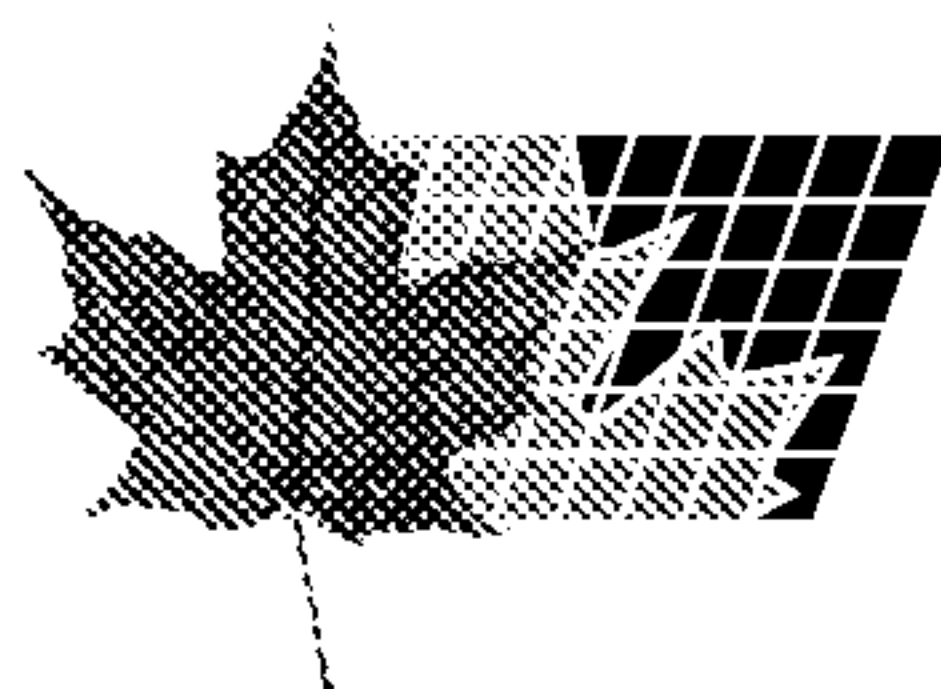




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(54) **PROCEDE DE PRODUCTION DE FLUORO-COMPLEXES**

**D'ALUMINIUM, LES FLUORO-COMPLEXES D'ALUMINIUM
AINSI OBTENUS AINSI QUE L'UTILISATION DU SECHAGE
PAR PULVERISATION ET DE POLYETHYLENE-GLYCOLS
POUR COMMANDER LA STRUCTURE DES FLUORURES
D'ALUMINIUM**

(54) **METHOD FOR THE PRODUCTION OF COMPLEX**

**FLUOROALUMINATE, THE FLUOROALUMINATE
PRODUCED THEREBY AND THE UTILIZATION OF SPRAY
DRYING AND POLYALKYLENEGLYCOLS FOR
CONTROLLING THE STRUCTURE OF
FLUOROALUMINATES**

(57) The invention relates to a method for producing a complex fluoroaluminate, wherein the fluoroaluminate is obtained by spray drying a suspension containing the fluoroaluminate, to the fluoroaluminate produced according to this method and to the use of said fluoroaluminate. The invention also relates to the utilization of spray drying for controlling the macroscopic structure of a fluoroaluminate and the utilization of a polyalkyleneglycol for identical purposes.



