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METHODS FOR GROWING ETHYLENE-DIAMINE TARTRATE CRYSTALS

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This invention relates to ethylenediamine tartrate crystals and more particularly to methods for growing the same.

One of the objects of this invention is to promote the growth rate of the crystals.

Another object of this invention is to provide crystals having essentially no tapering of the prism faces.

Still another object of this invention is to produce a more perfect crystal.

Yet another object of this invention is to stabilize the mother liquor against the appearance of spurious crystalline growth.

There are two common methods of growing ethylenediamine tartrate crystals, henceforth referred to as EDT crystals. These are: (1) by gradual reduction of temperature of a solution saturated at elevated temperature, and (2) by feeding a solution saturated at one temperature into a tank containing seed crystals kept at a constant temperature slightly lower than the saturation temperature. Due to the formation of the hydrate at temperatures below about 40.6° C., excessive decomposition of the heat sensitive EDT above about 50° C. and the therefore consequent narrow permissible temperature range, the first method has been replaced by the second in commercial production of the crystals. By the above-mentioned methods up to this time, it has been difficult to obtain an EDT crystal with parallel prism faces.

The saturated stoichiometric solution of EDT in water has a pH of about 5.9. Crystallizing from such an acidic normal solution results in a crystal at which the prism faces show approximately a 16 per cent taper. Adding an excess of ethylenediamine to bring the pH up to 7.2 has the effect of reducing crystal taper to about 3 per cent. However, adding an excess of ethylenediamine has the unsatisfactory effect of reducing growth rate and of increasing spurious crystal growth, and furthermore, results in the increase of polymerization of the EDT. Where this type solution is used, it is necessary to stop production when a concentration of polymerized polymer reaches about 8 per cent. The problem then has been to reduce the taper of the EDT crystal while at the same time allowing the crystallization process to proceed at a satisfactory rate.

In the past there have been theories advanced and solutions suggested to avoid this problem, all without success. The process of this invention advantageously improves crystal production in that by its use it is possible to grow a crystal with

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substantially no tapering of the prism faces while at the same time approximately doubling the growth rate and impeding the spurious crystal growth. Also, this process impedes formation of the polymerized EDT and allows the concentration of the polymerized EDT to rise to from 10 to 14 per cent before it becomes necessary to stop the process and clean the tanks, whereas, by methods before in use, the allowable concentration of the polymer was 8 per cent.

In the present invention the EDT crystals are grown from seeds in a saturated EDT mother liquor which liquor pH is maintained at any desired value with the addition of ammonium hydroxide. It has been found advisable to limit the pH range to from 6.5 to 8 since below 6.5 the resultant crystal is so severely tapered as to be commercially impracticable, and since at above 8, the formation of the polymer is a serious problem. Comparative data have been kept on crystal growth from a solution containing ammonium hydroxide in amounts necessary to maintain the pH at about 7.5 as compared with growth from a solution containing an excess of ethylenediamine in amounts necessary to maintain a pH of about 7.2. Sets of data were kept on constant temperature and dropping temperature runs. It was found that whereas the crystal produced from a solution containing an excess of ethylenediamine had prism faces which showed about a 3 per cent taper, crystals grown from the solution containing ammonium hydroxide showed essentially no taper. Whereas the growth rate in a solution containing an excess of ethylenediamine was only about 0.0294 inch per day, growth rates of up to 0.60 inch per day were achieved in a solution containing ammonium hydroxide. Furthermore, it was found practicable to allow the polymer concentration to rise from 10 to 14 per cent in the solution containing ammonium hydroxide, whereas in the solution containing an excess of ethylenediamine, but no ammonium hydroxide, the growth process had come to a substantial halt when the polymer concentration reached 8 per cent.

A detailed description of a dropping temperature process utilizing the present invention follows:

EDT seeds grown by known methods are mounted on a spider and placed in a tank. A saturated solution of EDT in water is made up at about 60° C. and is kept at that temperature. To this solution sufficient ammonium hydroxide is added to bring the pH to about 7.5. This alkaline solution is next poured into the tank which

latter has been preheated to 60° C. so that the tank and contents are now at the saturation temperature. The temperature is next dropped about 0.125° C. and is kept constant for about twelve hours at which time it is again dropped an equal increment. This process is continued for about 80 days until a temperature of 40° C. is reached, it being necessary to stop and drain the solution at any time the polymer concentration rises to from 10 to 14 per cent.

