

PATENT SPECIFICATION

1,180,921

NO DRAWINGS.

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COMPLETE SPECIFICATION.

Improvements in Separation Procedures.

We, THORIUM LIMITED, a British Company, whose Registered Office is at Austral House, Basinghall Avenue, E.C.2, in the City of London, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a process for the separation of the element yttrium (or a compound thereof), from rare-earth elements found associated with it.

Yttrium is always found in nature associated with rare-earth elements, and is chemically very similar to many of these. It is desirable to devise a separation procedure which will permit the production of high-purity yttrium compounds, since these are used, together with europium compounds, as constituents of the red phosphor used in color television receivers.

The present invention provides a batch method for the separation of a solution containing yttrium ions in a highly pure state from an initial solution containing yttrium ions and rare earth metal ions, which method comprises:—

- (I) (a) multistage countercurrent liquid-liquid extraction between:
 - (i) an aqueous solution of salts of rare earth metals and of yttrium, the salts being inorganic acid salts,
 - (ii) a hydrocarbon solution of a branched chain carboxylic acid the acid being branched at the carbon adjacent the —COOH group and the solution containing dissolved in it rare earth metal ions and yttrium ions;

the extraction being carried out under conditions such that any solutes which leave each end of the extraction apparatus are continuously returned to that end from which they were derived whereby in time a separation is attained in the system

- (b) stopping the countercurrent solvent flows when sufficient separation has been achieved and emptying one or more stages of the extraction system containing yttrium ions together with some rare earth metal ions as impurities
- (c) isolating the yttrium and rare earth values thus obtained and where necessary converting to nitrates
- (d) multistage countercurrent liquid-liquid extraction between:
 - (i) a solution of nitrates of the yttrium and rare earth elements thus obtained, in tributylphosphate,
 - (ii) an aqueous solution of said nitrates of yttrium and rare earth elements,

the extraction being carried out under conditions such that any solutes which leave each end of the extraction apparatus are continuously returned to that end from which they were derived whereby in time a separation is attained in the system

- (e) stopping the countercurrent solvent flows when sufficient separation has been achieved, and emptying one or more stages of the extraction apparatus containing a solution of suitably pure yttrium; or

(II) effecting the consecutive stages as set

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forth above but in stage (a) using the solvent pair (d) (i) and (d) (ii) and in stage (d) using the solvent pair (a) (i) and (a) (ii) wherein said salts are nitrates.

5 In commercial practice high purity yttrium salts can be obtained in two such cycles, although it is within the scope of the invention to use three or more cycles.

10 An advantage of performing the extraction in the first-mentioned order stated is that the yttrium is recovered from the end stages of the extractor system used for the second extraction step.

15 In the first step of this form of the process, the salts are preferably salts of hydrochloric or nitric acid, while the branched-chain tertiary carboxylic acid may be the mixture of C_9 , C_{10} and C_{11} tertiary carboxylic acids known under the Registered Trade Mark of "Versatic 911". This tertiary carboxylic acid is diluted with a hydrocarbon such as kerosene or xylene.

20 After the yttrium-rich stages have been removed from this first step they are preferably treated with a dilute mineral acid (preferably hydrochloric or nitric) so as to recover the yttrium in the aqueous phase at a concentration of 100–500 gm/litre. It is to be noted that both the organic and the aqueous phases may be subjected to this treatment. The yttrium-rich extract will also contain such elements as gadolinium and dysprosium and these may be located in both the aqueous and organic phases. These are thus extracted into this aqueous mineral acid solution.

25 The adjustment of the concentration of the aqueous solution entering stage (d) is important. Preferably a concentration of not more than 160 g/l, for example from 80 to 120 g/l of yttrium and rare earth elements as oxides is used, which allows a clean separation of yttrium from the elements associated with it in the extract from stage (b). In this stage also the tributyl phosphate may be diluted with suitable hydrocarbon solvents.

30 A feature upon which the practice of this invention depends is the utilization of the concept of operating such a conventional multi-stage liquid-liquid extraction unit at "total reflux". This concept is more fully explained in our British Patent No. 1,026,791. As is disclosed therein almost and of the standard forms of extraction apparatus can be used, but this form of working is particularly suited to the devices known as "mixer-settlers". It will be appreciated that in operation under total reflux, the system gradually approaches equilibrium. Extraction can be stopped when substantial equilibrium has been reached, or earlier, when the desired solute has been sufficiently separated.

