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**D E C I S I O N**  
**of 12 March 1997**

**Case Number:** T 0165/94 - 3.3.3

**Application Number:** 86906156.4

**Publication Number:** 0248030

**IPC:** C08K 5/00

**Language of the proceedings:** EN

**Title of invention:**  
Polyethylene composites containing a phase change material

**Applicant:**  
UNIVERSITY OF DAYTON

**Opponent:**  
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**Headword:**  
-

**Relevant legal provisions:**  
EPC Art. 56

**Keyword:**  
"Inventive step (yes) - non obvious alternative"

**Decisions cited:**  
-

**Catchword:**  
-



Case Number: T 0165/94 - 3.3.3

DECISION  
of the Technical Board of Appeal 3.3.3  
of 12 March 1997

**Appellant:** UNIVERSITY OF DAYTON  
300 College Park Avenue  
Dayton  
Ohio 45469 (US)

**Representative:** Warren, Anthony Robert  
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**Decision under appeal:** Decision of the Examining Division of the  
European Patent Office posted 3 November 1993  
refusing European patent application  
No. 86 906 156.4 pursuant to Article 97(1) EPC.

**Composition of the Board:**

**Chairman:** C. Gérardin  
**Members:** P. Kitzmantel  
J. A. Stephens-Ofner

## Summary of Facts and Submissions

I. This appeal, which was filed on 18 December 1993, lies against the decision of the Examining Division dated 3 November 1993, refusing European patent application No. 86 906 156.4 in the name of University of Dayton filed on 23 September 1986 as International patent application No. PCT/US86/01998, claiming the priority of 22 November 1985 from a US-application, and published under the No. WO 87/03290 (= EP-A-248 030). The appeal fee was paid on 21 December 1993 and the Statement of Grounds of Appeal was filed on 21 February 1994.

II. The decision under appeal was based on a set of 22 claims, Claims 2 to 19 having been filed with a letter dated 14 October 1992 and Claims 1 and 20 to 22 having been filed with a letter dated 7 July 1993. Independent Claims 1, 17 and 22 read as follows:

"1. A composite thermal energy storage material comprising a polyethylene matrix and, absorbed in and contained within the matrix, at least 10 wt.% of a crystalline straight-chain alkyl hydrocarbon phase change material having at least 14 carbon atoms in the molecule and a latent heat of fusion greater than 30 cal/g, characterised in that the polyethylene matrix is crosslinked to impart thereto a gel content of 10 to 90%."

"17. A thermal energy storage material comprising a non-polymeric cementitious matrix having dispersed therein pellets of a composite material according to any one of claims 13 to 16."

"22. The use of a composite material according to claim 21 as a wall or floor covering."

Claims 2 to 16 and Claim 21 related to preferred embodiments of the composite material according to Claim 1 and Claims 18 to 20 related to preferred embodiments of the thermal energy storage material according to Claim 17.

III. The decision under appeal held that the subject-matter of Claim 1 of the application in suit was novel but did not involve an inventive step over the disclosure of

- (1) JP-A-59 170 180 (considered in the form of Derwent Abstract No. 84-278399/45),

because it was considered obvious to a person skilled in the art to crosslink the polyethylene matrix of pellets and sheets which have been prepared from the polyethylene compositions specified in that document, if one wished to impart form stability to said pellets and sheets. Otherwise these compositions were identical to those according to the application in suit; in particular they comprised as phase change materials (hereinafter "PCM"s) the same low molecular weight polyethylene waxes which were used according to Claim 1. This conclusion was arrived at because the Applicant had failed to provide evidence for the alleged improvement of the materials according to Claim 1 over those disclosed in D1 with respect to the characteristics: wax content, stability towards thermocycling and heat efficiency on repeated use.

The subject matter of Claim 17 was also considered non-inventive, because concrete compositions comprising encapsulated PCMs have been known from document

- (2) US-A-4 504 402,

and because a technical effect resulting from the use of a PCM in the form of pellets instead of in encapsulated form was not shown.

