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PROCESS FOR THE CONTINUOUS PRODUCTION OF CRYSTALLINE SUBSTANCES

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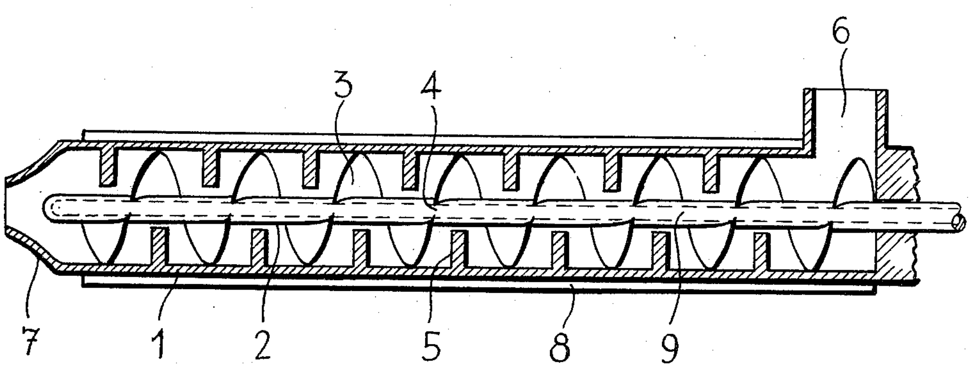

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PROCESS FOR THE CONTINUOUS PRODUCTION OF CRYSTALLINE SUBSTANCES

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The present invention relates to a process for the continuous production of crystalline substances.

For the continuous production of crystalline substances in a powdery or granulated form from crystallizable melts two different methods have been developed, i. e. the atomization in an air current and the solidification on a scraping drum. The atomization in an air current involves the drawback that relatively large rooms are required to enable the atomized drops to pass a sufficiently long way for depositing. The solidification on a scraping drum first yields the desired product in the form of flakes or scales, so that it must further be comminuted by a special operation when a powdery or finely granulated product is desired.

Substances having a low speed of crystallization, for example the hydrates of sodium meta-silicate containing at most 9 mols of water per 1 mol of Na_2SiO_3, can, however, be processed by these two methods only with difficulty, if at all. The atomization carried out, while cooling, does not succeed since the rate of fall of the drops, even when prolonged by suitable measures, is smaller than the time required for crystallization. The second method referred to, i. e. the solidification on a scraping drum may, in principle, be used; however, in view of the necessary long duration of the drops on the cooling surfaces the capacity of such apparatus is small or a disproportionately large apparatus is required to attain the desired effect.

For these reasons the crystallization of such slowly crystallizing substances could, hitherto, not be effected continuously but only by degrees. Thus, for example, the melts have been poured out into crystallizing containers, pans or dishes and allowed to solidify therein, an operation which takes many hours or even days. By overturning the containers, beating and knocking, the solidified melt has then been removed in the form of sheets, blocks or pieces which have then been comminuted to the desired fineness by means of a crusher or a mill. This process involves the drawback of requiring a great expenditure of time and energy and manpower.

It has also been proposed to process such slowly crystallizing melt by placing it onto a continuous belt-conveyor provided with regularly arranged interstices, for example a wire gauze, or a chain conveyor composed of several sections, in a manner such that the melt fills in the said interstices and is removed therefrom after solidification. By this process the product is obtained in the form of pieces (lumps) which must then further be comminuted when the size of the lumps does not satisfactorily meet the industrial requirements as regards the desired fineness.

Now we have found that crystalline material of powdery or granulated form can be obtained continuously from crystallizable melts in a simple manner and by one single operation by kneading the melt with an excess of already crystalline material of the same composition and formed by this process and, simultaneously, moving forward during the kneading process the entire material in

a continuous flow. The heat of crystallization evolved during the operation may be carried off the crystallizing material. It is of advantage to conduct this operation in a manner such that the heat evolved is dissipated to the walls of the apparatus, in which the process is effected. The process of this invention is advantageously conducted in such a manner that the material is mixed only in small zones, while being conveyed and, simultaneously, kneaded. It is likewise advantageous to operate in a manner such that an intimate mixing takes place in every section vertical to the conveying direction. Every constituent of substance passing through the apparatus is mixed, advantageously, only with such constituents of substance as, compared with the duration of the first constituent in the apparatus, have been introduced within a short time before or after the introduction of the first mentioned constituent.

The process of this invention can be used for crystallizing any kinds of crystallizable melts, for instance melts of inorganic or organic substances. The process according to this invention can suitably be used for treating rapidly crystallizing substances and can also be used with special advantage for treating slowly and even very slowly crystallizing substances part of which could hitherto not satisfactorily be processed for industrial purposes.

As substances suitable for being processed by the process of this invention, there may be named, tri-sodium phosphate, p-dichloro-benzene, sodium meta-silicate-9-hydrate, sodium meta-silicate-4-hydrate, sulfur, calcium nitrate, the hydrates of sodium carbonate, or sodium sulfate, or mixtures of washing agents which contain active substances in association with sodium carbonate containing crystal water and/or with sodium sulfate and/or sodium phosphate and/or sodium silicate.