65 Since the solvent systems used in stages

(a) and (d) are not the same, different methods of achieving total reflux may have to be used. In the case of stage (a) particularly useful methods are as follows. The aqueous solution leaving the extractor unit is treated with an alkali (by which term is included ammonia) and the carboxylic acid, so as to yield a solution of yttrium ions dissolved in excess carboxylic acid. The hydrocarbon solvent may be present during these operations. The hydrocarbon solution leaving the other end of the extractor unit is treated with an aqueous mineral acid solution thus decomposing the carboxylic acid salt in the organic phase, and transferring the rare-earth metal ions and yttrium ions to the aqueous mineral acid solution. After suitable concentration, if necessary, this solution is then returned to the same end of the extractor as that from which the organic solution was derived; the hydrocarbon solution of carboxylic acid can be used to obtain reflux at the other end of the extractor system. In the case of stage (d) particularly useful methods are as follows. The aqueous solution leaving the extractor unit passes through an evaporator to concentrate it, and the concentrate thus obtained is returned to the same end of the extractor unit as that from which it was derived. The organic solution leaving the extractor unit is passed to a smaller second mixer-settler unit, and contacted therein counter-currently with a much larger flow of water; this transfers all the rare earth metal ions and yttrium ions into the aqueous phase. After suitable concentration this is then returned to the extractor unit at the same end as that from which the organic solution was derived.

Other methods of achieving "total reflux", such as chemical precipitation (followed by filtration and redissolution in the other solvent), or total evaporation of solvent (followed by redissolution in the other solvent), may be used; and, optionally, recycling of the solvents — which is to be clearly distinguished from the refluxing of solutes treated above — may also be used. These problems are clearly explained in our British Patent No. 1,026,791 mentioned above.

The process of the present invention uses two separate solvent extraction steps, carried out under different conditions of solvent and extractant. Whilst yttrium, although not a lanthanide rare-earth metal displays a chemistry closely similar to those of the lanthanide rare-earth metal we have now surprisingly discovered that its behaviour in liquid-liquid solvent extraction systems is markedly dependent on:

- (a) the solvents in use;
- (b) the extractant in use.

In determining whether or not two species can be separated by liquid-liquid counter-current solvent extraction, the necessary

physical parameter is the separation factor. This is defined as the ratio of the partition coefficients of two species between the two solvents concerned, and is linked to the relative concentration in the extractor unit by the equation:

$$\frac{A_1/B_1}{A_n/B_n} = \beta^{(n-1)}$$

Where: A, B: components being separated;
 10 A_1/B_1 : is their ratio in the first mixer-settler;
 A_n/B_n : is their ratio in the nth mixer-settler

and β is the separation factor. (Varteresian and Fenske, Ind. Eng. Chem. 29, 270 (1937).) Even the use of apparently chemically similar extractant species causes marked changes in the determined value of β , as is shown in the following Table. This table shows the separation factors between yttrium and rare-earth metals normally associated with it, using aqueous nitrates as one phase, and a number of organic phases. The results are calculated relative to β for yttrium being 1.0.

