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**Datasheet for the decision  
of 8 February 2013**

**Case Number:** T 2039/09 - 3.3.05

**Application Number:** 06011301.6

**Publication Number:** 1741678

**IPC:** C02F 3/10, C12N 11/04

**Language of the proceedings:** EN

**Title of invention:**

Process and apparatus for producing entrapping immobilization pellets

**Applicant:**

Hitachi Plant Technologies, Ltd.

**Headword:**

Entrapping pellets/HITACHI

**Relevant legal provisions:**

EPC Art. 123(2), 84, 52(1), 54(1)(2), 56

**Keyword:**

"Inventive step (yes) "

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 2039/09 - 3.3.05

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.05  
of 8 February 2013

**Appellant:**  
(Applicant)

Hitachi Plant Technologies, Ltd.  
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Tokyo (JP)

**Representative:**

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**Decision under appeal:**

Decision of the Examining Division of the  
European Patent Office posted 25 May 2009  
refusing European patent application  
No. 06011301.6 pursuant to Article 97(2) EPC.

**Composition of the Board:**

**Chairman:** G. Raths  
**Members:** B. Czech  
C. Vallet

## Summary of Facts and Submissions

- I. The appeal is from the decision of the examining division refusing European patent application No. 06011301.6.
- II. The prior art documents cited in the course of the substantive examination of the case include:
- D3: JP 10-235384 A
- D6: T. Sumino et al.: "Immobilization of Nitrifying Bacteria by Polyethylene Glycol Prepolymer"; Journal of Fermentation and Bioengineering, Vol. 73, No. 1, 1992, pages 37 to 42; and
- D7: EP 1 304 200 A1.
- III. The examining division found that the process according claim 1 of the sole request (of 14 April 2009) then on file was not inventive in view of the disclosure of document D6, taken as closest prior art, considering that cubic pellets were known in the art from e.g. document D3.

Said claim 1 has the following wording (features added to claim 1 as filed highlighted by the board):

*"1. A process for producing entrapping immobilization pellets (70) in which microorganisms are entrapped and immobilized in an immobilizing agent, the process comprising:  
polymerizing a mixture containing the microorganisms and a solution of the immobilizing agent in a forming*

*frame (52) into a gel to prepare a pellet block (60) further comprising: cutting the pellet block (60) into lattices while fixing a periphery of the pellet block; and then cutting the lattice-shaped pellet block substantially into cubes to provide pellets (70)."*

IV. In its statement of grounds of appeal, the appellant maintained the request refused by the examining division. Under cover of said statement, the appellant also filed two sets of amended claims as auxiliary requests. It essentially argued that document D6 neither represented the closest prior art nor suggested a process as claimed.

V. The appellant was summoned to oral proceedings in accordance with an auxiliary request to this end. In a communication issued in preparation of the oral proceedings, the board raised objections under Articles 123(2) and 84 EPC. The board also questioned the meaning of some of the terms used in claim 1 and considered that when construed in the context of the description, claim 1 appeared to lack novelty over the following document, cited in the application in suit:

D8: JP 2003-235553 A.

Furthermore, the board questioned the presence of an inventive step in view of a combination of documents D8 and D7.

VI. Under cover of its response filed on 8 January 2013, the appellant filed twelve sets of amended claims as a new main request and auxiliary requests 1 to 11. It

considered that said new claims overcame all pending objections under Articles 123(2), 84, 54 and 56 EPC.

VII. Oral proceedings were held on 8 February 2013. In response to concerns expressed by the board with respect to the requests on file, regarding in particular their compliance with Articles 123(2) and 84 EPC as well as the issues of novelty and inventive step, the appellant submitted a single set of amended claims 1 to 5 as its sole and main request.