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(54) Title: METHOD FOR THE PRODUCTION OF COMPLEX FLUOROALUMINATE, THE FLUOROALUMINATE PRODUCED THEREBY AND THE UTILIZATION OF SPRAY DRYING AND POLYALKYLENEGLYCOLS FOR CONTROLLING THE STRUCTURE OF FLUOROALUMINATES (54) Bezeichnung: VERFAHREN ZUR HERSTELLUNG KOMPLEXER FLUOROALUMINATE, DIE HERGESTELLTEN FLUOROALUMINATE UND VERWENDUNG VON SPRÜHTROCKNUNG UND POLYALKYLENGLYKOLEN ZUR STEUERUNG DER STRUKTUR VON FLUOROALUMINATEN (57) Abstract The invention relates to a method for producing a complex fluoroaluminate, wherein the fluoroaluminate is obtained by spray drying a suspension containing the fluoroaluminate, to the fluoroaluminate produced according to this method and to the use of said fluoroaluminate. The invention also relates to the utilization of spray drying for controlling the macroscopic structure of a fluoroaluminate and the utilization of a polyalkyleneglycol for identical purposes. (57) Zusammenfassung Verfahren zur Herstellung eines komplexen Fluoroaluminats, wobei das Fluoroaluminat durch Sprühtrocknung aus einer das Fluoroaluminat enthaltenden Suspension erhalten wird, das durch dieses Verfahren herstellbares Fluoroaluminat und dessen Verwendung. Zusätzlich sind beansprucht die Verwendung der Sprühtrocknung zur Steuerung der makroskopischen Struktur eines Fluoroaluminats und Verwendung eines Polyalkylenglycols zum gleichen Zweck.				

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PROCESS FOR PREPARING COMPLEX FLUOROALUMINATES, THE
FLUOROALUMINATES PREPARED AND USE OF SPRAY DRYING AND
POLYALKYLENE GLYCOLS FOR CONTROLLING THE STRUCTURE OF
FLUOROALUMINATES

5

The present invention relates to a process for preparing a complex fluoroaluminate, with the fluoroaluminate being obtained from a suspension in which the fluoroaluminate is present by spray drying during the course of this process. The present invention likewise relates to a process in which the particle structure of this fluoroaluminate is controlled by means of a structure-influencing substance.

15 Fluoroaluminates are used in many areas of industry. Thus, for example, potassium tetrafluoroaluminate is used as an additive to abrasives, in glass production or as flux in industrial processes.

20 One way of preparing, for example, potassium tetrafluoroaluminate is disclosed in JP 08157212. In the process described there, aluminum hydroxide is reacted with 20% strength by weight hydrogen fluoride and the resulting solution, in which
25 tetrafluoroaluminic acid is present, is neutralized with KOH.

DE 31 16 469 describes a process in which an aqueous, HAlF_4 -containing solution is neutralized with KOH to form potassium tetrafluoroaluminate.

30 A problem which can occur in the preparation of tetrafluoroaluminates involves the particle structure, in particular the particle size of the fluoroaluminate obtained. Thus, for example, formation of coarse particles adversely affects the quality of the
35 fluoroaluminate required for certain applications in which amorphous structures in the lower μm range are necessary. Moreover, subsequently increasing the proportion of fines is uneconomical.

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A further problem which can occur in the preparation of tetrafluoroaluminates is the residual moisture which remains in the product and can have an adverse effect on the desired applications.

5 It is therefore an object of the present invention to provide a process for preparing fluoroaluminates which does not have these disadvantages.

10 The present invention provides a process for preparing a complex fluoroaluminate, wherein the fluoroaluminate is obtained from a suspension in which the fluoroaluminate is present by spray drying.

15 In this process, the suspension in which the fluoroaluminate is present is firstly fed to the spray dryer. It is preferably fed to the spray drying via a metering device.

20 In the process of the invention, there are in principle no restrictions in respect of the way in which the suspension in which the fluoroaluminate is present is sprayed. It is possible to use, for example, rotary disk atomizers, hydrodynamic introduction of the feed via single-fluid nozzles or introduction by means of compressed air via two-fluid nozzles. In one embodiment of the process of the invention, the
25 suspension in which the fluoroaluminate is present is, for example, fed via a controlled spindle pump to a spray dryer and introduced via a rotary disk atomizer having a diameter of 150 mm and a rotational speed of 16,000 rpm.

30 The product can likewise be discharged by all conceivable means. Examples which may be mentioned are cyclone discharge or discharge via one or more dedusting filter units.

35 The temperatures of the hot air stream employed in spray drying can be selected essentially freely and are in principle limited only by the melting point of the fluoroaluminate and the circumstances of the plant.