IV. With his Statement of Grounds of Appeal filed on 21 February 1994 the Appellant submitted a Main Request and an Auxiliary Request. In response to a suggestion of the Rapporteur in the communication of 22 August 1996 to amend Claim 1 of the Main Request, the Appellant, with his letter dated 21 October 1996, submitted an amended version of this claim, which, after minor editorial amendment (see minutes of telephone conversation of 7 March 1997, communication of 20 March 1997 and letter of confirmation by the Appellant dated 24 March 1997), has been replaced by the following final version of this Claim 1:

"1. A composite thermal energy storage material comprising polyethylene and at least 10 wt.% of a crystalline straight-chain alkyl hydrocarbon phase change material having at least 14 carbon atoms in the molecule and a latent heat of fusion greater than 30 cal/g, characterised in that the polyethylene is crosslinked to form a matrix having a gel content of 10 to 90% and in that said phase change material is absorbed in the matrix by its impregnation into the matrix."

Except for the deletion of Claims 21 and 22, which have been dropped by the Appellant in reaction to the aforementioned communication of the Rapporteur, and except for a minor amendment of Claim 5, the further claims of the Main Request are identical to those of the appealed decision.

Claim 1 of the Auxiliary Request, which comprises 22 claims, relates to a method of making a composite thermal energy storage material.

V. Together with his Statement of Grounds of the Appeal the Appellant submitted an English translation of document (1) and "Extracts A and B". In response to the Rapporteur's communication of 22 August 1996 he submitted "Annexes 1 to 3" and US-A-5 053 446:

- "Extract A": pages 2, 5 to 10, 16 to 19, Figures 1, 2, 6, 7, 8 (a) to 8 (g), 16a and 16b, from an undated test report of the Appellant;
- "Extract B": pages 23 to 25 from another undated test report of the Appellant;
- "Annexe 1": reports of tests (thermocycling evaluation and DSC (differential scanning calorimeter) data) carried out in laboratories of the Appellant in the years 1986 and 1987 (37 pages);
- "Annexe 2": reports of tests (DSC data) carried out in laboratories of the Appellant in the year 1986 (83 pages);
- "Annexe 3": reports of tests (weight gain) carried out in laboratories of the Appellant in the year 1986 (4 pages).

The Appellant essentially argued that the composites according to Claim 1 of the application in suit were distinguished from those according to document (1) not only by the polyethylene matrix being crosslinked but also by the incorporation of the PCM into the matrix material by absorption. By that a composite structure was established which could not result from the methods

of preparation disclosed in document (1), according to which the PCM was homogeneously mixed with the uncrosslinked polyethylene and subsequently melt-kneaded and extruded or moulded into the required shape.

Moreover, the thermal energy storage materials according to Claim 1 of the application in suit which contained a high proportion of PCM and remained form stable over a broad temperature range and through repeated melt/freeze cycles were not suggested by the homogeneous mixtures disclosed in document (1), because there the form stability was obtained by the use of an **uncrosslinked** polyethylene of very high molecular weight and there was no reason for the person skilled in the art to change to a **crosslinked** polyethylene matrix which could **not** be subjected to the mixing, melt-kneading and extrusion steps performed according to document (1).

Furthermore, document (1) did not suggest that the crosslinked matrix material could absorb large quantities of up to 80% of hydrocarbon PCM.

Concerning the newly filed evidence the Appellant contended the following:

- "Extract A" illustrated the form stability of pellets made from the compositions according to Claim 1 of the application in suit, and "Extract B" showed how the degree of cross-linking was related to the gel content;

- the data contained in "Annexe 1" with respect to samples 7 to 12 showed that once formed, the crosslinked polyethylene matrix material retained the hydrocarbon PCM material through many repeated cycles of melting and freezing (up to 1000 cycles between temperatures of -10 to 60°C).
- "Annexes 2 and 3" and Example 8 of US-A-5 053 446 contained information relevant to the issue of the interdependence of the degree of crosslinking and the capacity of the matrix material to absorb hydrocarbon PCM.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the Main Request or the Auxiliary Request.

In the event that neither of the two requests would be considered allowable by the Board, the Appellant requested that oral proceeding be held.

### Reasons for the Decision

1. The appeal is admissible.

#### *Main Request*

2. *Amendments (Article 123 (2) EPC)*

Claim 1 is essentially based on original Claim 1. The added characteristics are based on the following passages of the original application:

- the features that the straight-chain alkyl hydrocarbon PCM is crystalline, has at least 14 carbon atoms in the molecule and has a latent heat of fusion greater than 30 cal/g on the first paragraph of page 5,
- the lower and upper limits of 10% and 90% gel content of the crosslinked polyethylene matrix on the first paragraph of page 8, and
- the fact that the PCM is absorbed in the matrix by its impregnation into the matrix on the passage from page 12, line 28 to page 13, second paragraph.