The applicability of the process of this invention is, however, not restricted to the substances mentioned above.

The process of this invention can be conducted using any type of apparatus adapted to the specific purposes intended.

The apparatus shown diagrammatically in the accompanying drawing has proved particularly advantageous. The process of this invention is, however, not limited to the use of such apparatus.

In the accompanying drawing the numerals designate the following parts:

1 is the casing with rotating shaft **2** which simultaneously performs an axial to-and-fro movement (once per revolution). On shaft **2** spirals **3** are arranged which are interrupted by interstices **4**. The wall of casing **1** carries stationary kneading teeth **5** which as regards their size correspond to the said interstices arranged between the sections of the endless screws. By the arrangement of the interstices it is possible that during the motion described of the shaft a screw section passes the right-side of the kneading tooth and the following one passes the left-side of the kneading tooth; by such co-action of kneading teeth and screw-sections a kneading and, simultaneously, a conveying effect is produced. The melt is introduced into the apparatus at inlet **6**, and conveyed by the first spirals and kneaded in the main part of the apparatus with the mixture of melt and the crystals that have already been formed. The ratio of said crystals to the melt increases the more, the more the mixture, while being constantly kneaded, approaches outlet **7**. The heat of crystallization and the frictional heat evolved by the kneading process may be dissipated to cooling agents **8** and **9** through the walls of casing **1** or through the walls of shaft **2** which may be tubular, or the heat may be dissipated by means of the walls of casing **1** and the walls of the shaft **2**. The product leaves the apparatus in a solid powdery or granulated form at the outlet 7 which may be larger or smaller as shown or even be dispensed with.

The process of this invention can be conducted with special advantage using an apparatus with dimensions essentially larger in the conveying direction than in the vertical direction, for example, as large as about 5 to 20 times. The apparatus may have, for example, a casing extended in the conveying direction with a shaft rotating in the longitudinal direction and equipped with kneading elements. The casing may also be equipped with several shafts rotating with the same, a different or a periodically changing speed; the kneading elements, such as endless screws, interrupted screws, wings and teeth may intermesh and clean themselves reciprocally. The rotating movement of the shaft may be combined with to-and-fro motion in the axial direction; the interior of the casing may also be fitted with stationary kneading elements which interact with the movable elements.

The proportion of kneading to conveying effect is advantageously selected in this case in such a way that the kneading effect varies inversely with the crystallization speed of the substance to be crystallized. This may be attained, for example, by changing the form and/or the position of the kneading elements; such regulation, if desired, in association with a regulation of the number of revolutions of the kneading shaft enables to obtain the best output, regardless whether rapidly or slowly crystallizing melts are to be processed. The processing of especially slowly crystallizing substances may be facilitated by leading off the heat of crystallization evolved, at a temperature of maximum crystallization speed, and by taking care to avoid an undercooling below that temperature.

If the melt is passed continuously through an apparatus operating on these principles and otherwise being provided with any kind of equipment, a steady state is reached, i. e. the melt is constantly supplied at the inlet and the same amount of powdery or granulated solid substance constantly leaves the apparatus at the outlet. The proportion of crystalline substance to melt may remain temporarily constant in every section of the apparatus, whereas it varies locally and increases towards the conveying direction. Such increase may extend over the entire length of the apparatus or it may be limited to part of the apparatus.

The process of this invention may, for example, be conducted as follows: Three zones are arranged, i. e. a first zone in which the melt is cooled to the temperature of crystallization, a second zone containing a mixture of melt and crystals in which mixture the content of solid substance locally increases towards the conveying direction from zero to 100 percent and, finally, a third zone which serves to give the crystalline mass the desired final temperature and/or the desired fineness. The first and/or the third zone may, if desired, be dispensed with. At the transition point between first and second zone the crystals which have just been formed are constantly mixed with new melt. As seed crystals they favor the crystallization of the fresh melt introduced without it being necessary to reintroduce solid substance into the apparatus.

If the substance to be processed spontaneously forms a sufficient number of nuclei no crystals need be introduced into the apparatus when putting it into operation; if no nuclei or not enough nuclei are formed a crystalline substance of the same composition alone or in admixture with the melt is once introduced into the apparatus at the beginning of the operation. Thereby the steady state is more rapidly attained at which state agglomerated crystalline material in the end part of the kneader causes the resistance necessary to produce the kneading effect. Generally, it is sufficient to feed about one fourth to about one half of the kneading device with crystalline substance.

The heat exchange between the product within the apparatus and a cooling agent can, advantageously, be effected by dissipating the heat to the walls of the casing. It is, however, also possible to use for this purpose the shaft provided with the kneading element either alone or in combination with the walls of the casing provided said shaft is suitably constructed, for example of tubular construction, enabling the cooling agent to stream through. The cooling agent may be of gaseous nature; air may, for example, be used. It is, however, of advantage to use a liquid cooling agent, for example water, in order to effect a good heat transmission.