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The choice of temperature allows, inter alia, the drying capacity to be influenced and the residual moisture content of the spray-dried material to be controlled. This makes it possible to match the
5 moisture content of the fluoroaluminate to the demands made of the material by the user. In general, the temperatures are in a range from 100 to 500°C, which, for example, gives a relatively low residual moisture content of the fluoroaluminate of < 1% by weight.

10 It is of course also possible to employ two or more spray drying steps in the process of the invention.

It is likewise possible, in a modification of the process, to follow spray drying by a further drying
15 step according to the prior art, for example fluidized-bed drying.

A further advantage of the process of the invention is the influence on the particle structure of the fluoroaluminate which can be exercised by spray
20 drying. The choice of the dispersion device in the spray dryer makes it possible to influence the particle structure.

A further possible way of controlling the particle structure and in particular the particle size
25 distribution and also the floury appearance of fluoroaluminates generally required for practical use is to add a structure-influencing substance in the process of the invention at a suitable point in the preparation of the suspension in which the
30 fluoroaluminate is present.

The present invention therefore also provides a process as described above, characterized in that a structure-influencing substance is used in the preparation of the suspension in which the
35 fluoroaluminate is present.

A finely structured and floury appearance of the fluoroaluminate can be obtained when the fluoroaluminate is obtained from a solution which may

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comprise one or more precursors and the structure-influencing substance is added to the solution before the fluoroaluminate is obtained from the solution.

In particular, the structure-influencing substance is used in a process in which the fluoroaluminate is obtained as a solid by precipitation from the solution in which the fluoroaluminate is present.

The present invention accordingly provides a process, characterized in that it comprises the steps (i) to (iv):

- (i) preparation of a solution comprising a precursor of the fluoroaluminate;
- (ii) addition of the structure-influencing substance to the solution from (i);
- (iii) precipitation of the fluoroaluminate from the solution obtained from (ii) to give the suspension in which the fluoroaluminate is present;
- (iv) spray drying of the suspension obtained from (iii) to give the fluoroaluminate.

The precipitation is preferably carried out by addition of aqueous alkalis. Particular preference is given to precipitating the fluoroaluminate by addition of an aqueous potassium hydroxide solution.

The concentration of the aqueous potassium hydroxide solution is relatively uncritical and can extend from a very low concentration to the highest possible concentration. It is preferably in the range from 40 to 50% by weight.

It is likewise possible to use solutions which comprise not only KOH but also further K^+ -donating components. These may be, for example, K_2CO_3 or KCl.

In a preferred embodiment of the process of the invention, the solution is stirred during the precipitation of the fluoroaluminate. It is possible, if required, to optimize the particle structure of the fluoroaluminate by selection of a suitable stirrer.

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The temperature at which the fluoroaluminate is precipitated in the process of the invention is generally in the range from 0 to 100°C, preferably in the range from 60 to 90°C, particularly preferably in the range from 65 to 85°C, more particularly preferably in the range from 65 to 80°C and in particular about 70°C.

In the process of the invention, the pH of the suspension obtained after precipitation of the fluoroaluminate is set to a value which is preferably in the range from 4.5 to 7.0, particularly preferably in the range from 5.5 to 6.5 and in particular 6.

In the process of the invention, it is in principle possible to control the particle structure of all fluoroaluminates which can be prepared by the above-described process by the use of a structure-influencing substance.

According to the present invention, particular preference is given to controlling the particle structure in the preparation of tetrafluoroaluminates, in particular potassium tetrafluoroaluminate.

The present invention therefore provides a process as described above, characterized in that the fluoroaluminate is potassium tetrafluoroaluminate.

If the fluoroaluminate is prepared by the above-described process of the invention, the solution from step (i) can comprise any conceivable precursors from which this fluoroaluminate can be obtained.

In particular, the solution prepared in step (i) of the process of the present invention comprises tetrafluoroaluminic acid as precursor from which the potassium tetrafluoroaluminate preferably prepared is obtained.