Extraction	System Concentration	Relative Separation Factor									
		Yb	Y	Er	Ho	Dy	Nd	Gd	Sm		
Organic phase—tributyl phosphate (g/l in aqueous phase)	... 38.4	0.9	1.0	2.16	3.02	3.95	4.12	4.45	5.55		
	54.7	Yb	Y	Er	Ho	Dy	Nd	Gd	Sm		
		0.9	1.0	1.75	2.3	2.8	2.88	3.3	4.1		
Aqueous phase water	... 100.0	Yb	Y	Er	Nd	Ho	Gd	Dy	Sm		
		0.85	1.0	1.4	1.45	1.8	2.1	2.1	2.45		
	147.0	Yb	Y	Nd	Er	Ho	Gd	Dy	Sm		
		0.9	1.0	1.15	1.25	1.65	1.83	1.9	1.95		
	215.0	Yb	Y	Et	Sm	Gd	Ho	Dy			
		0.85	1.0	1.05	1.3	1.45	1.5	1.65			
	283.0	Nd	Y	Gd	Ho	Dy					
		0.55	1.0	1.2	1.4	1.47					
	about 480	Nd	Y	Er	Ho	Dy					
		0.39	1.0	1.15	1.2	1.3					
(sat'd aqueous sol'n)											
Iso-octanoic acid in xylene		Nd	Y	Sm	Gd	Er	Ho	Dy	Yb		
		0.68	1.0	1.38	1.38	1.93	1.94	1.98	2.1		
"C ₇ C ₃₀ " Acid in xylene		Nd	Y	Gd	Sm	Dy	Ho	Er	Yb		
		0.76	1.0	1.12	1.24	1.70	1.77	1.95	2.7		
Naphthenic acid 180 in "Shellsol" A		Nd	Y	Sm	Gd	Dy					
		0.88	1.0	1.58	1.74	2.4					
Naphthenic acid 230 in "Shellsol" A		Nd	Y	Gd	Sm	Dy					
		0.70	1.0	1.17	1.26	2.0					
"Versatic" acid "911" in "Shellsol" A		Nd	Y	Dy	Ho	Er	Yb				
		0.34	1.0	1.87	2.11	2.90	3.7				
"Versatic" acid 15/19 in "Shellsol" A		Pr	Y	Yb	Dy	Ho	Er	Yb			
		0.27	1.0	1.18	1.30	1.96	2.50	2.51			

The aqueous phase in every case was a solution of nitrates. The organic acids are all commercially available materials, "C₇C₂₀" acid comprises a mixture of predominantly secondary branched-chain carboxylic acids having from 7 to 20 carbon atoms per molecule. The naphthenic acids are secondary carboxylic acids derived from tetralin, having molecular weights of approximately 180 and 230 respectively. The "Versatic" acids are commercially available mixtures of tertiary branched chain carboxylic acids having from 9 to 11, and from 15 to 19, carbon atoms per molecule respectively. "Shellsol" is a commercially available material containing mainly a mixture of trimethyl benzenes.

These results clearly demonstrate that versatic acid 911, which is a tertiary branched chain carboxylic acid, gives the widest possible spread in the values of the separation factor. We have found that a given acid is only efficient if it

(a) is either largely insoluble in water (where water is to be the other solvent), or resides almost totally in the organic solvent under these working conditions (this sets a lower limit on the number of carbon atoms in the molecule, and it is preferred to use an acid having at least 8 carbon atoms per molecule),

(b) is capable of giving a suitably concentrated solution of rare earth ions in the organic solvent without causing a large increase in viscosity, (this effect becomes important above about 20 carbon atoms in the molecule). A single acid, or a mixture of acids, is equally effective.

The list of separation factors shows that the tertiary carboxylic acid "Versatic acid 911" separates samarium partially, and gives very little separation from gadolinium. Terbium (not listed) falls between gadolinium and dysprosium and also is not separated, whilst dysprosium and all the others are separated from yttrium. On the other hand, the tributyl phosphate process shows that the separation obtainable is markedly concentration variable, with two exceptions: yttrium is substantially unaffected, and erbium also stays close to yttrium.

Therefore by combining together the tributyl phosphate aqueous nitrates system and one of the organic carboxylic acid systems, separation of yttrium from the rare earth metals normally associated with it can be achieved.

In particular, by combining together a system using a branched chain carboxylic acid, and the tributyl phosphate-aqueous nitrates system at a suitable concentration, complete separation of yttrium from the rare earth metals normally associated with it can be achieved. Preferentially, the two stages

are employed in this order, but the reverse sequence is equally effective.

The concentration of the solutes in the aqueous phase of the tributyl phosphate/aqueous stage of the process depends in part on the purity of the desired yttrium compound. We obtain 99.999% pure yttrium oxide (after suitable isolation procedures) using a solute concentration in the aqueous phase of 100 g/l. Clearly, lower solute concentrations could result in yttrium of as good or better purity, but a limit on the dilution is set by the need to produce the yttrium compound in reasonable quantities. In general, we prefer a solute concentration in the aqueous phase of not more than 120 g/l when 99.999% purity is desired. When the product need only be of 99.99% purity, higher solute concentrations of up to, for example, 160 g/l or even higher may be employed.

