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**Datasheet for the decision
of 6 September 2012**

Case Number: T 0227/10 - 3.3.01
Application Number: 01917675.9
Publication Number: 1273579
IPC: C07D 319/12, B01D 9/02,
B01D 9/00
Language of the proceedings: EN

Title of invention:

Method of refining cyclic ester/KUREHA KAGAKU KOGYO KABUSHIKI
KAISHA

Applicant:

Kureha Corporation

Headword:

-

Relevant legal provisions:

EPC Art. 123(2), 56

Keyword:

"Main and auxiliary requests 1-4 - Inventive step (no) -
obvious alternative"
"Auxiliary request 5 - Inventive step - (yes)"

Decisions cited:

T 0106/84

Catchword:

-



Case Number: T 0227/10 - 3.3.01

D E C I S I O N
of the Technical Board of Appeal 3.3.01
of 6 September 2012

Appellant: Kureha Corporation
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 23 September 2009
refusing European application No. 01917675.9
pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: P. Ranguis
Members: J.-B. Ousset
C.-P. Brandt

Summary of Facts and Submissions

I. An appeal was lodged against the decision of the examining division to refuse the European patent application No. 01 917 675.9 **based on a main request and auxiliary request 1 to 4.**

II. **Claim 1 of the main request read:**

"1. A method of refining crystals of cyclic ester, comprising the steps of:

- supplying a crude cyclic ester as crystalline raw materials to a vertically extending cylindrical refiner (1) from an inlet (3) provided at a lower part of said refiner (1);
- agitating said crude cyclic ester by an agitator (2) provided in said refiner (1) while making said crude cyclic ester flow upwardly;
- forming a downflowing melt of crystal components as refined;
- bringing said upflowing crude cyclic ester and said downflowing melt into counter-flow contact so as to clean off the impurities adhered to the surface of the crystal and sweat out the impurities caught inside the crystal, whereby said crude cyclic ester is refined; and
- taking out, as products, the crystals thus refined from an outlet (4) provided at an upper part of said refiner (1)."

III. The examining division found that the subject-matter of the main request was not inventive in view of document

(6) as closest prior art and in combination, in particular, with document (1)

(1) US-A-3770386

(6) WO-A-96/31506

It held that the teaching of document (6) disclosed a detailed description of purification by melt-crystallization which, furthermore, had a number of particular advantages. The argument that this document established a technical prejudice against purification of cyclic esters by melt crystallization was, therefore, without any merit. The technical problem to be solved in view of document (6) was to provide a suitable device for the purification process by melt-crystallization suggested in document (6). It would have been obvious to solve this problem by using the well-known refining towers for purification of crystalline substances such as disclosed *inter alia* in document (1). The additional features contained in claims 1 of the auxiliary requests could not rebut this finding.

IV. In addition to documents (1) and (6), the following documents are in the appeal proceedings:

(2) GB-A-2064976

(3) EP-A-0446381

(4) EP-A-0789023

(5) EP-A-0242181

(7) US-A-5288881

(8) US-A-2668162

(9) US-A-3457280

(E1) "Temperature effect of cyclic ester degradation"

(E2) US-A-3597449

(E5) Berichte der Deutschen Chemischen Gesellschaft,
Vol I: Pages 745-752, "Über die Verseifung von
Glykolid und Lactid in saurer Lösung, H. Johansson
und H. Sebelius (1919),

(E6) Polymer Applications, Vol. 30, N.5 (1981),
pages 6-15 and translation into English of
pages 14-15.

(E7) "Melting point depression of glycolide".

V. The requests before the Board are the main request and auxiliary request 1-4 filed with letter dated 6 August 2012 and auxiliary request 5 filed during the oral proceedings which took place on 6 September 2012. For the sake of good order, the appellant was required to file again at the end of oral proceedings the complete sets of claims of the main request and auxiliary requests 1 to 4 (attached to the minutes).

Claim 1 of the main request and auxiliary request 1, apart a minor editorial change, namely, first line, "a crystal" instead "crystals" have the same wording as claim 1 of the main request before the Examining Division (see point II above).

Claim 1 of auxiliary request 2 differs from claim 1 of the main request in that the sentence "A method of refining a crystal of cyclic ester, comprising the steps of:" was replaced with "A method of refining a crystal of cyclic ester from a condensation product of a dicarboxylic acid and diol, or product made from hydroxycarboxylic acid dimer which was made circular by dehydrate-condensation, said method comprising the steps of:"

Claim 1 of auxiliary request 3 differs claim 1 of the main request in that "cyclic ester" was replaced with "glycolide".

