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D E C I S I O N
of 28 March 1996

Case Number: T 0181/93 - 3.3.3

Application Number: 86308517.1

Publication Number: 0222566

IPC: C08K 5/00

Language of the proceedings: EN

Title of invention:
Stabilised polyolefin composition

Applicant:
SUMITOMO CHEMICAL COMPANY, LIMITED

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (no) - obvious selection"

Decisions cited:
T 0813/93

Catchword:
-



Case Number: T 0181/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 28 March 1996

Appellant: SUMITOMO CHEMICAL COMPANY, LIMITED
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Decision under appeal: Decision of the Examining Division of the European Patent Office posted 1 September 1992 refusing European patent application No. 86 308 517.1 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
R. E. Teschemacher

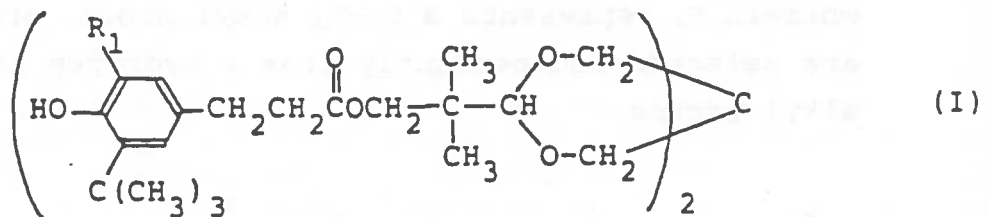
Summary of Facts and Submissions

I. This appeal, which was filed on 25 September 1992, lies against the decision of the Examining Division dated 1 September 1992, refusing European patent application No. 86 308 517.1 in the name of Sumitomo Chemical Company, Limited filed on 31 October 1986, claiming the priority of 8 November 1985 from two JP-applications, and published under No. 222 566. The appeal fee was paid on 30 September 1992 and a Statement of Grounds of Appeal was filed on 11 January 1993.

II. The decision under appeal was based on the set of 10 claims as originally filed, independent Claim 1 reading as follows:

Claim 1:

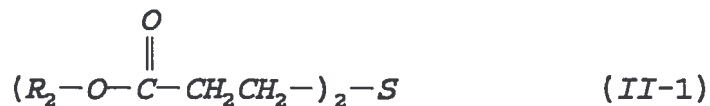
"A polyolefin composition which comprises (A) 100 parts by weight of a resin composition comprising 100 to 35 wt.% of polyolefin and 0 to 65 wt.% of inorganic filler; (B) at least 0.01 part by weight of phenolic compound selected from those of formula (I)



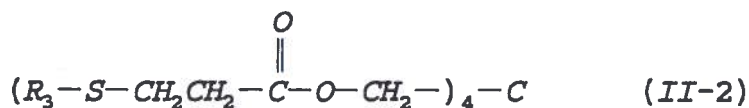
wherein R₁ represents a C₁-C₃ alkyl group, (C) at least 0.01 parts by weight of metal deactivator having one or more -C-N- groups in the side chain, and



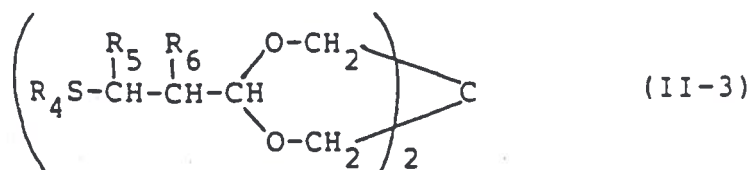
(D) 0.01 to 1 part by weight of sulfur-containing compound selected from those of formulae (II-1), (II-2), (II-3) and (II-4):



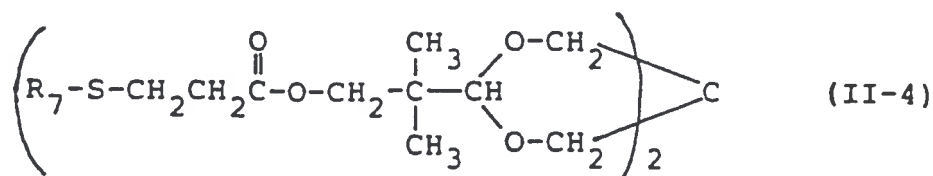
wherein R₂ represents a C₄-C₂₀ alkyl group,



wherein R₃ represents a C₄-C₂₀ alkyl group,



wherein R₄ represents a C₃-C₁₈ alkyl group, and R₅ and R₆ are selected independently from a hydrogen atom and alkyl groups,



wherein R₇ represents a C₃-C₁₈ alkyl group."