Claims 2, 3, 5, 8, 10 and 13 to 20 are based, in this sequence, on original Claims 7, 4, 10, 11, 13, 3, 5, 6, 8, 16, 22, 27 and a combination of original Claims 28 to 31.

Claims 4, 6, 7, 9, 11 and 12 are based, in this sequence, on the following passages in the original application:

- page 14, lines 4 to 6,
- page 14, lines 20 to 23,
- page 15, lines 8 to 11,
- page 16, lines 21 to 25,
- page 14, line 28 to page 15, line 11, and
- page 15, lines 12 to 20.

The requirement of Article 123(2) EPC is therefore complied with by all claims.

3. *Novelty (Article 54 EPC)*

- 3.1 Document (1), now considered in the form of an English translation of the JP-A-59-170180, relates to thermal energy storage materials similar to those according to the application in suit.

In particular, it discloses compositions which comprise, homogeneously dispersed, 5 to 60 parts by weight of an ultra-high molecular weight polyethylene of an intrinsic viscosity  $[\eta]$  of at least 5 dl/g and 95 to 40 parts by weight of a polyethylene wax of a molecular weight of from 300 to 10 000. These compositions may be prepared by homogeneous mixing of the two components, followed by melt blending and shaping by extrusion, injection moulding or compression moulding (see "Scope of Claim" and page 5, first paragraph).

According to the only Examples 1, 2 and 3 ultra-high molecular weight polyethylene of  $[\eta] = 17$  dl/g (20, 40 and 20 parts by weight, respectively) and paraffin wax (molecular weights of 900, 5000 and 700, respectively; melting points 109°C, 134°C and 93°C, respectively) were mixed in a Henschel mixer and the so prepared blends extruded to strands which were then subjected to thermal tests.

The strands are reported (i) to have an outstanding shape stability when fused and (ii) to be able to retain the polyethylene wax over repeated melt/freeze cycles (see last paragraph of page 1, second paragraph of page 3, second paragraph of page 5 and examples).

3.2 The subject-matter of present Claim 1 is distinguished from that disclosed in (1) by

- (i) the use of crosslinked polyethylene and
- (ii) the presence of the hydrocarbon PCM within the polyethylene matrix in absorbed, not in melt-mixed form.

3.2.1 While the distinguishing character of the aforementioned feature (i) was recognized in the appealed decision, that of feature (ii) was denied. In particular it was held that the materials disclosed in document (1), irrespective of their preparation by homogeneous mixing, melt-kneading and extrusion to pellets or sheets, must, in view of the reference in the Derwent abstract of document (1) to the melting of the wax, have a composite structure. The Board agrees to this conclusion of the Examining Division because document (1) does not disclose that the heat storage materials disclosed therein are homogeneous in the sense that they consist of a single phase.

3.2.2 However, the absorption of a polyethylene wax into a crosslinked polyethylene matrix, must, in order to take up the wax, lead to an expansion of the structure of the matrix material that is different from the structure of a wax-containing uncrosslinked polyethylene, because in the latter case the constraint caused by the crosslinking is lacking.

3.2.3 The Board concurs with the finding in the appealed decision, which was not contested by the Appellant, that the feature of present Claim 1 that the hydrocarbon PCM has a latent heat of fusion greater than 30 cal/g, though not literally disclosed in (1), does not distinguish the waxes used according to (1) from the PCMs employed according to the application in

suit. This finding of the Examining Division was based on the obvious conclusion that a polyethylene wax having a molecular weight of 300 (this being the lower molecular weight limit for the waxes used according to (1)), which corresponds to a straight chain alkyl hydrocarbon having the formula  $C_{21}H_{44}$  (molecular weight 296) having a melting point of about 40°C, could not be different with respect to its latent heat of fusion from the "commercially available waxes" having similar melting points, which are exemplified as useful PCMs on page 13, last paragraph of the application in suit.

3.2.4 In view of the conclusion reached in point 3.2.2 above, the subject-matter of Claim 1 of the application in suit is thus novel over document (1). The same conclusion applies to the subject-matter of the further independent Claim 17 which relates to a material comprising pellets of the composite material according to Claim 1.

4. *Inventive step (Article 56 EPC)*

4.1 Problem and solution

4.1.1 According to the application in suit the problem to be solved by the alleged invention was the provision of a thermal energy storage material that has a good thermal form stability and a high capacity for the PCM. Thermal form stability is defined in the application in suit as the resistance of pellets of the material against sticking to one another at temperatures up to about 150°C (page 6, line 7 to page 7, line 13).