Claim 1 of auxiliary request 4 differs from claim 1 of the main request in that the sentence "A method of refining a crystal of cyclic ester, comprising the steps of:" was replaced with "A method of refining a crystal of cyclic ester from a condensation product of a dicarboxylic acid and diol, or product made from hydroxycarboxylic acid dimer which was made circular by dehydrate-condensation, wherein the crystal of a crude cyclic ester is adhered by at least one high-boiling point organic solvent having a boiling point greater than the melting point of said crystal, said method comprising the steps of:"

Claim 1 of the fifth auxiliary request reads:

"1. A method of refining a crystal of cyclic ester produced by depolymerising hydroxycarboxylic acid oligomer, wherein no solvent is adhered to the crystal or a high-boiling point organic solvent is adhered to the crystal, said method **consisting** (emphasis added by the Board) of the steps of:

- supplying a crude cyclic ester as a crystalline raw materials to a vertically extending cylindrical refiner (1) from an inlet (3) provided at a lower part of said refiner (1);
- agitating said crude cyclic ester by an agitator (2) provided in said refiner (1) to make said crude cyclic ester flow upwardly;

- forming a downflowing melt of a crystal component as refined;
- bringing said upflowing crude cyclic ester and said downflowing melt into counter-flow contact, whereby said crude cyclic ester is refined; and
- taking out, as products, the crystals thus refined from an outlet (4) provided at an upper part of said refiner (1)."

VI. The board pointed out in its communication annexed to the summons to oral proceedings that in view of document (6) as closest prior art, the person skilled in the art would have considered document (1), when trying to find an alternative method to purify easily degradable cyclic esters.

VII. The arguments of the appellant submitted in the written proceedings and during oral proceedings can be summarized as follows:

The cyclic esters are easily degradable depending on time and temperature as confirmed by documents (E1) , (E2), (6) bridging paragraph pages 2-3 . Furthermore, document (6) points out the difficulty to transfer the processes for purifying cyclic ester compositions, *inter alia* melt-crystallization, in controlled laboratory conditions to a commercial process because they are complex or have impractical operating parameters and the cyclic esters may degrade resulting in poor purity. Document (6) proposes a method of purification, wherein the feed material including cyclic esters is subjected to an adsorption step in which impurities are adsorbed from the feed material. A prejudice existed, therefore, against applying a

continuously refining tower to easily degradable cyclic esters. It is true that some further recovery processes including among other purification by melt-crystallization were proposed. This melt-crystallization was however merely cited as one of alternative methods in the course of **post-adsorption** treatments. Furthermore, the claimed refining method and the melt crystallization method of document (6) are based on a different refining principle. The method of document (6) must be a batch method, which is less efficient than a continuous method. In addition, none of the disclosures appear to suggest performing the melt crystallization in a counter-flow contact.

Therefore, the problem to be solved in view of document (6) can be seen in the provision of a simpler and industrially better adapted method for purifying easily degradable cyclic esters. The solution was to suppress the main purifying step on an adsorbent, and adapting the melt crystallization step destined for post-treatment in document (6) to a single-step continuously refining method, carried out in a vertically extending cylindrical refiner on a counter-current basis. It was surprising that the use of downflowing melt in a refining tower in a gravitational manner with a tight control of temperature and exposed time could lead to a purification of these unstable cyclic esters.

Documents (1), (2), (3) or (5) related to refining **stable** substances (p-dichlorobenzene, p-xylene and naphthalene) in an upright tower apparatus and designing an optimal tower structure. Thus, though the tower apparatuses were known at least since 1973, document (1), there was no teaching to use them for

refining easily degradable cyclic esters. By contrast, the preferred purification method in the prior-art was re-crystallization. Many documents (not in the proceedings, note of the Board) published in 1968, 1969, 1988 and 1994 illustrated this method. Finally, in 1997, document (4) addressed the problem of refining the dimeric cyclic esters that tend to undergo ring opening polymerization. The proposed solution consisted of distilling the cyclic esters in the presence of a high-boiling polar organic solvent. Document (4) confirmed the general trend not to use the refining tower method but crystallization methods, or a distilling method. The development in the past led away from the use of a refining tower for unstable cyclic esters. T 106/84 was cited to show that a sharp change of direction should be considered as involving an inventive step.

Regarding auxiliary request 3 limited to glycolide, document (6) discloses a method of purification only for tetramethyl glycolide and lactide. Not for glycolide. Tetramethyl glycolide and lactide are more hydrophobic and more bulky than glycolide as confirmed by documents (E5) and (E6, translation). Therefore, it was not obvious to employ the claimed method for the purification of glycolide which is hydrolysed more easily than tetramethyl glycolide and lactide.

Regarding auxiliary request 4, as stated in documents (1) to (3), the impurities removed from the refining towers have usually a lower melting point than that of the crystals to be refined. It would have been thought impossible to remove, from easily degradable cyclic esters, a solvent having a boiling point higher than

the melting point of the crystals of such cyclic esters, when a continuously refining tower is used.