The present invention therefore also provides a process as described above which is characterized in that the precursor of the fluoroaluminate is tetrafluoroaluminic acid.

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Such a solution comprising tetrafluoroaluminic acid can, for the purposes of the process of the invention, be prepared by all methods known from the prior art.

5 According to the present invention, this solution is preferably prepared from hydrated aluminum oxide and an aqueous solution of hydrogen fluoride. Use is generally made of commercial hydrated aluminum oxide. The Al_2O_3 content of the hydrated aluminum oxide
10 is preferably in the region of 65% by weight. It is of course also possible to use hydrated aluminum oxides having a lower concentration, for example those obtained from recycling plants.

 The hydrated aluminum oxide and the aqueous
15 solution of hydrogen fluoride are mixed with one another in such amounts that the molar ratio of Al:F is generally in a range from 1:3.9 to 1:4.5, preferably in a range from 1:4.0 to 1:4.4, particularly preferably in a range from 1:4.1 to 1:4.3 and in particular about
20 1:4.2.

 The concentration of the resulting solution of tetrafluoroaluminic acid is, in the process of the invention, set so that it is generally in a range from 5 to 40% by weight, preferably in a range from 10 to
25 30% by weight and particularly preferably in a range from 15 to 20% by weight. Depending on the concentration of the hydrated aluminum oxide used and the aqueous solution of hydrogen fluoride, it may be necessary to add additional solvent to the
30 tetrafluoroaluminic acid solution so as to bring the concentration to within the ranges described.

 Here, it is in principle possible to use all solvents which are suitable for this purpose and which do not interfere in the later isolation of the
35 fluoroaluminate. Preference is given to using water as solvent in the process of the invention.

 In step (ii) of the process of the invention, the structure-influencing substance is added to the

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solution comprising the precursor of the fluoroaluminate, in particular the tetrafluoroaluminic acid.

However, it is likewise conceivable to add the structure-influencing substance either to the hydrogen fluoride solution or to the solution in which the hydrated aluminum oxide is present or to both solutions prior to the preparation of the solution in which the precursor of the fluoroaluminate is present.

10 The structure-influencing substance can be added as a solid or as a liquid, depending on the physical state of the structure-influencing substance.

In the case of a solid structure-influencing substance, it is preferably firstly dissolved in a suitable solvent before the addition.

For the present purposes, the term "suitable solvent" means that the structure-influencing substance dissolves in this solvent and the solvent does not interfere in the later precipitation of the fluoroaluminate.

It is of course also possible to use a mixture of two or more suitable solvents. Particular preference is given to using water as solvent.

It is likewise conceivable to suspend a solid structure-influencing substance in a suitable liquid or in a suitable liquid mixture and to add the resulting suspension to the solution comprising the precursor of the fluoroaluminate.

Should the addition of the structure-influencing substance to the solution comprising the precursor of the fluoroaluminate be exothermic, it may be necessary to remove all or some of the heat generated by methods known from the prior art.

The amount of structure-influencing substance added to the solution obtained from step (i) is, in the process of the invention, calculated so that, based on the theoretical yield of fluoroaluminate, the concentration of the structure-influencing substance in

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the solution is generally in the range from 0.01 to 1% by weight, preferably in the range from 0.05 to 0.5% by weight and in particular in the range from 0.1 to 0.2% by weight.

5 For the purposes of the present invention, polyalkylene glycols has been found to be a particularly useful group of substances by means of which the particle structure of fluoroaluminates can be controlled.

10 The present invention accordingly provides a process as described above which is characterized in that the structure-influencing substance is a polyalkylene glycol.

15 Examples of polyalkylene glycols which may be mentioned are: polyethylene glycol, polypropylene glycol, polytetrahydrofurans, polypropylene glycol ethoxylates or polyethylene glycol propoxylates.

20 Depending on the desired particle structure, different polyalkylene glycols can be used. It is naturally also possible to use a mixture of two or more thereof.