It is believed that tertiary branched-chain carboxylic acids have not previously been employed in liquid/liquid extraction systems. Their advantages, particularly the very wide spread of relative separation factors found for rare earth metal ions in them, are clearly shown in the preceding description.

The following Examples 2 and 3, taken together illustrate the invention:

EXAMPLE 1

Preparation of an Yttrium concentrate:

Solutions were prepared by the methods given below, from an oxide mixture of the following composition:

Oxide	%
Y ₂ O ₃	43.3
Sm ₂ O ₃	0.7
Gd ₂ O ₃	6.7
Dy ₂ O ₃	12.7
Ho ₂ O ₃	2.0
Er ₂ O ₃	7.1
Yb ₂ O ₃	2.5
Pr and Nd oxides	25.0

To 1500 gm of this mixture was added 200 gm of mixed Pr₂O₃ in order to provide a mixture more closely approximating commercial raw material.

(a) Organic phase: An equal volumes mixture of Versatic Acid 911 and "Shellsol" A was contacted with a concentrated solution of the mixture of oxides dissolved in aqueous nitric acid, and transfer of the metal ions of the organic phase obtained by adding .880 ammonium hydroxide solution. Contacting was continued until the organic phase contained the equivalent of 40 gm/litre of dissolved oxides.

(b) Aqueous phase: A solution of 250 gm/litre of oxides in 5.3 molar nitric acid in water was made up.

The mixer-settlers used held about 300 ml; for the 25 stage unit used about 1,500 gm. of oxides was required. After filling, flows of 52 ml./min. of organic phase, and 12 ml/min. of aqueous phase were set up, and the extractor operated at total reflux for 22 hours. Analysis of the stages (aqueous phase only given; organic phase differs only by the relevant partition coefficient) gave the following results.

Unit No.	Analysis, % as oxides								
	Pr	Nd	Sm	Gd	Y	Dy	Ho	Er	Yb
1	31.8	64.3							
4	17.3	75.7	1.2	4.8					
6	7.6	58.1	3.5	25.9	9.8				
7	4.0	33.6	3.9	32.4	25.6				
10	0.2	2.4	1.5	17.4	75.0	3.3			
13		0.2	0.3	4.3	86.0	8.5	0.7	0.1	
16				1.0	79.5	16.8	1.7	0.9	
19					63.1	25.6	4.0	5.2	
22					38.2	29.0	6.6	18.3	7.8
25					29.2	23.2	6.9	23.7	17.0

Maximum purity of yttrium reached is 86.0 in stage 13.

These data represent internal ratios; since yttrium is of a lower molecular weight, whilst solution molarity is preserved in all of the stages (excepting the end 2 or 3 which are perturbed by the reflux processes), variations in solution strength in gm/litre do occur, the loading being a minimum at maximum yttrium content.

The important impurities are neodymium and erbium; this 25 stage unit does not provide a sufficiently pure concentrate.

EXAMPLE 2

Preparation of a usable yttrium concentrate.

The above experiment was repeated, using a mixture of primarily neodymium, samarium, dysprosium, erbium, holmium, yttrium and gadolinium oxides. The aqueous solution of nitrates was made up to 1.33 molar, as $\text{Re}(\text{NO}_3)_3$, the term "Re" being accepted in this art as meaning any, or a mixture of, rare earth ions, and in this particular instance for convenience in writing the formula, includes yttrium ions. When equilibrium had been reached, analysis of the stages gave the following results (aqueous phase only given), for the 50 stage unit used,

Unit No.	Analysis, as oxides						
	Nd	Sm	Gd	Y	Dy	Ho	Er*
1	Perturbed due to reflux						
4	32.3	33.9	33.8	0.1			
7	4.4	37.2	57.5	0.9			
10	0.4	27.5	65.9	6.2			
13		13.8	51.8	34.3	0.1		
16		3.2	18.9	77.5	0.3		
19		0.4	3.4	95.3	1.0		
22			0.6	97.5	1.8		
25			0.1	96.6	3.2	0.1	
28				94.8	5.1	0.1	
31				91.5	8.2	0.3	
34				87.0	12.4	0.5	
37				78.9	19.9	1.2	0.1
40				65.8	31.3	2.6	0.3
43				45.7	46.4	5.8	2.1
45				30.6	53.4	9.8	7.2
47				16.8	51.8	11.4	20.1
50				4.1	30.5	10.2	55.2

* contains other earths up to Lu_2O_3 inclusive.