Claims 2 to 10 concerned preferred embodiments of the polyolefin compositions according to Claim 1.

III. The decision under appeal held that the claimed subject-matter was novel over the cited prior art but did not involve an inventive step over the combined disclosures of documents

(1) EP-A-155 847 and

(2) R. Gächter et al. "Taschenbuch der Kunststoff-Additive", 1979, pages 69 to 76, Carl Hanser Verlag München.

The polyolefin stabilizer compositions disclosed in document (1) differed from those according to Claim 1 only by the selection of certain metal deactivators. However, this selection was obvious in the light of document (2) and there was no evidence to show that the selected metal deactivators would bring about any technical effect not obtained with deactivators of a different kind.

IV. In the course of the written appeal proceedings the Appellant submitted various alternative sets of claims. From these, during the oral proceedings held on 28 March 1996, he maintained the following sets:

Main request: Claims 1 to 11 submitted on 13 October 1995 as (then) main request;

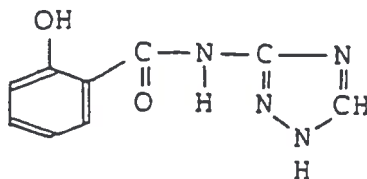
Auxiliary request I: Claims 1 to 10 submitted on 13 October 1995 as (then) auxiliary request;

Auxiliary request II: Claim 1 to 10 submitted on 28 February 1996 as (then) auxiliary request.

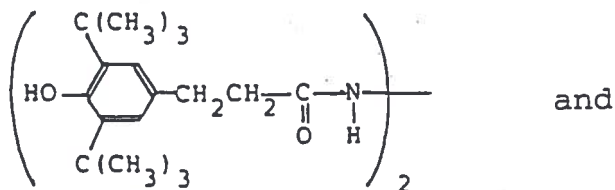
Claim 1 of the main request differs from the main claim as originally filed (see point II above) only in the definition of component (C), which is as follows:

"(C) at least 0.01 parts by weight of metal deactivator selected from

(a)

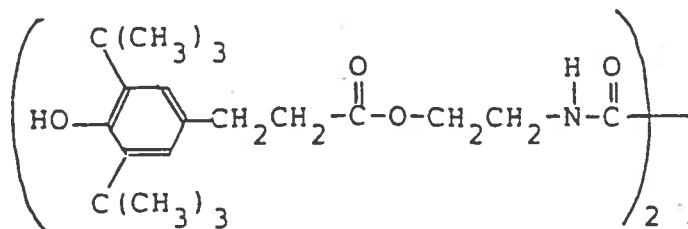


(b)



and

(c)



The respective Claims 1 of the auxiliary requests I and II differ from Claim 1 of the main request only by their further restriction of component (C) to

- compounds (a) and (b) (auxiliary request I) or
- compound (b) (auxiliary request II).

Apart from their respective Claims 1, all requests comprise a number of dependent claims (main request: 10, auxiliary requests: 9 each) which relate to preferred embodiments of the compositions of Claim 1.

- V. The Appellant argued essentially that the choice of the metal deactivator compound(s), to which the respective Claims 1 are now restricted, and their (its) combination with the further stabilizer components (B) and (D) were non-obvious over the combined disclosure of documents (1) and (2).

In support of his arguments he filed the results of new comparative experiments which in his view would demonstrate that by the use of these selected metal deactivator compound(s) polypropylene compositions could be obtained having considerably improved values of TEIP (thermal embrittlement induction period) and YI (yellowness index). In the Appellant's view, these unpredictable results showed that the metal deactivator compound(s) to which Claim 1 was now restricted represented a true selection from the extensive list of alternatives disclosed in document (2).

In this respect the Appellant referred also to decision T 0813/93 (dated 17 October 1994, not published in the OJ EPO), where it was stated that the selection of one solvent from a vast number of possibilities was non-obvious.

- VI. The Appellant requested that the appealed decision be set aside and that a patent be granted on the basis of the main or of the auxiliary requests I or II, all as set out in point IV above.