Claim 1 according to said new main request reads as follows (features added to claim 1 as filed highlighted by the board):

*"1. A process for producing entrapping immobilization pellets (70) in which microorganisms are entrapped and immobilized in an immobilizing agent, the process comprising:  
polymerizing a mixture containing the microorganisms and a solution of the immobilizing agent in a forming frame (52) into a gel to prepare a pellet block (60)  
**further comprising:  
cutting the pellet block (60) into a lattice-shaped pellet block while fixing a periphery of the pellet block; and then cutting the lattice-shaped pellet block substantially into cubes to provide pellets (70),  
wherein as the immobilizing agent a polyethylene glycol dimethacrylate prepolymer is used, having a molecular weight of 4000 to 12000 g/mol, at a concentration of 3 to 10 mass%,  
wherein the pellet block (60) has a deformation ratio of 70% or more  
wherein the pellet deformation ratio is represented by***

*the following Formula 1*

*(Formula 1): pellet deformation ratio (%) =  
(H0 -H1)/H0x100,*

*wherein H0: initial pellet thickness before compression,  
wherein H1: pellet thickness immediately before  
breakdown of the pellet gel, and wherein the pellet  
deformation ratio is determined using a rheometer."*

The dependent claims 2 to 5 according to the new main request relate to specific embodiments of the process of claim 1.

- VIII. Having regard to this request, the appellant essentially argued that the amended claims met the requirements of Articles 84 and 123(2) EPC, and that the claimed subject-matter was novel and inventive in view of the prior art including documents D8, D7 and D6. More particularly, none of the documents on file disclosed or suggested the preparation of entrapping immobilisation pellets by a process with all the specific features of present claim 1 at issue.
- IX. The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of claims 1 to 5 according to the main request filed at the oral proceedings.

## **Reasons for the Decision**

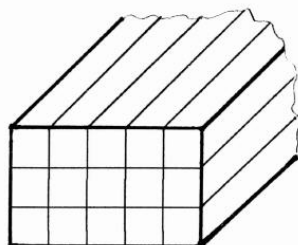
1. Admissibility of the appellant's request
  - 1.1 The proposed amendments to the claims can be regarded as straightforward attempts to deal with objections

raised by the board in its communication and/or at the oral proceedings.

1.2 The board, in the exercise of the discretion conferred on it by Article 13(1)(3) RPBA, therefore decided to admit the appellant's request to the proceedings despite its late filing.

2. Clarity of the amended claims (Article 84 EPC)

2.1 Amended claim 1 at issue refers to "*cutting the pellet block (60) into a lattice-shaped pellet block*", which "*lattice-shaped block*" is subsequently cut "*into cubes*". In contrast to the wording of the respective claims 1 of the request previously on file, the present wording clearly expresses that the block is first cut into a two dimensional array of parallel stripes or columns of substantially square cross section, said array being subsequently cut into cubic pellets in the last cutting step. The skilled person understands without doubt that in the context of claim 1 the expression "*lattice-shaped pellet block*" designates such an array of stripes or columns present before the final cutting step, which can be schematically represented in the following manner:



An example of a "*lattice-shaped pellet block*" is also shown in Figure 3A of the application in suit as it emerges from cutting tool 66B.

2.2 In response to a corresponding objection by the board, the appellant amended claim 1 by including indications concerning the determination of the claimed "*deformation ratio*". From claim 1 as amended, the skilled person gathers that the determination of said "*deformation ratio*" is done "*using a rheometer*" and involves measuring the pellet thickness and relating it to the thickness of a pellet compressed to a point immediately before its breakdown. Therefore the board accepts that in the context of claim 1 in its present wording, the expression "*deformation ratio of 70% or more*" of the "*pellet **block** (60)*" refers to the "*deformation ratio*" value of the material as measured on pellets cut from the block.

2.3 In the board's judgement, the amended claims at issue are thus not objectionable under Article 84 EPC.

3. Allowability of the amendments (Article 123(2) EPC)

3.1 Claim 1 at issue stems from a combination of the features of claims 1, 2 ("*deformation ratio*" addressed) and 6 (refers to "*cutting into a lattice-shaped pellet block*"). The erroneous reference to lattices (plural) as contained in claim 6 of the application as filed (and in the first full paragraph on page 16 thereof) was not taken up into claim 1 at issue.

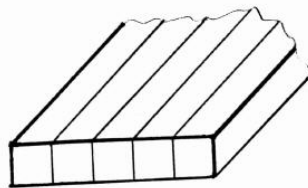
3.2 The indications concerning the determination of the "*deformation ratio*" and the preference for deformation ratio values of "*70 % or more*" are disclosed on page 4, penultimate paragraph, page 5, lines 12 to 13, and on page 20, lines 3 to 9, respectively. In the examples contained in the application as filed, "*polyethylene*



*glycol dimethacrylate*" was used as the immobilisation agent, see page 17, table 1. From figures 7 and 8, page 5, second paragraph, and the two last paragraphs on page 20, of the application as filed, it can be derived that when this particular immobilisation agent is used, a prepolymer concentration in the range of "3 to 10 mass%" and a prepolymer having a molecular weight in the range of "4,000 to 12,000" permit achieving the required deformation ration of 70 % or more.