Thus units 11-36 contain yttrium at a purity suitable for the second stage of the process.

The final solutions contain no free nitric acid.

EXAMPLE 3

- 5 Purification of an yttrium concentrate
After suitable chemical treatment to provide the concentrate as nitrates, the following solutions were made up:

- 10 (a) Organic phase: tributyl phosphate, diluted with an equal volume of Shell-sol A, containing 21 gm/litre of solutes, expressed as oxides (approximately 40 gm/litre as nitrates, depending on the precise yttrium content).
- 15 (b) Aqueous phase: aqueous solution of nitrates, containing 100 gm/litre as oxides (approximately 200 gm/litre of nitrates, depending on the precise yttrium content).
- 20

This mixture was then charged into a 50 unit extractor system, and extraction at total reflux carried out; reflux was obtained by:

Aqueous solution: concentration to 380 gm/litre of $\text{Re}(\text{NO}_3)_3$.

Organic solution: extraction with a large volume of water, and concentration of this solution to 190 gm/litre of $\text{Re}(\text{NO}_3)_3$.

The flow rates used were 10 ml/min. of the organic phase and 4.6 ml/min of the aqueous phase.

At equilibrium, analysis of the aqueous phases in the units gave the following results:

	Unit No.	Y_2O_3	Er_2O_3	Others
40	1	greater than 99.99	nil	nil
	25	greater than 99.99	nil	nil
	30	greater than 99.98	0.01	0.01
	35	greater than 99.95	0.01	0.04
	40	greater than 99.88	0.01	0.11
45	45	greater than 99.35	0.02	0.63
	48	96.4	0.03	3.57
	50	87.8	0.05	12.15

- Typical analysis of the rare-earth mixture present with the yttrium in the refluxed solutes, i.e. in the outgoing organic phase, are as follows:

Element	Content, % ... (relative)
Pr	less than 1.0
55 Nd	less than 1.0
Sm	10.0
Eu	1.1
Gd	13.3
60 Tb	14.9
Dy	7.6
Ho	ca 0.4
Er	0.2
Y	ca 51.0

- 65 Having regard to the provisions of Section 9 of the Patent Act, attention is directed to the claims of Patent No. 51035/69 (Serial No. 1,180,922).

WHAT WE CLAIM IS:—

- 70 1. A batch method for the separation of a solution containing yttrium ions in a highly pure state from an initial solution containing yttrium ions and rare earth metal ions, which method comprises:—

- (I) (a) multistage countercurrent liquid-liquid extraction between:
- 75 (i) an aqueous solution of salts of rare earth metals and of yttrium, the salts being inorganic acid salts,
- 80 (ii) a hydrocarbon solution of a branched chain carboxylic acid the acid being branched at the carbon adjacent the $-\text{COOH}$ group and the solution containing dissolved in it the rare earth metal ions and yttrium ions.
- 85

the extraction being carried out under conditions such that any solutes which leave each end of the extraction apparatus are continuously returned to that end from which they were derived whereby in time a separation is attained in the system

90

- (b) stopping the countercurrent solvent flows when sufficient separation has been achieved and emptying one or more stages of the extraction system containing yttrium ions together with some rare earth metal ions as impurities
- 95
- (c) isolating the yttrium and rare earth values thus obtained and
- 100

Thus units 11—36 contain yttrium at a purity suitable for the second stage of the process.

EXAMPLE 3

- 5 Purification of an yttrium concentrate
After suitable chemical treatment to provide the concentrate as nitrates, the following solutions were made up:

- 10 (a) Organic phase: tributyl phosphate, diluted with an equal volume of Shell-sol A, containing 21 gm/litre of solutes, expressed as oxides (approximately 40 gm/litre as nitrates, depending on the precise yttrium content).
- 15 (b) Aqueous phase: aqueous solution of nitrates, containing 100 gm/litre as oxides (approximately 200 gm/litre of nitrates, depending on the precise yttrium content).
- 20

The final solutions contain no free nitric acid.

This mixture was then charged into a 50 unit extractor system, and extraction at total reflux carried out; reflux was obtained 25 by:

Aqueous solution: concentration to 380 gm/litre of $\text{Re}(\text{NO}_3)_3$.