A further important property of the material, which is not separately mentioned in the original application, but which is self-evident in view of the intended use as heat storage material in the building sector, is

the maintenance of the material's favourable properties during use, which requires maximal retention of the PCM in the matrix (low oozing out) through many repeated cycles of melting and freezing (see page 2, last paragraph of the Statement of Reasons of Appeal).

- 4.1.2 In view of the fact that no comparative evidence is available on the basis of which it could be concluded that the materials according to present Claim 1 are superior, with respect to the above properties, to those disclosed in document (1), which represents the closest state of the art, the technical problem underlying the application in suit can only be seen in the provision of an **alternative** thermal energy storage material having a good form stability, a good resistance against oozing out of the PCM and the matrix material of which has a high capacity for the PCM.
  
- 4.1.3 According to Claim 1 of the application in suit the afore-mentioned aspects of the technical problem are to be solved by a thermal energy storage material comprising a matrix of crosslinked polyethylene having a gel content of 10 to 90%, having absorbed therein a crystalline straight-chain alkyl hydrocarbon PCM having at least 14 carbon atoms in the molecule and a latent heat of fusion greater than 30 cal/g.
  
- 4.1.4 In view of the considerations which follow, it can be concluded that the technical problem set out in point 4.1.2 has been effectively solved in all its aspects by the subject-matter of present Claim 1, i.e. that its subject-matter defines an alternative thermal

energy storage material to that disclosed in document (1) having, under the conditions of repeated melt/freeze cycles, a good form stability, a good capability to retain the PCM and a high capacity for PCM.

- 4.1.4.1 Pellets made from the material according to Claim 1 of the application in suit exhibit, in terms of resistance against sticking, a reasonable form stability, and are also resistant to oozing out of the wax, as can be inferred from the results of samples No. 7 to 12 reported under the heading "Initial Thermocycling Samples" on sheet 9 of "Annexe 1". All six tested samples show after 100 melt/freeze cycles good sticking and oozing results (no sticking at all or slight sticking; no oozing or some oozing). On average these two properties gradually deteriorate when the number of thermocycles is enhanced to 200, 500 and 800, but remain relatively good even at 1000 thermocycles. This can be inferred from the fact that at this high test rate the sticking results still range from "no change" (sample No. 7) over "slight sticking" (sample No. 12) to "sticking" (sample No. 8) and the oozing results range from "no change" (sample No. 7) over "very little oozing" (sample No. 11) to "some oozing" (samples No. 9 and 10).

Pellets from materials according to Claim 1 of the application in suit (for the compositions of samples No. 7 to 12 see sheets 1 to 3, especially Table 1 on page 2 of "Annexe 1" and the relevant explanations thereto in the paragraph bridging pages 1 and 2 of Appellant's submission dated 21 October 1996) thus meet the desired standards of form stability and PCM retention capacity. This part of the existing technical problem is therefore solved.

4.1.4.2 According to Claim 1 of the application in suit the materials shall comprise at least 10 wt.-% of PCM. Preferably, pellets of the material should absorb 40 to 80 wt.-% of the PCM (page 13, lines 15 to 17 of the application in suit).

In "Annexe 3" weight gain data are reported for materials comprised of crosslinked HDPE (Marlex<sup>(R)</sup> 6006) which was impregnated with "Witco 45-A", a wax exemplified as useful PCM on page 13, line 29 of the original application. On sheet No. 2 of "Annexe 3" an average weight gain of 88,26% is indicated for 10 batches.

It can thus be accepted that the crosslinked polyethylene matrix used according to Claim 1 of the application in suit has a high capacity for hydrocarbon PCMs as defined in this claim. This part of the existing technical problem is therefore also solved.

#### 4.2 Obviousness

This issue turns on the question whether document (1), alone or in combination with further state of the art, was able to suggest to the person skilled in the art the alternative thermal energy storage materials specified in Claim 1 of the application in suit.

4.2.1 Owing to the use of ultra-high molecular weight polyethylenes the materials disclosed in document (1) have an "outstanding shape-stability when fused" (page 3, second paragraph). It follows from the indication of a minimum intrinsic viscosity of 5 dl/g necessary for shape retention (page 3, last paragraph, second sentence) and even more from the warning that crosslinking impaired the properties of polyolefins as

heat storage materials (page 3, first paragraph) that the way to achieve form stability according to document (1) is by enhancing the molecular weight and **not** by crosslinking.

