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D E C I S I O N
of 12 December 2000

Case Number: T 0613/98 - 3.3.3

Application Number: 93901328.0

Publication Number: 0617713

IPC: C08J 7/02

Language of the proceedings: EN

Title of invention:
Method for removing residual additives from elastomeric articles

Applicant:
SCHERING CORPORATION

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - unobvious solution"

Decisions cited:
-

Catchword:
-

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Case Number: T 0613/98 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 12 December 2000

Appellant: SCHERING CORPORATION
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 28 January 1998 refusing European patent application No. 93 901 328.0 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: C. Gérardin
Members: P. Kitzmantel
A. Lindqvist

Summary of Facts and Submissions

I. This appeal, which was filed on 23 March 1998, lies against the decision of the Examining Division dated 28 January 1998, refusing European patent application No. 93 901 328.0 filed as PCT/US92/10742 on 17 December 1992 in the name of SCHERING CORPORATION, and published under No. WO-A-93/12161 (EP-A-0 617 713). The appeal fee was paid together with the Notice of Appeal and the Statement of Grounds of Appeal was filed on 2 June 1998.

II. The decision under appeal was based on a set of 14 claims of a main request filed with a submission dated 24 July 1996 and on 8 claims of an auxiliary request submitted during oral proceedings before the Examining Division on 10 December 1997.

(i) Independent Claims 1 and 9 of the main request read as follows:

"1. A method for cleaning a vulcanised elastomeric article containing phthalate and/or PAH impurities integrated throughout its matrix comprising contacting the elastomeric article with at least one supercritical fluid under conditions and for a time sufficient to remove phthalates and/or polynuclear aromatic hydrocarbons (PAHs) contained therein whereby the cleansed vulcanised elastomeric article has a lower phthalate and/or PAH impurity content than a comparable vulcanised elastomeric article cleaned conventionally by refluxing in FREON P11 for 72 hours."

"9. A cleaned vulcanised elastomeric article

having a lower phthalate and/or PAH content than a comparable elastomeric article cleaned conventionally by refluxing in FREON P11 for 72 hours wherein prior to cleaning the elastomeric article contained phthalate and/or PAH impurities integrated throughout its matrix."

The further Claims 2 to 8 and 10 to 14 were dependent on, respectively, Claims 1 and 9.

(ii) The set of claims of the auxiliary request differed from that of the main request solely by deletion of Claims 9 to 14 from the latter.

III. The arguments of the decision under appeal may be summarized as follows:

(i) The subject-matter of Claims 9 to 14 of the main request was not novel over the known cleansed elastomeric articles referred to on pages 1 and 2 of the application in suit, because - even if it was arguably accepted that their purity was enhanced thereover - this feature could not be determined on the cleansed article.

(ii) Novelty could also not be conceded if Claim 9 was regarded as a product-by-process claim, because a higher degree of purity did not qualify as a distinguishing feature.

(iii) The decision under appeal held, moreover, that the subject-matter of Claim 1 of both requests lacked inventive step, because it was obvious in view of the disclosure of document

D3: Kirk-Othmer Encyclopedia of Chemical Technology, Supplement, 1984, pages 872 to 877

to enhance the amount and rate of removal of phthalate and PAH impurities by replacing the known Freon solvents by supercritical fluids.

(iv) In the Examining Division's view, documents

D2: Encyclopedia of Polymer Science and Engineering, vol. 16, 1989, pages 368, 369 and 387, and

D4: Chemtech, January. 1986, pages 52 to 56

did not militate against the said replacement, because these documents indicated that any damage that might be caused to the articles by the treatment with a supercritical fluid could be avoided by appropriate selection (1) of this fluid and (2) of the decompression conditions.

IV. At the oral proceedings held on 12 December 2000, following the discussion of the set of claims filed together with the Statement of Grounds of Appeal, the Appellant filed an amended set of three claims, which read as follows:

"1. A method of cleaning a vulcanised nitrile rubber article in the form of a gasket, valve, seat, flap, stopper or plug for use in a metered dose delivery device containing phthalate and/or polynuclear aromatic hydrocarbon (PAH) impurities integrated throughout its matrix, the method comprising contacting the

elastomeric article with supercritical carbon dioxide under conditions of pressure of 50 to 400 atm, a temperature from 30° to 50°C and for a time of 1 to 4 hours."

"2. The method of Claim 1 wherein the phthalate being removed is dibutyl phthalate or diisooctyl phthalate."

"3. The method of Claim 1 or Claim 2 wherein the metered dose delivery device is an aerosol container containing chlorofluorohydrocarbons or fluorohydrocarbon propellants."

V. The arguments presented by the Appellant in the Statement of Grounds of Appeal, the letter dated 14 July 2000 (submitted in response to the Rapporteur's communication dated 18 May 2000) and during the oral proceedings may be summarized as follows:

(i) As compared with conventional cleaning procedures by solvent extraction with fluorocarbon type liquids the present invention provided nitrile rubber articles having lower contents of phthalate and/or PAH impurities without sacrificing their mechanical and physical properties by a method which was simpler, faster, more economical and environmentally safer.