Organic solution: extraction with a large volume of water, and concentration of this solution to 190 gm/litre of $\text{Re}(\text{NO}_3)_3$. 30

The flow rates used were 10 ml/min. of the organic phase and 4.6 ml/min of the aqueous phase. 35

At equilibrium, analysis of the aqueous phases in the units gave the following results:

Unit No.	Y_2O_3	Er_2O_3	Others
40	1	greater than 99.99	nil
	25	greater than 99.99	nil
	30	greater than 99.98	0.01
	35	greater than 99.95	0.01
	40	greater than 99.88	0.01
45	45	greater than 99.35	0.02
	48	96.4	0.03
	50	87.8	0.05
			12.15

Typical analysis of the rare-earth mixture present with the yttrium in the refluxed solutes, i.e. in the outgoing organic phase, are as follows:

Element	Content, % ... (relative)
55 Pr	less than 1.0
Nd	less than 1.0
Sm	10.0
Eu	1.1
Gd	13.3
60 Tb	14.9
Dy	7.6
Ho	ca 0.4
Er	0.2
Y	ca 51.0

65 Having regard to the provisions of Section 9 of the Patent Act, attention is directed to the claims of Patent No. 51035/69 (Serial No. 1,180,922).

WHAT WE CLAIM IS:—

- 70 1. A batch method for the separation of a solution containing yttrium ions in a highly pure state from an initial solution containing yttrium ions and rare earth metal ions, which method comprises:—

- (I) (a) multistage countercurrent liquid-liquid extraction between: 75
(i) an aqueous solution of salts of rare earth metals and of yttrium, the salts being inorganic acid salts, 80
(ii) a hydrocarbon solution of a branched chain carboxylic acid the acid being branched at the carbon adjacent the $-\text{COOH}$ group and the solution containing dissolved in it the rare earth metal ions and yttrium ions. 85

the extraction being carried out under conditions such that any solutes which leave each end of the extraction apparatus are continuously returned to that end from which they were derived whereby in time a separation is attained in the system 90

- (b) stopping the countercurrent solvent flows when sufficient separation has been achieved and emptying one or more stages of the extraction system containing yttrium ions together with some rare earth metal ions as impurities 95
(c) isolating the yttrium and rare earth values thus obtained and 100



- where necessary converting to nitrates
- (d) multistage countercurrent liquid-liquid extraction between:
- 5 (i) a solution of nitrates of the yttrium and rare earth elements thus obtained, in tributylphosphate, (ii) an aqueous solution of said nitrates of yttrium and rare earth elements,
- 10 the extraction being carried out under conditions such that any solutes which leave each end of the extraction apparatus are continuously returned to that end from which they were derived whereby in time a separation is attained in the system
- 15 (e) stopping the countercurrent solvent flows when sufficient separation has been achieved, and emptying one or more stages of the extraction apparatus containing a solution of suitably pure yttrium; or
- 20 (II) effecting the consecutive stages as set forth above but in stage (a) using the solvent pair (d) (i) and (d) (ii) and in stage (d) using the solvent pair (a) (i) and (a) (ii) wherein said salts are nitrates.
- 25 2. A method as claimed in claim 1
- wherein solvent (a) (ii) is a hydrocarbon solution of a tertiary carboxylic acid.
- 30 3. A method as claimed in claim 2 in which the tertiary carboxylic acid has from 8 to 20 carbon atoms per molecule.
- 35 4. A method as claimed in any one preceding claim wherein the hydrocarbon is a mixture of trimethylbenzenes.
- 40 5. A method as claimed in any of claims 1 to 4 wherein the salts in aqueous solution (a) (i) are nitrates.
- 45 6. A method as claimed in any one preceding claim wherein the tributyl phosphate is used in solution in an organic solvent.
- 50 7. A method as claimed in claim 6 wherein said organic solvent is a mixture of trimethylbenzenes.
- 55 8. A method as claimed in any one preceding claim wherein the nitrate concentration (d) (ii) is not greater than 160 g./l., measured as oxide.
9. A method as claimed in claim 8 wherein said concentration is from 80 g./l. to 120 g./l. measured as oxide.
10. A method as claimed in claim 1 and substantially as herein described in Examples 2 and 3.
11. Yttrium when separated by the method claimed in any one of claims 1 to 10.

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