The main disadvantage in the dropping temperature method of growing EDT crystals is the high decomposition rate above 50° C. and the consequent, slow growth rate.

The preferred process utilizing this invention is by growth of the crystals from a supersaturated solution kept at constant temperature. A detailed description of one such process follows:

EDT seeds grown by known methods are mounted on a spider and placed in a tank. A saturated solution of EDT in water is made up at about 43° C. Such a saturated solution contains about 63 weight per cent of EDT. To this solution sufficient ammonium hydroxide is added to bring the pH to about 7.5. This alkaline solution is next run into a superheating tank in which the temperature is kept at about 46° or 47° C. The solution then passes into a crystallizing tank which tank, including a spider and crystals, has been preheated for several hours preceding (12 hours has been found to be ample) at a temperature of about 54° C. The whole crystallizer tank and contents is then kept at a temperature of about 42° C. so as to result in a supersaturation of about 1° C. Excess EDT is added at regular intervals to the saturating tank which EDT passes through the superheating tank and then into the crystallizer. The temperature of the crystallizing tank is kept at the growing temperature, which as above noted, is in the neighborhood of 42° C. This process is allowed to continue for periods of about three months, it being necessary to stop and drain the solution at any time that polymer concentration rises to from 10 to 14 per cent.

A more detailed description of this constant temperature process and of one type of equipment which has proved to be very satisfactory, may be found in United States Patent No. 2,459,869.

Examples have been given of the application of this invention to one dropping temperature process and one constant temperature process. The invention is obviously applicable to any process for the growing of EDT crystals where it is desirable to reduce taper, to increase growth rate and to receive the other benefits outlined above. It is intended to include all methods of growing EDT crystals in the presence of ammonium hydroxide.

What is claimed is:

1. A process comprising growing ethylenediamine tartrate crystals from seeds in a mother liquor supersaturated with ethylenediamine tartrate, which solution is maintained alkaline at a pH of from 6.5 to 8 by the presence of ammonium hydroxide and at a temperature of from about 40.6° C. to 50° C.

2. A process comprising growing ethylenediamine tartrate crystals from seeds in a mother liquor supersaturated with ethylenediamine tartrate, which solution is maintained alkaline at a pH of about 7.5 by the presence of ammonium hydroxide and at a temperature of from about 40.6° C. to 50° C.

3. A process comprising growing ethylenediamine tartrate crystals from seeds in a solution saturated with ethylenediamine tartrate at elevated temperatures by gradually reducing the temperature of the solution from about 50° C. to about 40.6° C. so as to keep it supersaturated at all times, which solution is maintained at a pH of from 6.5 to 8 by the presence of ammonium hydroxide.

4. A process comprising growing ethylenediamine tartrate crystals from seeds in a solution saturated with ethylenediamine tartrate at elevated temperatures by gradually reducing the temperature of the solution from about 50° C. to about 40.6° C. so as to keep it supersaturated at all times, which solution is maintained at a pH of about 7.5 by the presence of ammonium hydroxide.

5. A process comprising growing ethylenediamine tartrate crystals by feeding a solution saturated with ethylenediamine tartrate at one temperature into a tank containing seed crystals kept at a constant temperature in the range of from 40.6° C. to 50° C. which temperature is slightly lower than the saturation temperature and in which the solution is maintained at a pH of from 6.5 to 8 by the presence of ammonium hydroxide.

6. A process comprising growing ethylenediamine tartrate crystals by feeding a solution saturated with ethylenediamine tartrate at one temperature into a tank containing seed crystals kept at a constant temperaure in the range of from 40.6° C. to 50° C. which temperature is slightly lower than the saturation temperature and in which the solution is maintained at a pH of about 7.5 by the presence of ammonium hydroxide.

7. A process comprising growing ethylenediamine tartrate crystals by which a saturated ethylenediamine tartrate solution is made up at a temperature of about 43° C., superheated to 46 or 47° C., and then passed into a crystallizer which crystallizer together with its contents is then kept at a constant temperature of about 42° C. and in which the pH is maintained alkaline at about 7.5 by the addition of ammonium hydroxide, and in which the solution in the crystallizer is kept supersaturated by the addition of ethylenediamine tartrate solution saturated at about 43° C. at regular intervals.

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No references cited.