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DECISION
of 14 May 1997

Case Number: T 0501/93 - 3.3.3

Application Number: 88907030.6

Publication Number: 0324842

IPC: C08L 81/02

Language of the proceedings: EN

Title of invention:
Conductive plastic composites

Patentee:
NESTE OY

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56, 84

Keyword:
"Novelty (yes); after amendment"
"Inventive step (yes)"
"Clarity (yes) - natural and ordinary meaning"

Decisions cited:
-

Catchword:
-

Case Number: T 0501/93 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 14 May 1997

Appellant:	NESTE OY SF-06850 Kulloo (FI)
Representative:	Browne, Robin Forsythe, Dr. Urquhart-Dykes & Lord Tower House Merrion Way Leeds LS2 8PA West Yorkshire (GB)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office dated 5 January 1993 refusing European patent application No. 88 907 030.6 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: R. J. Gérardin
Members: J. Young
J. A. Stephens-Ofner

Summary of Facts and Submissions

I. The appeal lies from a decision of the Examining Division, dated 5 January 1993, to refuse European patent application No. 88 907 030.6 (publication No. 0 324 842), based on International application No. PCT/FI88/00122, having an International Filing Date of 29 July 1988 and an International Publication Date of 9 February 1989 (International Publication No. WO 89/01015), and claiming a priority date of 29 July 1987 (FI 873308). The Notice of Appeal was received on 26 February 1993, the fee being paid on 8 March 1993.

II. The decision under appeal was based on a set of Claims 1 to 18 filed on 2 April 1992, Claim 1 of which read as follows:

> "A conductive material which consists of a polymer matrix or a polymer substrate, an internally conductive, doped polymer, and optionally plastics additives, characterized in that the internally conductive, doped polymer is a solidified melt of a poly(3-alkyl thiophene).

Claims 2 to 10 were dependent claims directed to elaborations of the material according to Claim 1.

Claim 11, an independent claim, read as follows:

> "A process for the preparation of a conductive material, wherein a polymer matrix or polymer substrate is brought together with a conductive polymer and optionally with plastics additives and the conductive polymer is doped at some stage, characterized in that

the poly(3-alkyl thiophene) is shaped in molten state together with the polymer matrix on the polymer substrate to produce a finished material.".

Claims 12 to 18 were dependent claims directed to elaborations of the process of Claim 11.

According to the decision, whilst Claim 1 did not fulfil the requirements of Article 123(2) EPC in respect of a minor point of its wording, the sole ground of refusal relied upon was lack of novelty of the claimed subject-matter in view of:

D1: WO-A-87/00677; and
D2: WO-A-85/05728.

In particular, the product defined by Claim 1, a product-by-process claim, was a composite of an electrically conductive polythiophene or substituted polythiophene and a polymer substrate. The allegation that it was a homogeneous material not obtainable by the method of D1 was (a) not relevant, since the structure of the composition was not a feature of Claim 1, (b) not consistent with the wording of Claim 1, which covered heterogeneous compositions, and (c) not convincing, because the allegation was unsupported by any evidence. Consequently, the subject-matter of Claim 1 lacked novelty in the light of D1.

Furthermore, the disclosure of D2 related to blends of substituted polythiophene and a polymer matrix or substrate which were able to be shaped thermoplastically, and "thermoplastic" had been admitted to mean shapable in molten state. The allegation that D2 related to copolymers consisting of chemically bound blocks A and B did not establish a distinction, firstly because the claims did not exclude

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chemical bonding between the "polymer matrix" and the "doped polymer", and secondly because D2 also referred specifically to "blends", i.e. mixtures of two or more polymers which could be shaped thermoplastically. Consequently, the subject-matter of Claims 1 and 11 lacked novelty over the disclosure of D2.

The decision furthermore pointed out that the subject-matter of the dependent claims related to features which were also disclosed in D1 and/or D2.

III. The Appellant filed, together with the Statement of Grounds of Appeal on 11 May 1993, a new, restricted set of Claims 1 to 8 and argued that the method now claimed addressed the problem of providing an easily melt processable polymer which was also electrically conductive. Combination of the electrically conductive polymer with another polymer component was easier if the conductive polymer was in the molten state and more complete than would be the case if the electrically conductive polymer was in the form of a non-fusible powder or if it was polymerised in situ. Prior to this, however, it had been believed that electrically conductive polymers lacked thermoplastic properties.