25 It is likewise possible to use polyalkylene glycols having different molar masses. Thus, for example, it is conceivable to use polyalkylene glycol which is made up of molecules having a uniform degree of polymerization. It is naturally also conceivable to use mixtures consisting of collections of molecules having different molar masses.

30 Should it be necessary for the purposes of the way in which the process is carried out and/or the desired particle structure of the fluoroaluminate, it is of course also possible to use mixtures of two or more different polyalkylene glycols of which each can be molecularly uniform or polymolecular in the process
35 of the invention.

 The polyalkylene glycols used in the process of the invention can be prepared by all methods known from the prior art. An overview of the most important

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preparative methods may be found, for example, in Ullmanns Encyklopädie der technischen Chemie, Volume 19, 4th Edition, Verlag Chemie, Weinheim, 1980, pp. 31 to 38, the relevant contents of which are hereby fully
5 incorporated by reference into the present application.

Preference is given to using polyethylene glycol as structure-influencing substance in the process of the invention.

The present invention therefore also provides a
10 process as described above, characterized in that the polyalkylene glycol is a polyethylene glycol.

In general, the molar mass of the polyethylene glycol used is in the range from 200 to 40,000 g, preferably in the range from 400 to 25,000 g and in
15 particular about 20,000 g.

The present invention accordingly also provides a process as described above, characterized in that the polyethylene glycol has a molar mass in the range from 200 to 40,000 g.

20 The present invention likewise provides a complex fluoroaluminate which can be prepared by a process comprising step (I) below:

(I) Obtaining the complex fluoroaluminate from a suspension in which the fluoroaluminate is present
25 by spray drying.

In a preferred embodiment of the process of the invention, complex fluoroaluminates having a particle diameter which is generally in the range from 1 to 150 μm , preferably in the range from 1 to 100 μm , are
30 obtained.

Furthermore, the complex fluoroaluminates prepared according to the invention have a particle diameter distribution which has a reduced proportion of oversize particles compared to fluoroaluminates
35 prepared by processes of the prior art. In general, the maximum of the particle diameter distribution is in the range from 5 to 17 μm , preferably in the range from 7

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to 15 μm and more preferably in the range from 9 to 13 μm .

A further advantage of the process of the invention is that the reduced proportion of oversize
5 particles makes it possible to prepare fluoroaluminates for which further mechanical processing can be dispensed with.

The process of the invention offers, inter alia, the advantage that it gives complex
10 fluoroaluminates which have a lower melting point than complex fluoroaluminates prepared by a process according to the prior art. For example, the process of the invention makes it possible to prepare potassium tetrafluoroaluminate having a melting point which is in
15 the range from 540 to 550°C and is thus significantly below the melting point of the KAlF_4 - K_3AlF_6 eutectic. This melting point is far below the previously known melting points, which are in the range from about 560 to 575°C for commercial products.

20 The present invention accordingly also provides a process as described above which is characterized in that the potassium tetrafluoroaluminate has a melting point in the range from 540 to 550°C.

Since potassium tetrafluoroaluminate is used
25 predominantly as flux in hard soldering processes, the low melting range is of particular economic and industrial importance.

The present invention accordingly provides for the use of a complex fluoroaluminate which can be
30 prepared by a process as described above or a complex fluoroaluminate which can be prepared by a process comprising the step (I) as described above in the field of metallurgy.

The present invention likewise provides for the
35 use of a complex fluoroaluminate as described above, characterized in that it is used as flux, in particular in hard soldering processes.

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The present invention further provides for the use of spray drying for controlling the macroscopic structure of a complex fluoroaluminate.

The invention likewise provides for the use of
5 a polyalkylene glycol for controlling the macroscopic structure of a complex fluoroaluminate.

Furthermore, the invention also provides for the use as described above, characterized in that the polyalkylene glycol used is polyethylene glycol.

10 The present invention additionally provides for the use as described above, characterized in that the complex fluoroaluminate is potassium tetrafluoroaluminate.

The present invention is illustrated by the
15 following examples.