(ii) Account being taken of the conditions of pressure, temperature and treatment time specified in the worked example, the claimed subject-matter was disclosed in the original specification in a manner sufficiently clear and complete for it to be carried out by a person

skilled in the art.

(iii) The skilled person faced with the problem of developing an alternative method for the provision of rubber articles which are sufficiently clean for medicinal or pharmaceutical use would not consider any of the cited documents D1 to D4, which all related to different technical fields.

(iv) Even if considered, the cited prior art was unable to suggest that by using supercritical carbon dioxide nitrile rubber articles could be prepared having an impurity content which is lower than according to the conventional solvent extraction technology without any deterioration of the mechanical and physical properties of the cleansed articles.

(v) This argument was valid in particular with respect to documents D2 and D4, which disclosed that by prolonged contact of elastomeric seals with supercritical carbon dioxide their integrity was damaged by swelling, inflation and embrittlement.

(vi) Document D1 (FR-A-2 638 098), although relating to the extraction of impurities from rubber particles by supercritical carbon dioxide, was no relevant prior art, because it was only concerned with unvulcanised rubber.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the set of three claims submitted during oral

proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

Claim 1 is supported by its original version, by original Claims 3 ("supercritical carbon dioxide"), 7 ("nitrile rubber") and 9 ("article in the form of a gasket, valve, seat, flap, stopper or plug for use in a metered dose delivery device") as well as by the statements in the original description on page 4, lines 4 to 33 ("vulcanized ... article") and on page 6, lines 14 to 26 ("conditions of pressure of 50 to 400 atm, a temperature from 30° to 50°C and for a time of 1 to 4 hours").

Claims 2 and 3 are supported by original Claims 2 and 10.

The claims, thus, comply with the requirement of Article 123(2) EPC.

3. *Article 84 EPC*

The claims also meet the requirements of Article 84 EPC. In particular, Claim 1 is adequately supported by the description in that it specifies all the process features required in combination to achieve the desired result, which provides a clear definition of the claimed subject-matter.

4. *Article 83 EPC*

The application in suit also complies with the requirement of sufficiency of disclosure according to this article.

On the basis of the information contained in the original specification, particularly the worked Example set out on pages 11 to 14, the Board is satisfied that the invention as specified in Claim 1 can be carried out, i.e. that the skilled person is enabled by this information to extract phthalate and PAH impurities from vulcanised nitrile rubber articles with supercritical carbon dioxide at the specified temperature, pressure and time conditions without damaging the integrity of the article.

5. *Citations*

5.1 Document D1

This document discloses a method of cleaning a rubber powder by extracting the impurities contained therein, including solvents, monomers, oligomers and by-products with an extracting agent, e.g. carbon dioxide, possibly in a supercritical state (cf. Claims 1, 3, 5 and 7; page 9, line 26 to page 10, line 25; page 11, Example 1).

Although this is not explicitly stated in D1, it is evident from the discussion of the deficiencies of the state of the art purification techniques in D2, especially the problem of the sticking together of the particles, that the rubber to be used is unvulcanised (cf. page 2, lines 1 to 8; page 3, lines 7 to 12).

5.2 Document D2

The cited Section "Supercritical Fluid" of this encyclopedia states on page 368, penultimate paragraph that "Supercritical fluids exhibit a unique combination of solvent and transport properties and have been used to ... extract residual solvents, monomers, and catalysts from solid and molten polymers" (see also page 371, 4th paragraph).

Carbon dioxide in supercritical state is exemplified as agent for the extraction of cyclic siloxane species from silicone polymers and of low molecular weight organic impurities from polystyrene and from atactic polypropylene (cf. page 372, penultimate paragraph; page 378, first paragraph and lines 1 to 4 of second paragraph).

On page 387, penultimate paragraph it is stated: "In the case of elastomeric materials used for seals and O-rings in supercritical-fluid processes, the absorption of a gas at high pressure and the subsequent release of the dissolved gas at ambient pressure (when an extraction is completed, for example) causes many polymeric sealing materials to degrade (54) [this literature reference is the present document D4]. When the system pressure is reduced, adsorbed gases nucleate into bubbles and cause blistering of the seals. In addition, the supercritical fluid can extract the plasticizing agents in the seals and thus cause embrittlement."

5.3 Document D3

The Section "Supercritical Fluids" of this encyclopedia

states in the paragraph bridging pages 875 and 876: "Although a supercritical fluid has a density approaching that of a liquid for high solvent capacity, the diffusivity is orders of magnitude greater giving improved mass-transfer rates. In addition, the lower viscosity also provides advantages such as enhanced solid settling rates during precipitation. For these reasons, supercritical solvents are superior to liquids for penetrating the micropores of a solid structure such as coal" (see also page 877, fifth paragraph).

On page 877, fourth paragraph the importance of the nontoxic, nonhazardous character of supercritical carbon dioxide for the use in the food and pharmaceutical industries is emphasized, as opposed to liquid extraction methods which leave toxic residues even after cumbersome elimination stages.