In support of the latter submission, reference was made to D2, and also to three further documents, which, although published after the priority date of the application in suit and thus not forming state of the art in the sense of Article 54 EPC, contained numerous references to previous studies about processability and conductivity of poly(3-alkylthiophenes):

A: *Österholm, J.E. et al.*, "Melt and Solution Processable Poly(3-alkylthiophenes) and their Blends", Synthetic Metals, *28* (1989) C435 to C444;

B: *Gustafsson, G.*, "Processable Conducting Poly(3-alkylthiophenes", *J.L. Brédas and R. Silbey (eds.)* "Conjugated Polymers", Kluwer Academic Publishers, 1991, 315 to 362; and

C: *Leclerc, M. et al.*, "Structural analysis of poly(3-alkylthiophene)s", Makromol. Chem. 190, 1989, 3105 to 3116.

IV. Following the issue, on 2 July 1996, of a communication by the Board, in which attention was directed *inter alia* to the need to establish a distinction over D2 as regards the nature of the alkyl polythiophene, the Appellant filed, on 29 August 1996, two further revised sets of Claims 1 to 8 and two sets of revised pages 3, 3a of description, forming a main and auxiliary request respectively. A minor amendment of Claim 1 and the description on page 3 was requested in a subsequent letter dated 29 April 1997, received on 2 May 1997. Claim 1 of the main request, including this amendment, reads as follows:

"A process for the preparation of an electrically conductive polymer composite, wherein a first polymer is brought together with a second electrically conductive polymer and the polymer composite is doped, characterised in that the second electrically conductive polymer is a melt processable poly(3-alkyl thiophene) and that the first polymer and the second electrically conductive polymer are brought together by shaping and/or processing the second electrically conductive polymer, that is the melt processable poly(3-alkyl thiophene), in its molten state."

Claims 2 to 8 are dependent claims directed to elaborations of the process according to Claim 1.

V. The Appellant requested that a patent be granted on the basis of pages 1, 2, 4 to 8 as originally filed and pages 3 and 3a filed with the main request of the letter of 29 August 1996 together with Claims 1 to 8 also filed on that date, with the amendments indicated in the letter dated 29 April 1997 or, failing this, on the basis of the auxiliary request, filed on 29 August 1996.

Reasons for the Decision

1. The appeal is admissible.

2. *Admissibility of amendments; main request*

Claims 1 to 8 correspond generally to Claims 11 to 18 underlying the decision under appeal. No objection under Article 123(2) EPC was raised in the decision under appeal to any of the latter claims, and none is evident to the Board.

2.1 Claim 1 in its present version differs from that of Claim 11 referred to above, apart from the deletion of an optional feature, in that:

(a) the term "conductive material" has been replaced by "an electrically conductive polymer composite";

(b) the "polymer matrix or substrate" has been generalised to "a first polymer", with the conductive poly (3-alkyl thiophene) polymer being termed "a second electrically conductive polymer";

(c) the definition of the latter has been restricted to "melt processable poly(3-alkyl thiophene)" species;

(d) the shaping and/or processing step now requires only the second, electrically conductive polymer to be in molten state as the two polymers are brought together;

(e) the phrase "the conductive polymer is doped at some stage" has been amended to read "the composite is doped"; and

(f) the word "characterised" has been inserted at line 4 of the claim.

2.1.1 Amendment (a) is supported by Claim 11 as originally filed, which referred to the preparation of "a conductive polymer composite", read in conjunction with the introductory description, which refers, in the context of internally conductive polymers, to "electric current" (page 1, second paragraph). Thus, it is clear that "conductive" was originally intended to mean "electrically conductive".

2.1.2 Amendment (b) is an allowable generalisation from the alternatives presented, since it is clear from the description on page 3, last paragraph, and page 4, first two paragraphs, that the only common feature is that of a polymer.