Examples

Example

20 1000 g of technical-grade hydrated aluminum oxide having an Al_2O_3 content of 65% by weight were reacted at room temperature with 2150 ml of an aqueous hydrogen fluoride solution having an HF concentration
25 of 42.6% by weight.

The solution obtained had, after addition of water, an HAlF_4 concentration of 18.0% by weight.

2.7 g of a polyethylene glycol having a molar mass of 20,000 g were added to the HAlF_4 -containing
30 solution.

1300 ml of a 45% strength by weight aqueous KOH solution were added to the above solution over a period of 10 minutes, with the temperature of the solution being maintained at 70°C.

35 The pH of the resulting suspension was set to 6 using an electronic pH measurement.

The suspension was subsequently spray dried at a temperature of 130°C.

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The particle size distribution of the potassium tetrafluoroaluminate obtained was determined using an HR 850-B granulometer from Cilas Alcatel (Figure 1a).

5 In Figures 1a and 1b, the diameter d in μm is plotted on the abscissa and the amount of material D below this size is plotted in % on the ordinate.

Comparative Example

10 The comparative example was carried out in the same way as the above example. The only difference was that no polyethylene glycol was added.

15 As in the above example, the particle size distribution was determined using an HR 850-B granulometer from Cilas Alcatel (Figure 1b).

20 Comparison of the particle size distributions of the example and the comparative example clearly shows that the proportion of oversize particles was significantly reduced when the structure-influencing substance polyethylene glycol was added.

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Claims

1. A process for preparing a complex fluoroaluminate, wherein the fluoroaluminate is
5 obtained from a suspension in which the fluoroaluminate is present by spray drying.
2. The process as claimed in claim 1, characterized in that a structure-influencing substance is used in the preparation of the suspension in which
10 the fluoroaluminate is present.
3. The process as claimed in claim 2, characterized in that it comprises the steps (i) to (iv):
 - (i) preparation of a solution comprising a
15 precursor of the fluoroaluminate;
 - (ii) addition of the structure-influencing substance to the solution from (i);
 - (iii) precipitation of the fluoroaluminate from the
20 solution obtained from (ii) to give the suspension in which the fluoroaluminate is present;
 - (iv) spray drying of the suspension obtained from (iii) to give the fluoroaluminate.
4. The process as claimed in claim 3,
25 characterized in that the precursor of the fluoroaluminate is tetrafluoroaluminic acid.
5. The process as claimed in any of claims 2 to 4, characterized in that the structure-influencing substance is a polyalkylene glycol.
- 30 6. The process as claimed in claim 5, characterized in that the polyalkylene glycol is a polyethylene glycol.
7. The process as claimed in claim 6, characterized in that the polyethylene glycol has a
35 molar mass in the range from 200 to 40,000 g.
8. The process as claimed in any of claims 1 to 7, characterized in that the fluoroaluminate is potassium tetrafluoroaluminate.

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9. The process as claimed in claim 8, characterized in that the potassium tetrafluoroaluminate has a melting point in the range from 540 to 550°C.

5 10. A complex fluoroaluminate which can be prepared by a process comprising step (I) below:

(I) Obtaining the complex fluoroaluminate from a suspension in which the fluoroaluminate is present by spray drying.

10 11. The use of a complex fluoroaluminate which can be prepared by a process as claimed in any of claims 1 to 9 or a complex fluoroaluminate as claimed in claim 10 as auxiliary in the field of metallurgy.

15 12. The use as claimed in claim 11, characterized in that the complex fluoroaluminate is used as flux, in particular in hard soldering processes.

13. The use of spray drying for controlling the macroscopic structure of a complex fluoroaluminate.

20 14. The use of a polyalkylene glycol for controlling the macroscopic structure of a complex fluoroaluminate.

