purity of ⁶Li and ⁷Li purified by the described method

Element	/ ^{ug} / Lithium-6	g Li Lithium-7
Na	10	25
K	5	25
Mg	15	15
Ca	< 5	< 5
Si	50	<u><</u> 50
Fe	20	<u>≤</u> 50
other metals	not detectable by emission spectrography	



Fig.1 K_d-values of Li, Na, K, Be, Mg, Ca on Dowex 50W_X16 in hydrochloric acid medium





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Alfred Nobel

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