2.1.3 Amendment (c) is supported by Claims 1, 2, 8, 11 and 13, and the description on page 3, penultimate paragraph, which make it clear that the "poly (3-substituted thiophene)" is applied in molten state, read in conjunction with the description at the top of page 4, which refers to an advantageous composite in which the active component is "poly(3-alkyl thiophene)".

2.1.4 Amendment (d) is supported by the embodiment generally referred to on page 2, first complete paragraph and more particularly described in Example 11, in which the second electrically conductive polymer is applied in molten form to a substrate. It is thus clear that it is not necessary that both polymers are processed in molten state, provided that, when they are brought together, at least the second electrically conductive polymer is so shaped and/or processed.

2.1.5 Amendment (e) is supported by the description on page 4, third complete paragraph.

2.1.6 Amendment (f) is supported by original Claim 1.

2.2 Claims 2 to 8 correspond to Claims 12 to 18 respectively underlying the decision under appeal, except that the amended terminology in Claim 1 has been adopted consistently and the appendancies have been changed to reflect the renumbering adopted.

2.3 The amendments to the description merely correspond to those made in Claim 1, as well as an acknowledgment of the contents of D1 and D2.

2.4 Thus, no objection arises under Article 123(2) EPC.

3. *Clarity of Claim 1*

Objection had been raised by the Examining Division that the word "composite" in Claim 1 was confusing, since the term "composite" was normally used for laminated materials, which were not believed to be part of the subject-matter of the application (communication dated 11 April 1991, page 1, last paragraph).

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The Board cannot concur with this view. The dictionary definition of "composite" is "made up of various parts or elements" (The Shorter Oxford English Dictionary", Volume I, Oxford University Press, 1973). Furthermore, one embodiment of the description evidently involves a lamination step (section 2.1.4, above). The term "composite" according to such a definition would, in this connection, clearly include a laminate. There is thus no reason to depart from the natural and ordinary meaning of the word "composite" in the present case, as defined in a standard dictionary. In summary, the term "composite" in Claim 1 is clear.

4. *The closest state of the art; the technical problem*

The application in suit is concerned with the preparation of electrically conductive polymer composites having good mechanical processing characteristics, in which one of the components of the composite is an internally electrically conductive polymer (page 2, third paragraph). Such composites are, however, known from D2, which is considered to be the closest state of the art.

4.1 According to D2, a disadvantage of polymers with a conjugated π-system, which become electrically conductive after doping (i.e. oxidation or reduction), is that they adopt a structure of rigid microcrystalline domains which detracts from all the mechanical and processing qualities normally associated with synthetic polymers. An intrinsically conductive polymer of this type can neither be melted nor dissolved (D2, page 1, lines 12 to 31).

4.2 Intrinsically conductive polymers which have, as far as possible, good processing characteristics are, according to D2, copolymers or blends essentially composed of a polymer component (A) not having a π-system, and a polymer component (B) having a conjugated π-system, the polymer component (A) being provided with redox active groups capable of oxidising or reducing component (B) (page 2, lines 16 to 27; Claim 1).

4.3 The polymer component (A) may be mixed, in known manner, with the polymer component (B) or with monomers which yield this component, and then chemically or electrochemically oxidised or reduced (Claim 13).

4.4 The conductive polymer (B) may be a substituted or unsubstituted polythiophene (page 3, lines 16 to 17; Claim 8).

4.5 The polymer component (A) may, for instance, be a reaction product of ferrocene with polyvinyl chloride, polybutadiene, polyacrylate, or polymethacrylate, or with a copolymer of maleic acid with styrene, or of butadiene with styrene or with poly (chloromethylated styrene) (page 4, line 32 to page 5, line 9; Claim 16).

4.6 Compared with this state of the art, the technical problem can be seen as the provision of an improved electrically conductive polymer composite having superior mechanical working properties and a wider range of applicability.

4.7 The solution proposed according to Claim 1 of the application in suit is to select, as the conductive polymer component (B), a melt processable poly(3-alkyl thiophene), whereby the choice of polymer component (A) is no longer limited to polymers having active redox groups.