15. The use as claimed in claim 14, characterized in that the polyalkylene glycol used is polyethylene glycol.

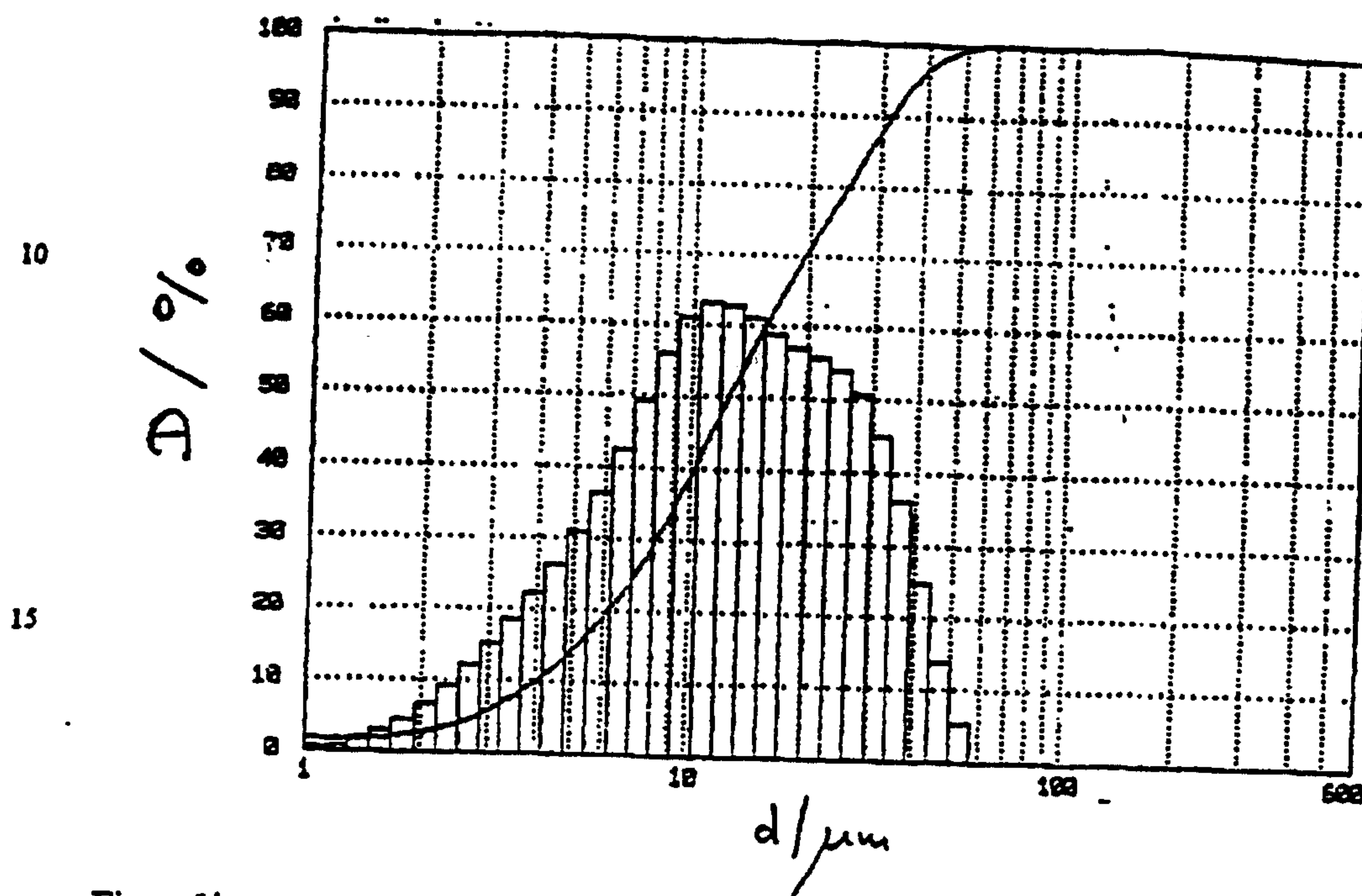
25 16. The use as claimed in any of claims 11 to 15, characterized in that the complex fluoroaluminate is potassium tetrafluoroaluminate.