4. Novelty

Document D8 neither discloses the specific prepolymers nor the molecular weight, concentration and deformation ratio ranges recited in claim 1 at issue. Moreover, D8 does not disclose a "*lattice-shaped pellet block*" within the meaning of claim 1 at issue (see point 2.1. above) as an intermediate product in the pellet preparation process. D8 merely describes a sheet of gelled material cut into parallel stripes, i.e. having an appearance that can schematically be illustrated in the following manner (compare to point 2.1 above):



This schematic drawing shows that the sheet cut into stripes can be regarded as a "*block*" in the broadest sense of the term, but differs from the "*lattice-shaped block*" obtained as an intermediate product in the process of claim 1 at issue. Whereas the cut sheet shown above comprises a single layer of parallel stripes or columns, the "***lattice-shaped block***" referred

to in claim 1 comprises several adjacent layers of parallel stripes or columns, as shown in the example schematically represented under point 2.1 above.

In this connection, the board observes that the acknowledgement of D8 in the application as filed (page 2, second full paragraph) is thus erroneous insofar as it refers to such a sheet of material cut into parallel stripes as "*lattices*" and as a "*lattice-shaped pellet block*" (see application as filed, page 2, lines 18 and 19).

4.1 The board is also satisfied that none of the other prior art documents cited discloses a process with all the features of present claim 1.

4.2 The subject-matter of claim 1 and, consequently, of claims 2 to 5 dependent thereon, is thus novel (Articles 52(1) and 54(1)(2) EPC).

5. Inventive step

5.1 The invention relates to a process for producing pellets comprising entrapped and immobilised microorganisms.

5.2 The closest prior art is undisputedly represented by the disclosure of document D8 which is cited in the description of the application in suit (page 2, second full paragraph and Figure 11).

5.2.1 As acknowledged in the application in suit, the process described in D8 (see e.g. figures 1 and 4) comprises forming a sheet from a mixture of an unspecified

polymeric material and microorganisms. In paragraph [0016] of D8 (see the machine translation available from the JPO website) it is indicated that the raw material mixture is cast onto a belt conveyor 26 and gels, thereby immobilising the microorganisms within the gelled sheet. The sheet is formed to its final thickness between the lower belt conveyor 26 and an upper belt conveyor 28. Paragraphs [0017] to [0022] of D8 describe how the formed sheet, moving along on backing plate 30, is then cut by rotary blades 36 into endless stripes parallel to the transport direction. Finally, said stripes of material are cut into cubic pellets (see translation of claim 3) by cutting unit 38 comprising blades extending in a direction perpendicular to the transport direction.

5.2.2 According to D8 (translation of paragraphs [0011] and [0013], this process is suitable for the mass production of entrapping immobilisation pellets of cubic shape at high speeds. Pellets of "excellent form accuracy" (see PAJP abstract) are obtained.

5.3 According to the application as filed (page 3, second full paragraph), the technical problem was to provide a process for the inexpensive and high-speed mass-production of entrapping immobilisation pellets which are highly stable.

More particularly, the process according to the invention is stated to solve problems associated with the process described in D8 (see application as filed, page 3, lines 3 to 29). The use of a "*forming frame*" is stated to reduce the contact of the polymerising mixture with air and to lead, therefore, to a better

pellet strength and to less variance in pellet quality than the sheet-forming process of D8. Moreover, in the process according to D8, some material which is not polymerised is stated to remain on the sheet surface, which material is discharged as COD when the pellets are used for waste water treatment. The performance of the pellets in waste water treatment is thus negatively affected.