4.8 It is self-evident that incorporation of an electrically conductive polymer component is easier if the conductive polymer is in the molten state, and more complete than would be the case if the electrically conductive polymer were in the form, say, of a non-fusible solid (section 4.1, above). This is corroborated by document C, according to which "Only those conducting polymers which are able to combine good electrical and mechanical properties together with processability from solution or melt... have chances to be used toward the production of electronic or optical devices." (paragraph bridging pages 3105, 3106).

4.9 It is clear from the examples of the application in suit, that poly(3-octyl thiophene) can be moulded in a molten state to form a conductive polymer composite based on a typical non-conductive polymer, such as polyethylene or polyethylene terephthalate, not containing redox groups (Examples 6, 11; pages 7, 8).

4.10 Although the only poly(3-alkyl thiophene) exemplified is poly(3-octyl thiophene), it is clear from the evidence of documents A and B that the relevant effect is not limited to poly(3-alkyl thiophene)s in which the alkyl group is octyl.

4.10.1 In particular, according to document A, poly(3-alkylthiophenes) having long, flexible hydrocarbon chains in the 3-position of the thiophene ring yield poly(3-alkylthiophenes) which are solution and melt processable (page C436, lines 7 to 10).

4.10.2 Furthermore, according to document B, "Poly(3-alkylthiophenes, with alkyl side chains longer, than butyl, are both solution and melt processable" (page 327, section 4.1, "Processability").

4.10.3 Consequently, it is evident that substitution of the thiophene with any suitably long alkyl group at the 3-position, such as, for instance, an octyl group, will result in an effective level of melt processability in the conductive polymer.

4.10.4 Whilst the precise length of the alkyl chain is not the subject of a specific limitation in Claim 1, the combination with the functional requirement of the species being "melt processable" fulfils, in the Board's view, the same purpose.

4.10.5 In this connection, it is not considered that the skilled person would be presented with an intolerable burden in establishing that such species in which the alkyl chain is too short are not melt processable, and therefore do not fall within the terms of the proposed solution of the technical problem. Indeed, the publication, shortly after the application in suit, of the relevant details in documents A and B corroborates this (sections 4.10.1, 4.10.2, above).

4.11 Furthermore, it is clear from the general disclosure and examples of the application in suit that any conventionally compatible non-conductive polymer can be mixed with the melt processable component, and in particular that the presence of active redox groups is not necessary.

4.12 In summary, it is credible that the claimed measures provide an effective solution of the stated problem.

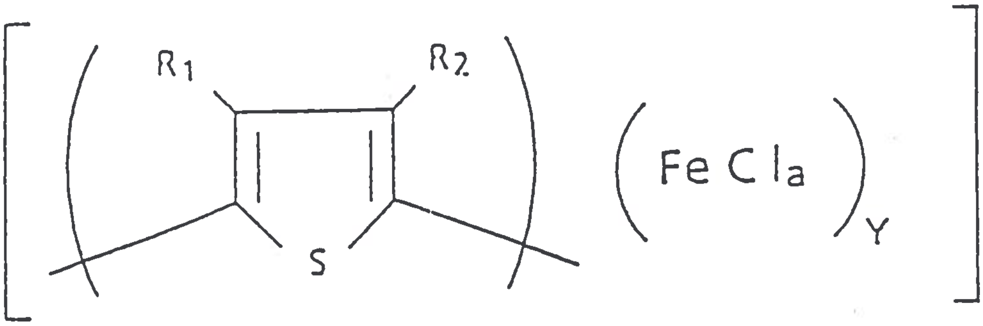
5. *Novelty*

This was the sole ground of refusal relied upon in the decision under appeal.

5.1 The closest state of the art, document D2, merely refers to polymers and copolymers of a substituted or unsubstituted thiophene. There is no disclosure of a poly(3-alkylthiophene). Furthermore, although D2 refers generally to mixing the polymer component (A) not having a conjugated π-system in known manner with the (conductive) polymer component (B) or with monomers which yield the component (B), only the latter method, in which the conductive polymer is formed *in situ*, is described in further detail. In other words, there is no example in which the conductive polymer according to D2 is processed in melt form. Consequently, there is no disclosure of any melt processable conductive polymer, let alone a melt processable poly(3-alkylthiophene).