Reasons for the Decision

- 1. The appeal is admissible.

Claim 1 of all Requests

- 2. *Amendments*

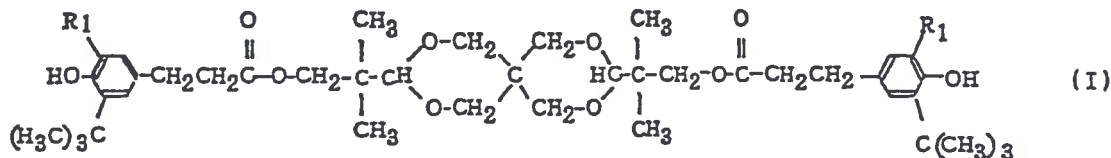
The metal deactivator compounds, to the use of which the Claim 1 of the main and the auxiliary requests have been narrowed down, were disclosed on pages 7 and 8 of the original application as compounds No. 1 (compound (a) according to the main request and the auxiliary request I), No. 3 (compound (b) according to the main request and sole metal deactivator compound according to the auxiliary request II) and No. 4 (compound (c) according to the main request).

The requirements of Article 123(2) EPC are therefore met by these amendments.

- 3. *Novelty*

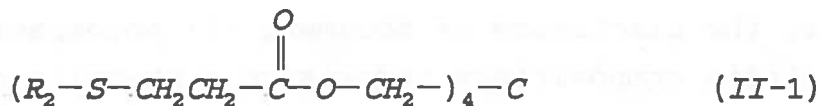
- 3.1 *Prior art*

As the application in suit, document (1) relates to an improvement of the thermal and oxidation stability of synthetic resins including polyolefins. According to (1) this is accomplished by the use of a combination of at least one phenol type compound of the formula (I)

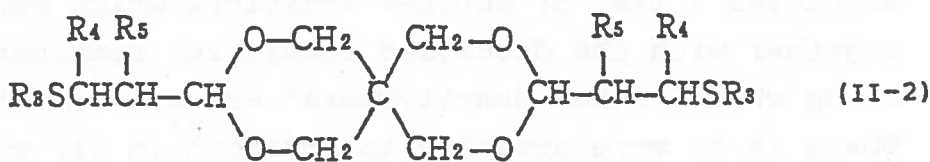


wherein R_1 represents a C_1 - C_3 alkyl group,

with at least one sulfur type compound (II) selected from



wherein R_2 represents a C_4 - C_{20} alkyl group, and



wherein R_3 represents a C_3 - C_{18} alkyl group, and R_4 and R_5 are selected independently from a hydrogen atom and C_1 - C_6 alkyl groups; the weight ratio of the phenol type compound(s) (I) to the sulfur type compound(s) (II) is preferably 1 to 0,5 to 15 (see Claims 1, 6, 8 to 10; page 2, line 15 to page 3, line 17; page 8, lines 9 to 17).

The definition of the phenolic type compound (I) according to document (1) is identical to formula (I) of the phenolic compound (B) according to Claim 1 of all of the present requests. In the same way the sulfur type formulae (II-1) and (II-2) of document (1) correspond fully to the formulae (II-2) and (II-3) according to Claim 1 of said requests.

Procedure 1 of document (1) (pages 11 to 15 in conjunction with Table 1 on page 4) discloses blends (Examples 1 to 5, Table 4) comprising 100 parts by

weight of a polypropylene resin, 0,05 parts by weight of a phenol type stabilizer (I-1) [$R_1 = CH_3$] and 0,1 to 0,3 parts by weight of a sulfur type stabilizer (II-1-1) [$R_2 = C_6H_{13}$], (II-1-2) [$R_2 = C_{12}H_{25}$] or (II-1-3) [$R_2 = C_{18}H_{37}$].

Hence, the disclosure of document (1) encompasses polyolefin compositions comprising a phenolic compound (B) and a sulfur-containing compound (D) in the amounts required by Claim 1 of all of the present requests.

Furthermore, on page 7, first paragraph, document (1) discloses a list of further additives which may be used together with the described stabilizer combination, among which "metal deactivators" are also mentioned. There is no more detailed information in (1) concerning this optional embodiment. It is, however, self-evident (and was not contested by the Appellant) that, in the existing context of thermal and oxidation stabilization, the purpose of adding a metal deactivator can only have been that to alleviate the negative influence (activity) which metals may have on the effectivity of the said stabilization.