5.4 As a solution to this problem the application now proposes the process according to claim 1 at issue, which is characterised in particular in that it comprises (emphasis added by the board) "*polymerizing a mixture containing the microorganisms and a solution of the immobilizing agent in a forming frame (52) into a gel to prepare a pellet block (60)*" and then "*cutting the pellet block (60) into a **lattice-shaped pellet block** while fixing a periphery of the pellet block; and then **cutting the lattice-shaped pellet block** substantially **into cubes** to provide pellets (70), wherein as the immobilizing agent a **polyethylene glycol dimethacrylate prepolymer** is used, having a **molecular weight of 4000 to 12000 g/mol**, at a **concentration of 3 to 10 mass%**, wherein the pellet block (60) has a **deformation ratio of 70% or more**".*

5.5 Concerning the success of the solution, and more particularly the improved "stability" and/or "quality" of the pellets, allegedly achieved in an inexpensive manner, compared to the pellets obtained by the process of document D8, the board observes the following.

5.5.1 It can readily be gathered from the application as filed (see e.g. page 3, last two paragraphs; page 4, lines 14 to 17; page 14, lines 5, 6, 12 and 22 to 24; page 19, third paragraph) that said improvements may be achieved provided the "*forming frame*" used meets certain criteria as to its construction, shape and/or volume-to-surface ratio, in order to reduce the contact area of the polymerising mixture with ambient air.

5.5.2 Claim 1 at issue is, however, silent about the constructional features, shape and volume of the "*forming frame*" to be used.

As indicated in the board's communication, the sheet-forming upper and lower conveyors belts 26 and 28 shown in document D8 can also be considered as a "*forming frame*" and the sheet obtained can be considered as a "*block*" in the broadest sense of these terms.

5.5.3 Consequently, the board does not accept that the process according to claim 1 at issue will generally lead to the stated improvements over the process described in D8.

5.6 Therefore, the technical problem to be solved starting from the disclosure of D8 must be reformulated in a less ambitious manner. It can be seen in providing a further process for the high-speed mass-production of cubic entrapping immobilisation pellets.

5.7 The board accepts that this problem is indeed successfully solved by the process of claim 1. Irrespective of the precise shape of the forming-frame used, the process claimed obviously lends itself to a

high-speed mass-production of pellets, since a high number of pellets can simultaneously be obtained in the final step of cutting a "*lattice-shaped block*" within the meaning of claim 1 (see point 2.1 above) into cubes. Moreover, the experimental results reported in the application corroborate that when the specific materials and process conditions recited in claim 1 are used, deformation ratios of 70 % or more may be achieved, such optimised deformation ratios permitting a very fast removal of the gel block from the forming-frame (see Figure 6 and page 20, first full paragraph, of the application as filed).

- 5.8 Hence, it remains to be assessed whether or not the claimed process is obvious in the light of the prior art.
- 5.8.1 D8, taken alone, does not suggest cutting a gelled block of material into a "*lattice-shaped block*" in the sense of claim 1 at issue (see point 2.1 above), let alone using a prepolymer solution as specifically defined in claim 1 at issue and ascertaining that a deformation ratio of 70 % or more is achieved.
- 5.8.2 Document D7 (see page 3, paragraph [0005]) discloses a process for cutting blocks of polymeric, preferably aqueous gels into smaller particles of defined, preferably cubic, shape. Various applications of such gel particles are mentioned, including in "waste water treatment" in general (page 2, line 11), but not specifically as pellets with entrapped immobilised micro-organisms.

More particularly, the mechanical process features of claim 1 are all disclosed in document D7 (see e.g. claims 1 and 11 and Figure 1): The polymeric gel block is first fed to a static grid- or lattice-shaped ("gitterförmig") cutting element whilst being kept dimensionally stable via a frame ("Lade 2" in Figure 1 and claim 11) which is "*fixing a periphery*" of the block in the sense of claim 1. The resulting three-dimensional block of parallel strands of gel each having a square cross-section, which is undisputedly a "*lattice-shaped*" block in the sense of claim 1, is subsequently cut into a plurality of essentially equal, cubic shape.