Consequently, the subject-matter of Claim 1 is novel in the light of D2.

5.2 According to D1, an electrically conductive polythiophene or substituted polythiophene is characterised in that it has been doped or treated with $FeCl_3$ or with compounds containing $FeCl_4$ anions (Claim 1). The polythiophene or substituted polythiophene may have the following structure:



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where R_1 and R_2 are either both hydrogen atoms or one or both are an inorganic substituent or an organic substituent such as CH_2CN, $0CH_3$, CH_3, C_2H_5 or C_6H_5, a = 3 to 5, Y = 0.0001 to 0.50 and X > 4 (Claim 2).

The resulting polythiophene may be laminated to or applied as a coating on the surface of substrates consisting of other materials, such as plastics or metals, or between them. The polythiophene may equally be used in the shape of electrically conductive granules or powder (page 3, lines 29 to 36).

Thus, whilst D1 refers to poly(3-alkyl thiophene) species in which alkyl is methyl or ethyl, it is clear from the evidence of the Appellant that such species will not be melt processable. Consequently, they do not fall within the terms of a "melt processable poly (3-alkyl thiophene)" as defined in Claim 1 (section 4.10.2, above).

Consequently the subject-matter is novel over the disclosure of D1.

5.3 Hence the subject-matter of Claim 1, and, by the same token, of Claims 2 to 8, which are directly or indirectly dependent on Claim 1, is novel.

6. Although the sole ground of refusal was lack of novelty, the view was expressed in the decision under appeal that there would also be a lack of inventive step (Reasons for the decision, point 6.). In view of the arguments provided by the Appellant on that issue, the Board considers it appropriate to deal with it also.

7. *Inventive step*

It has to be considered whether the person skilled in the art, starting from D2 and endeavouring to provide an electrically conductive polymer composite of superior mechanical properties and wider applicability, would have expected that the provision of a substantial alkyl chain at the 3-position of the thiophene polymer would result in the conductive polymer itself being melt processable, and furthermore compatible with polymers not having active redox groups, thus opening the way to a wider variety of composites.

7.1 Document D2 does not disclose any melt processable conductive polymer (section 5.1, above), let alone suggest that such a melt processable polythiophene may be obtained by substitution of the thiophene molecule with an appropriate alkyl group at the 3-position.

Consequently, there is neither any hint of the precise effect on which the application in suit is based, nor of the means by which such an effect might be provided.

7.2 As to the disclosure of D1, this is not concerned with the stated problem of providing melt processability, but, on the contrary, with providing thermal stability. Indeed, in the examples of D1, the polythiophene is always in the form of an evaporated film or a dry powder, which does not involve melt processing. There is no reason, therefore, why the skilled person should regard its disclosure as relevant to the solution of the stated problem in the first place.

7.3 Even if the skilled person did, for some reason, attempt to make use of the teaching of D1, there is nothing to suggest the use of poly(3-alkyl thiophene) species having an alkyl group higher than ethyl. On the contrary, the most highly preferred substituted polythiophene, and the only one exemplified according to D1, is one in which the substituent is methyl (page 4,lines 6 to 14; Example 7).

Consequently, there is no hint either of the stated problem or its solution in D1.

7.4 In other words, the subject-matter of Claim 1 does not arise in an obvious way from the state of the art. On the contrary, the wide applicability of the conductive polymers due to the lack of any necessity of having active redox groups present in the non-conductive polymer, must be regarded as an unexpected technical advantage.

Consequently, the subject-matter of Claim 1 involves an inventive step. By the same token, the subject-matter of dependent Claims 2 to 8 also involves an inventive step.

8. In view of the above finding, it is not necessary for the Board to consider the auxiliary request of the Appellant.

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Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is referred back to the Examining Division with the order to grant a patent on the basis of the main request, that is pages 1, 2, 4 to 8 of the application as originally filed and published, and pages 3 and 3a filed with the main request of the letter of 29 August 1996 together with Claims 1 to 8 also filed on that date, with the amendments indicated in the letter dated 29 April 1997 (section V., above).

The Registrar:

E. Görgmaier

The Chairman:

C. Gérardin

C. Gérardin