3.2 Difference between the claimed subject-matter and the prior art

The only difference of the subject-matter of the application in suit as claimed with respect to that of document (1) resides in the additional presence of at least 0,01 part by weight of **certain** metal deactivators as defined in Claim 1 of the respective requests.

3.3 Since document (1) is completely silent about these particular metal deactivator compounds, the subject-matter of Claim 1 of all of the present requests is novel over this document.

4. *Problem and solution*

4.1 According to the discussion of the state of the art in the application in suit (page 1, last paragraph to page 3, line three from the bottom) the known stabilizer systems have not been sufficiently effective in suppressing the thermal deterioration of polyolefins when these are in contact with heavy metals, like copper.

For the skilled person being confronted with the need for further thermal stabilizers for polyolefins which should be more effective in this respect document (1) is a very promising starting point, since it offers a particularly advantageous combination of two stabilizers, one of the phenol type and the other one of the sulfur type, and suggests already the addition of metal deactivators, without, however, specifying them.

4.2 The problem underlying the alleged invention, when starting from document (1) as most relevant piece of the prior art, was thus the optimization of the thermal stabilization of polyolefin compositions provided by the stabilizer combination offered by document (1) against that kind of deterioration triggered by the presence of heavy metals, e.g. copper.

4.3 According to Claim 1 of the existing requests the solution of this problem is to be seen in the selection of certain specified metal deactivator compounds. This is confirmed by the results contained in the Experimental Report filed on 13 October 1995. Therein the TEIP and YI values achieved by the addition of the metal deactivator compounds used according to the present requests, designated C-1, C-2 and C-3 (corresponding in this order to the metal deactivator compounds (c), (a) and (b) according to the main

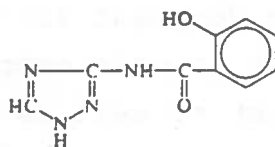
request), have been compared with the TEIP and YI values obtained by using other conventional metal deactivator compounds C-4, C-5, C-6, C-7 and C-8, as well as the values achieved in the absence of any metal deactivator (Run No. 9). These experiments demonstrate that the compounds C-1, C-2 and C-3 are particularly effective in suppressing the thermal embrittlement; this is shown by the high TEIP values of 10, 12 and 17 days as compared with values of from 1 day (control Run No. 9) to 9 days for the comparative experiments. The YI values do not vary significantly.

5. *Obviousness*

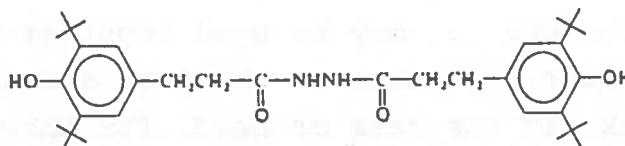
- 5.1 Document (2) is part of the textbook "Taschenbuch der Kunststoff-Additive" published in 1979. The disclosure contained in this document with respect to metal deactivators is highly relevant to the meaning of this term in document (1).

At page 70, paragraph 1, of document (2) it is reported that sterically hindered phenolic stabilizers as well as aromatic diamine stabilizers can only insufficiently retard the metal-catalyzed thermal oxidation of polyolefins, but that metal deactivators, when used together with said stabilizers, provide a much better stabilization. The fact that for an effective stabilization against metal-catalyzed degradation of polyolefins metal deactivators are to be used in combination with sterically hindered phenolic or aromatic amine antioxidants is again emphasized in the same document (2) at page 71, first paragraph of the Chapter 2.3 "Wirkungsweise von Metalldeaktivatoren bzw. Kupferinhibitoren".

At pages 74 to 76 of document (2) eleven major classes (a) to (k) of compounds are set out which had been proposed as metal deactivators in the previous 15 years (cf. "2.5 Stoffklassen"). Among the exemplified metal deactivators are compound (IX)



and compound (XI)



(+ = tert. butylgroup)

5.2 A comparison of the structure of these compounds (IX and (XI) with that of the metal deactivators used according to the Claims 1 of the present requests reveals that compound (IX) corresponds to deactivator (a) according to the main and the first auxiliary requests and compound (XI) corresponds to deactivator according to the main and the first auxiliary requests as well as to the sole deactivator used according to the second auxiliary request.