Concerning the chemical aspects of the process according to claim 1 at issue, document D7 is almost silent and, therefore, of little relevance: It is merely indicated in D7 that the disclosed processing method is suitable for various kinds of polymeric gels, including aqueous methacrylic (co-)polymer gels. It can also be assumed that the deformation ratio of the polymer blocks prepared is such that the cutting operation can actually be successfully performed. However, applying the cutting process described to a gel block obtained by polymerising, specifically, a solution of a "**polyethylene glycol dimethacrylate prepolymer**" is neither mentioned nor suggested in D7, let alone to a gel having a "**deformation ratio of 70 % or more**" which is obtained from a solution of such a prepolymer "*having a molecular weight in the range of from 4000 to 12000 g/mol*" in a concentration of from "**3 to 10 mass%**".

5.8.3 Document D6 relates to the preparation of entrapping immobilisation pellets. More specifically, nitrifying bacteria for water purification purposes are entrapped in a gel obtained by radical polymerisation of polyethylene glycol ("PEG" hereinafter) prepolymers. The suitability of several different PEG prepolymers was investigated, taking into account *inter alia* the deformation ratio (defined as in the application suit) of the gel pellets. (see D6: page 38, left-hand column, section "Deformation rate").

According to D6 (page 37, right-hand column, last full paragraph), the pellets are obtained by extruding the polymer gel entrapping the microorganisms from a PVC tube and cutting the extruded gel "into columnar pellets of Ø2 x 2, Ø3 x 3, or Ø8x5 mm".

From these indications, the board understands that the gel is extruded in form of a single strand of circular cross section, the pellets only being obtained one at a time by cutting across the extruded strand of material. As far as the transformation of the gel material into pellets is concerned, document D6 thus suggests, if at all, to simultaneously extrude several strands of polymerised material and to cut these into pellets in a single cutting operation. D6 thus teaches away from a method as disclosed in D8, wherein a shaped mass in form of a sheet of material is transformed into pellets by subsequent cutting operations performed on said sheet and which can be assumed to require different considerations when it comes to the design of the cutting tools and the required gel properties.



Moreover, concerning the chemical aspects of the teaching of document D6, the board observes the following:

- i) All the pre-polymers investigated in D6, including the dimethacrylates, have molecular weights far outside of the claimed range of "4000 to 12000", the highest numerical value reported being **1684** (see D6: Table 1 on page 38);
- ii) The PEG prepolymer concentration used in the experiments reported in D6 (see page 37, line 4 from the bottom, was **18 %** (w/v), i.e. substantially higher than the "3 to 10 mass%" prescribed by claim 1 at issue; and
- iii) last but not least, none of the pellets described in D6 (see Tables 2 and 3 on page 39) has a deformation ratio of "70 % or more" as required by claim 1 at issue, the highest reported value being **63 %** and, in the case of gels made from PEG dimethacrylate prepolymers, **49.1 %** (D6: Table 2, sample G2).

From the above, the board concludes that the skilled person starting from a process as disclosed in D8 and being confronted with the stated technical problem, was not prompted by the disclosure of document D6 to solve said problem by preparing a block of gel material with a deformation ratio of 70 % using the raw materials and process conditions defined in claim 1, and to cut this block into a "lattice-shaped" block first, and then into cubes.

- 5.8.4 Moreover, the board considers that without the benefit of hindsight, the skilled person would not envisage modifying the process of document D8 by adopting the gel forming and cutting steps described in D7 and, at

the same time, using raw materials and chemical gel forming conditions differing radically from those disclosed as in document D6, in order to solve the stated technical problem.

5.8.5 Document D3 discloses the preparation of cubic carrier particles for the immobilisation of microorganisms (see figures 1 a-c and the PAJP abstract of D3) by cutting up a block of melt-bonded polypropylene fibres. However, these cubic particles are not of the "entrapping" type formed of polymeric gel. Instead, they are intended to carry microorganisms attached only to the surface of the bonded fibres making up the cubic particle. Document D3 is thus less relevant than both D6 and D7.

5.8.6 The board is also satisfied that none of the other prior art documents cited in the search report contains further relevant information that could render the subject-matter of claim 1 obvious.

5.9 The board concludes that the subject-matter of claim 1 and, consequently, of claims 2 to 5 dependent thereon, involves an inventive step (Articles 52(1) and 56 EPC).

6. Hence, the appellant's (main) request is allowable.

**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 first instance with the order to grant a patent on the basis of claims 1 to 5 according to the sole request filed at the oral proceedings and a description, including figures 1 to 11, to be adapted where appropriate.

The Registrar

The Chairman

C. Vodz

G. Rath