PCT/EP99/07292
Riedel-de Haen GmbH

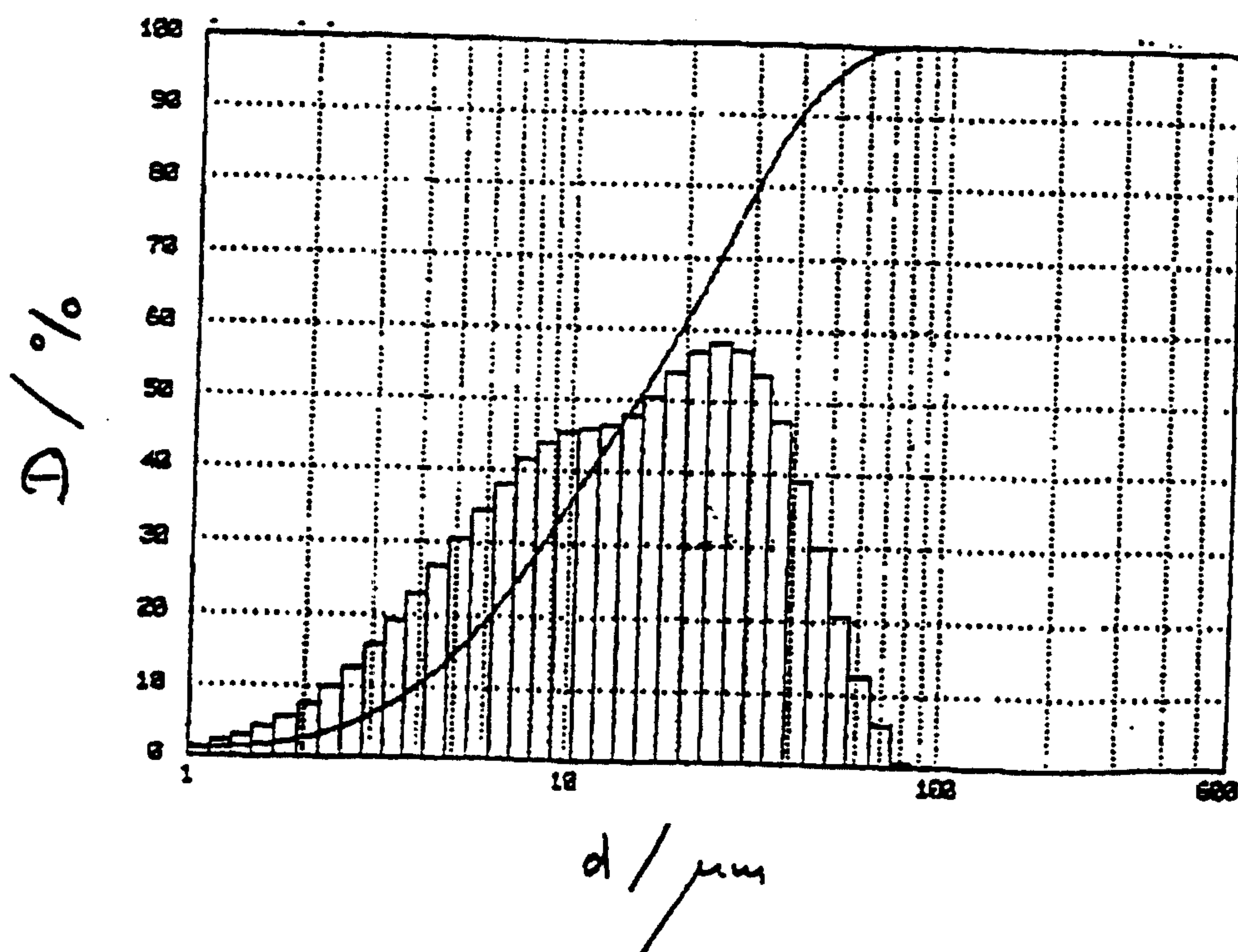
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s Figur 1a



Figur 1b



GEAENDERTES BLATT