The metal deactivator compound (c) according to the main request is not disclosed in document (2); its structure is, however, in line with the recommendation given in (2) for the deactivator class h) to combine sterically hindered phenols and metal complexing groups (see (2) page 75).

5.3 It remains to be decided whether the selection of the metal deactivator compounds specified in the present requests involves an inventive step. In this respect it has to be borne in mind that Claim 1 of all requests comprises the metal deactivator compound (b) (compound (XI) according to document (2)) and that conclusions drawn with respect to this compound are thus applicable to the main request as well as to auxiliary requests I and II.

5.4 In essence, the statement in document (1) page 7, first paragraph "In applying the stabilizer of the present application, other additives such as metal deactivators ... may be used together with said stabilizer." amounts to the very straightforward advice to look, in the case of need, for known metal deactivators which are sufficiently effective. The skilled person is, thus, directed to investigate the relevant state of the art, here document (2), and to carry out the necessary experiments.

This is a routine task that does not require inventive skill.

Moreover, document (2) explicitly states (cf. the four last lines on page 76) that two of the selected compounds, namely (IX) and (XI), belong to the small group of only five compounds which are said to be of technical relevance (i.e. have acquired "Technische Bedeutung").

Since, before embarking on costly experimental investigations, one skilled in the art will normally consider in the first place those alternatives which, for technical and/or economical reasons are likely to be

the most profitable ones, it appears that in the present instance the suitability of compounds (IX) and (XI) as metal deactivators will be discovered after very few experiments.

- 5.5 The Appellant's argument that, in view of their unexpectedly good performance, the selection of the metal deactivators (a), (b) and (c) was non-obvious, is not convincing.

Any chemist and certainly one skilled in the art of plastic compositions is aware of the fact that **different** compounds, in spite of their common ability to act as stabilizers, must invariably have **different** stabilizing activities. This, because any chemical reaction is highly affected by changes in the structure of one of the reactants. In the present case of thermooxidative polyolefin degradation processes the reaction situation is particularly complex, since several chemical moieties interact (i.a. hydrocarbon, oxygen, metal ions, phenol type and sulfur type stabilizers).

It is therefore not at all surprising that the compounds tested in the Appellant's Experimental Report provide different TEIP values (and less different YI values). Indeed, the contrary would be very surprising.

The discovery of some metal deactivators from the host of compounds disclosed in document (2) which are particularly effective is therefore only the result one skilled in the art will obtain automatically when performing his routine investigations (see preceding paragraph 5.4).

- 5.6 Decision T 0813/93, cited by the Appellant, is not at variance with the above conclusions. Therein it was found that the choice of N-methylpyrrolidone (NMP) as solvent for the recrystallization of hexanitrostilbene was non-obvious over a certain document (citation (3)) where dimethylformamide (DMF) was used for the same purpose, because the skilled person "would have been confronted with a vast number of possibilities [other aprotic polar solvents] without any guidance which one to select." (reasons 4.3 and 4.4). That case is different from the present in various aspects. Firstly, said citation did not suggest that another aprotic polar solvent would be able to provide better results than NMP and, secondly, the number of alternative solvents was considered to be vast and without guidance, whereas here (document (2)) there is a very distinct preference for few compounds, among them two of the metal deactivators chosen by the alleged invention.
- 5.7 In view of the above considerations the Board concludes that no more than routine experimentation was required for a skilled person looking for a solution to the problem defined in Section 4.1.2 above to find out the suitability as metal deactivators of compounds (a) and (b) according to the present requests (compounds (IX) and (XI) in document (2)) when used in combination with the phenol type and sulfur type stabilizers recommended in document (1).
6. Since compound (XI) is to be used according to Claim 1 of the main as well as of the auxiliary requests I and II, the obviousness of its use entails that none of these requests complies with the requirements of Article 56 EPC. The same conclusion is to be drawn for the main request and for auxiliary request I with respect to compound (IX), the use of which is obvious as well.

7. Since the remaining claims of the requests must share the fate of their respective main claims, there is no need for further comments on them.
8. All requests must therefore be refused.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:


E. Gorgmayer

The Chairman:


C. Gérardin