VIII. The appellant requested that the decision under appeal be set aside and that a patent be granted either on the basis of the main request or on the basis of one of auxiliary requests 1 to 4 all filed with the appellant's letter dated 6 August 2012 or on the basis of auxiliary request 5 as filed during oral proceedings on 6 September 2012.

IX. At the end of the oral proceedings, the decision of the board was announced.

Reasons for the Decision

1. The appeal is admissible.
2. Valid text of the application

This text corresponds to the application as originally filed (pages 1 to 4, 6, 7, 10, drawings 1/2 and 2/2) and the amended version filed with letter dated 6 August 2009 (pages 5, 8, 8a, 9, 9a, 11 and 11a).

Main and auxiliary requests 1 and 2

3. Inventive step
 - 3.1 Document (6) discloses a process to purify cyclic esters. Therefore, the Board concurs with the examining division that this document aims at the same objective as the claimed invention and can be considered as the

closest state of the art to define the technical problem to be solved. The appellant although contending that document (6) might not even be an appropriate prior art with regard to the claimed invention, did not submit any other prior art instead thereof.

This document states first that several processes have been proposed in controlled laboratory conditions for purifying cyclic ester compositions, including melt crystallization. Those processes suffer many inconvenience so that they cannot be operated on a commercial scale. It was also pointed out that the cyclic esters being purified could degrade (see page 2, line 14 to page 3, line 2). As a solution it was proposed a purification method wherein the feed material including cyclic esters is subjected to an adsorption step (see claim 1). After this adsorbing step, the cyclic esters can be further purified by melt crystallisation as purifying process. The melt crystallisation has various advantages over solvent crystallisation processes and there are several configurations of equipment for melt crystallization. Those familiar with the art will be able to pick an appropriate configuration for a given application (see page 20, first and second full paragraphs). Cyclic esters include those derived from lactic acid and glycolic acid (see page 7, line 16) and those derived from hydroxycarboxylic acids (see page 6, lines 17 to 18).

- 3.2 Claim 1 properly construed by the examining division does not exclude due to the wording "comprising" a previous initial step for purifying the crystalline material to be treated according to the claimed process.

As already noted by the examining division, claim 3 requires a high-boiling solvent adhered to the crystals, which is typical for a preceding purification step according to document (4). Furthermore, the claimed process is not limited to any scale and this alleged additional feature cannot be considered.

- 3.3 In the absence of evidence showing a different or better technical effect provided by the claimed invention vis-à-vis document (6), the problem underlying the present application can be regarded in the provision of a further continuous process to purify crude cyclic esters, namely to remove mother liquid impurities adhered to the crystal and to those taken inside it.
- 3.4 In view of the example and the general description it is considered that the problem was actually solved.
- 3.5 It remains to be decided whether or not the claimed solution is obvious in view of the state of the art.
- 3.5.1 Any argument related to the judicious control in the present invention of temperature and time necessary to maintain the cyclic esters in a state with no heat degradation is without any merit since the claim 1 does not recite those conditions which are, in addition not disclosed in the application. Therefore, document (E8, E1) which represents the thermal decomposition of cyclic esters depending on heat and time has no bearing with the claimed solution.
- 3.5.2 The person skilled in the art seeking to find a further process to solve the problem underlying the present

would know from document (6) that melt crystallisation is particularly appropriate for cyclic esters as a further purification step, after the adsorption step. Moreover, he would also know from this document that several configurations of equipment can be used to perform the said purification by-melt crystallization (see point 3.1 above). Unless a prejudice would have prevented him, he would have considered the equipment known in the prior art.

3.5.3 As properly noted by the examining division melt-crystallization, although after the adsorption step, is envisaged in document (6) and even said to have "a number of particular advantages". The alleged "technical prejudice" is, therefore, without any merit. Furthermore, that document (4) relates to a process involving distilling out the dimeric cyclic ester together with a high-boiling polar organic solvent cannot confirm a general trend teaching away from using melt crystallization as further purification step.

3.5.4 The alleged sharp change of direction relied upon, by reference to T 106/84, cannot be acknowledged because contrary to the former case cited, where the PCT resistors in the claimed packing machine functioned as stringent control means, technical effect not known from the previous uses, in the present case the melt-crystallisation as a post-purification step of cyclic esters was known from document (6).