For conducting the cooling agent, it is often advisable to use an apparatus equipped with a cooling jacket, it being particularly advantageous if said cooling jacket can be subdivided into single zones so that the substance passing through the apparatus may come into contact, on its way, with cooling walls of different temperature. Thus, it is possible to adapt the heat transmission to the various requirements set forth by subdividing the working zone into two or three zones having different functions. It may thus be desirable more strongly to cool the initial part of the apparatus in order to give the mass on a relatively small distance the best temperature of crystallization. It is, however, also possible more strongly to cool the terminal part of the apparatus in which the final product is given its storage temperature. A reduction of the length of the first and/or the third zone is thus reached, so that the apparatus can be used more advantageously for its specific purpose, i. e. the kneading of the melt and crystals, and its capacity is increased thereby. According to the requirements in each particular case it may also be advisable to dissipate the cooling temperatures in the different zones in another way. For example, at a relatively low temperature of crystallization it may be advantageous to lead off from the inlet of the apparatus only a small amount of heat in order to prevent crystallization and clogging in those parts of the apparatus which serve to introduce the material into the specific working zone. In such cases it may be useful to cause the cooling medium to flow in counter-current to the material.

The following examples serve to illustrate the invention but they are not intended to limit it thereto, Examples 1 and 2 illustrating the process of this invention when applied to rapidly crystallizing substances and Examples 3 and 4 illustrating the process when applied to slowly crystallizing substances.

Example 1

A melt of trisodium phosphate heated to about 80° C. and containing 10.6 mols of water per 1 mol of Na_3PO_4 is introduced continuously into a kneading apparatus at a rate of 4.2 kilograms per hour. Said apparatus has a rotating shaft performing simultaneously periodic axial displacements and on which shaft spirals interrupted by interstices are arranged; on the inner side of the casing kneading teeth are arranged which correspond to said interstices. The terminal part of the casing having an internal diameter of 4.7 cm. and the tubular kneading shaft of 35 cm. length are cooled with cooling water of 20° C., and the inlet of the casing is cooled with cooling water of 50° C. The crystalline product continuously leaves the open outlet of the kneading apparatus in the form of a fine powder.

Example 2

The kneading apparatus described in Example 1 is continuously fed at a rate of 2.4 kilograms per hour with a melt of p-dichlorobenzene heated to 56° C., all parts of the apparatus that can be cooled being cooled with water of 25° C. p-Dichlorobenzene is produced continuously in the form of a fine powder.

Example 3

Into the kneading apparatus described in Example 1 molten sodium-meta-silicate-9-hydrate of 50° C. is introduced continuously at a rate of 4.1 kilograms per hour. The casing and the tubular kneading shaft are cooled with water of 25° C. At the beginning of the kneading operation about 10 grams of crystallized sodium meta-silicate 9-hydrate are once added to the melt; from that time there is only added the melt. At the open outlet of the kneading apparatus crystallized meta-silicate continuously leaves the kneader in a powdery or finely granulated form.

Example 4

Molten sodium-meta-silicate-4-hydrate of about 80° C. is processed continuously at a rate of 2.7 kilograms per hour under the conditions described in Example 1. The temperatures of the cooling water amount at the inlet of the casing to 50° C., at the terminal part and at the tubular kneading shaft to 40° C. At the beginning of the kneading operation about 50 grams of a mixture of equal parts of melt and already crystalline substance are added; from that time there is only added the melt. The product is obtained continuously in a powdery to granulated form.

Example 5

Molten sulfur of 120–130° C. is introduced continuously into the apparatus described in Example 1. The kneading shaft and the part of the casing near the inlet are cooled with water of 50° C., and the terminal part of the casing is cooled with water of 20° C. At 40 revolutions per minute of the kneading shaft the output amounts to 3 kilograms per hour. Sulfur is obtained in the form of a fine powder with a small portion of a gritty constituent. By increasing the number of revolutions to 110

revolutions per minute, the output may be increased to 10 kilograms per hour.

We claim:

1. A process for the continuous production of crystalline material ranging from powdered to granulated form from crystallizable melts, which comprises continuously and intimately kneading the melt with an excess of crystals of said material which have already formed and with the crystals forming, while dissipating the liberated heat of crystallization.

2. A process for the continuous production of crystalline material ranging from powdered to granulated form from crystallizable melts, which comprises continuously and intimately intensively kneading the melt with an excess of crystals of said material, whereby the melt is incorporated into said excess of crystallized material by intimate trituration, while dissipating the liberated heat of crystallization.

3. A process for the continuous production of crystalline material in ground form from crystallizable melts, which comprises continuously and intimately intensively kneading the melt with an excess of crystals of said material at one zone, simultaneously continuously advancing the kneaded mass to a second zone, the proportion of melt decreasing during said advancing until only dry solid material remains, continuously dissipating the liberated heat of crystallization from said zones, and continuously withdrawing the dry ground crystalline material from said second zone.

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