There is no information in document (1) which could induce the person skilled in the art to use crosslinked (lower molecular weight) polyethylene instead of uncrosslinked ultra-high molecular weight polyethylene.

This conclusion applies not only to "ordinary" thermal form stability, i.e. shape retention on heating, but also to the special definition of form stability used according to the application in suit, which defines this property in terms of the resistance of pellets of the material against sticking to one another (cf. point 4.1.1 supra; Example 2 of document (1)).

Neither does document (1) contain any hint at the incorporation of the hydrocarbon PCM, instead of melt mixing with, by impregnation into the (crosslinked) polyethylene matrix.

Since there is no further state of the art in the proceedings which contained any suggestion to the solution of the existing problem (cf. point 4.1.2 supra), the Board holds that this solution, as specified in Claim 1 of the application in suit, was non-obvious.

- 4.2.2 The conclusion drawn in the penultimate paragraph of point 5 of the Reasons of the appealed decision, with reference to the "problem" stated in the first paragraph of the same point, namely that, in view of common general knowledge (reference was made to document (3): Encyclopaedia of Polymer Science and

Engineering, Vol. 4, "Cross-linking", John Wiley & Sons), it was obvious that the problem "to impart form stability to the pellets and sheets, so that they do not deform or stick to one another" could be solved by the use of a crosslinked polymer, is wrong.

Firstly, this definition of the "problem" is inconsistent with the fact that the materials disclosed in the closest prior art, i.e. document (1), **are** form stable, with the consequence that this "problem" did not exist. Secondly, even if, for the sake of argument, one accepted that in view of common general knowledge, with respect to form stability, ultra-high molecular weight polyethylene and crosslinked (lower molecular weight) polyethylene would have been obvious **alternatives**, the skilled person would be left without any information as to how the PCM should be incorporated into such a matrix material. As rightly set out by the Appellant on page 3, paragraph 4 of the Statement of Reasons of the Appeal, homogeneous mixing followed by melt-kneading and extrusion could not be carried out with a thermally form-stable crosslinked matrix material. Thus, putting on a par, with respect to form stability, ultra-high molecular weight polyethylene and crosslinked (lower molecular weight) polyethylene is a hindsight argument not supported by either (1) or (3). Thirdly, there is not the slightest hint in document (1) at the incorporation of the hydrocarbon PCM into the polyethylene matrix by impregnation.

- 4.2.3 Furthermore, it is without consequence for the recognition of the non-obviousness of the subject-matter of Claim 1 of the application in suit (cf. point 4.2.1 supra) that there is no evidence for an **improvement**, with respect to the materials disclosed in document (1), of the form stability, the capability

of the material for retention of the PCM and the capacity of the matrix to hold substantial quantities of PCM. Once the choice of a mere **alternative solution** is non-obvious, there is no need for any improvement.

- 4.3 It follows that the subject-matter of Claim 1 of the application in suit involves an inventive step.
- 4.4 Owing to the fact that the compositions according to Claim 17 comprise the thermal energy storage materials specified in Claim 1, this subject-matter must also be inventive.
- 4.5 The inventivity of the subject-matter of Claims 2 to 16 and 18 to 20 follows from their dependency upon the respective independent Claims 1 and 17.
- 4.6 Therefore, the subject-matter covered by the set of claims of the Main Request complies with the requirements of Article 56 EPC.
5. Since the claims of the Main Request are also in agreement with the other requirements of the EPC, particularly with Articles 84 and 123(2), this request is allowable.
6. There is, thus, no need to deal with the Auxiliary Request, nor is there any need to hold oral proceedings.
7. Since the description of the application is not yet in agreement with the claims of the Main Request, the Board decides to make use of its powers under Article 111 EPC and remits the case to the Examining Division for this purpose.

## Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division with the order to grant a patent on the basis of the following claims:
  - Claim 1 as set out in the Rapporteur's communication of 20 March 1997 (cf. Section IV supra) and confirmed by the appellant on 1 April 1997
  - Claims 2 to 4 and 6 to 20 as decided upon in the appealed decision,
  - Claim 5 as submitted with the Statement of Grounds of Appeal filed on 21 February 1994

and a description yet to be adapted.

The Registrar:



E. Görgmaier

The Chairman:



C. Gérardin