3.5.5 Hence, the person skilled, having no prejudice against the use of melt-crystallization as further purification step in the context of the teaching of document (6), would have looked at document (1), which describes a

process to purify **continuously** a crystalline substance (see column 1, lines 12-13). It is true that the material teaching of document (1) is confined to stable substances (e.g. p-dichlorobenzene). However, the instability of cyclic esters being not an obstacle to melt-crystallization (see document (6)), a device such as disclosed in document (1) would have been *prima facie* appropriate for implementing the purification by melt-crystallization as taught in document (6). Document (1) describes the supply of the crude crystal from the charging mouth **6** (lower part of the tower) into the purification tower by means of a screw conveyor **7**. The raw materials charged into the tower are purified while being transported to the upper portion of the tower by means of the stirrer **5**. The heating and melting means **1** positioned in the upper portion of the tower is maintained at the temperature of the melting point of the crystal component, or above whereby the purified crystal is melted to a liquid. A portion of the liquid descends back down through the tower and contacts the raw material crystal rising in the tower. One portion of the purified material reaching the upper portion is removed by means of products outlet **2** and the remaining liquid portion is returned to the inside of the tower (see column 5, lines 21 to 57 and Fig. 1). The person skilled in the art applying the teaching of document (6), namely adsorption step then melt-crystallization, in combination document (1) would have designed without inventive ingenuity a process falling within the definition of claim 1 and for this reason the subject-matter of claim 1 of the **main request and first auxiliary request** does not involve an inventive step. Documents (2), (3) and (5) disclose other crystal

refining apparatuses and cannot rebut but only reinforce, if needed, this finding.

- 3.5.6 For the same reason the second auxiliary request must fail since the added feature, "cyclic ester from a condensation product of a dicarboxylic acid and diol, or product made from hydroxycarboxylic acid dimer which was made circular by dehydrate-condensation" is disclosed in document (6) (see point 3.1 above).

Auxiliary request 3

4. Inventive step

- 4.1 Even though the Board would admit that glycolide is more easily hydrolysable than lactide or tetramethyl glycolide as stated by the appellant in view of the fact that the two latter are more hydrophobic and more bulky than glycolide, i.e. documents (E5), (E6), it remains that document (6) discloses a method for purifying cyclic esters of formula X_2A which is a linear dimer molecule of an X_1A molecule wherein X_1A can be glycolic acid. Therefore X_2A corresponds to glycolide (see pages 6, line 16 to page 7, line 23). For the same reasons set out above (see point 3 above, main request) claim 1 of the third auxiliary request does not involve an inventive step.

- 4.2 Documents (7), (8) and (9) were submitted by the appellant to support that claim 1 of this request does not infringe Article 123(2) EPC. This is however not an issue in the present decision.

Auxiliary request 4

5. Inventive step

5.1 High boiling polar organic solvents used together in the distillation of cyclic ester have a boiling point within the range of 230-450°C as taught by document (4). It was noted during oral proceedings that diethylene glycol dibenzoate (see document (4), page 8, example 2) had a melting point of 24°C. That is lower than the melting point of glycolide.

5.2 The person skilled would have applied without inventive ingenuity the teaching of document (1) which requires that the impurity has a lower melting point than that of the crystalline substance. For this reason the subject-matter of claim 1 of the auxiliary 4 does not involve an inventive step and since the Board can only decide on a request as a whole, auxiliary request 4 is to be rejected.

6. Auxiliary request 5

6.1 The expression "produced by depolymerising hydroxycarboxylic acid oligomer" is based on page 8, lines 8 to 9. The expression "wherein no solvent is adhered to the crystal" is based on page 8, line 7. The expression "a high-boiling point organic solvent is adhered to the crystal" is based on page 8, line 25 to 26. In view of the example on page 9, lines 21 to 25, the person skilled in the art would directly and unambiguously derive that the claimed process consists only in the steps described in claim 1.

The requirements of Article 123(2) are fulfilled.

6.2 Inventive step.

6.2.1 Document (6) remains the closest state of the art and the technical problem to be solved can also be seen in the provision of a further continuous process to purify crude cyclic esters, namely to remove mother liquid impurities adhered to the crystal and to those taken inside.

6.2.2 Document (6) teaches that crude cyclic esters are purified by necessarily, first, subjecting the feed material including cyclic esters to an adsorbing step, then to a purification step which may be a melt-crystallization. Document (4) teaches a different method, namely crystallization or distilling method. None of the prior art cited invites the person skilled in the art to use as **sole** step a melt-crystallization involving the continuous melt-crystallization processes disclosed in documents (1), (2), (3) or (5) to solve the above defined problem. The subject-matter of Claim 1 of the auxiliary request 5 involves an inventive step. The same applies to sub-claims 2 to 5.

7. Remittal

The case is remitted to the department of first instance for adaptation of the description to the subject-matter of auxiliary request 5 and incorporation of the relevant prior art according to Rule 42(1)b).

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the department of the first instance with the order to grant a patent on the basis of auxiliary request 5 (claims 1-5) as filed during oral proceedings on 6 September 2012 and after any necessary consequential amendment of the description.

The Registrar

The Chairman

M. Schalow

P. Ranguis