5.4 Document D4

This paper relates to the use of elastomeric seals for applications, e.g. in the food and petroleum industry, which involve the use of supercritical fluids, e.g. carbon dioxide. It is set out that the strong solvent capacities of such fluids lead to some swelling of the seals and to inflation upon release of the gas pressure, which may cause voids, blisters and even fractures (page 52, left hand column, first three paragraphs; page 54, left hand column, last three paragraphs to right hand column, first paragraph; page 55, left hand column, last paragraph).

6. *Novelty*

6.1 None of the citations discloses a method of cleaning a

vulcanised nitrile rubber article from phthalate and/or PAH impurities with supercritical carbon dioxide.

The subject-matter of Claim 1 is, therefore, novel over the cited documents.

6.2 Owing to the different extraction agent supercritical carbon dioxide the subject-matter of Claim 1 is also novel over the (unidentified) state of the art referred to on page 2, lines 9 to 12 of the description of the application in suit, which relates to the liquid-solid extraction of phthalate and/or PAH impurities from rubber articles with conventional solvents or fluorocarbon type solvents.

6.3 The acknowledgement of novelty also applies *a fortiori* to the subject-matter of the dependent Claims 2 and 3.

7. *Closest prior art, problem and solution*

7.1 The prior art which comes closest to the claimed subject-matter is the liquid-solid extraction method with conventional or fluorocarbon type solvents referred to in point 4.2 supra, because none of the citations D1 to D4 relates to the removal of phthalate and/or PAH impurities from vulcanised nitrile rubber articles.

7.2 According to the application in suit the problem underlying its subject-matter comprises *inter alia* the provision of a method for the removal of impurities from elastomeric articles, which - in comparison with conventional techniques - not only is occupationally and environmentally safer as well as more rapid, but is also more efficient in removing phthalate and PAH

impurities, which are contained therein without impairing the integrity of the article or its physical properties (cf. page 2, line 23 to page 3, line 30; page 10, lines 18 to 19 and lines 24 to 27).

7.3 According to present Claim 1 the solution of the various aspects of the afore-mentioned problem is to be achieved by the use of supercritical carbon dioxide as extraction agent at the indicated temperature, pressure and time conditions.

7.4 Table I of the worked Example set out on pages 11 to 14 of the description of the application in suit shows that the extraction of impurities, particularly dibutyl phthalate and diisooctyl phthalate, from nitrile rubber valve components with supercritical carbon dioxide for four hours is more effective than the extraction by refluxing in the fluorocarbon type solvent Freon(R)P11 for 72 hours.

Table II of this Example indicates that the cleaning of unprocessed rubber with supercritical fluid did not change the hardness and the physical properties of the treated rubber.

Table III discloses that the residual amounts of non-volatile residues and PAH after the cleaning of unprocessed rubber were lower after four hours treatment with supercritical fluid than after conventional cleaning.

By these data it is established that the problem set out in point 7.2 supra has effectively been solved by the measures specified in Claim 1.

8. *Obviousness*

8.1 The issue of inventive step turns on the question whether it is obvious to solve the technical problem underlying the subject-matter of present Claim 1 as set out in point 7.2 supra by the replacement as extraction agent of known conventional and fluorocarbon type solvents (cf. point 6.2 supra) by supercritical carbon dioxide under the conditions specified in said claim.

8.2 In the judgment of the Board the present solution of the existing technical problem was unobvious.

In arriving at that conclusion the Board did consider the disclosure of documents D1 to D4, because - contrary to the Appellant's contention - they are all relevant to the technical field under discussion, be it because they concern the cleaning of rubber by extraction with supercritical carbon dioxide (D1), the interaction of supercritical carbon dioxide with elastomeric seals (D4) or the properties of supercritical fluids in general (D2 and D3).

Since the subject-matter of Claim 1 is concerned with the cleaning of rubber articles, their possible use for medical or pharmaceutical applications (cf. page 3, lines 17 to 20) does not mean that the competent expert for the assessment of its inventiveness is a pharmaceutical chemist. Rather the skilled person, whose knowledge is to be considered here, is still the polymer specialist.

8.3 The afore-mentioned conclusion of non-obviousness is based on the following considerations.

8.3.1 Although the skilled person is aware from general common knowledge, as reported in documents D2 and D3 (cf. points 5.2 and 5.3 supra), that supercritical fluids possess excellent solvent and transport properties, these documents are silent on the cleaning of vulcanised elastomers by extraction of impurities, phthalates and PAHs inclusive, with such fluids. D2 only relates to the extraction of uncrosslinked polymers, e.g. polyethylene, polystyrene and polypropylene, whereas D3 discloses the use of the supercritical fluid extraction for coal and in the food and pharmaceutical industries (D2: page 368, lines 2 to 6 of penultimate paragraph; page 372, lines 7 to 10; page 376, lines 1 to 4; page 378, penultimate paragraph and first sentence of last paragraph; D3: page 876, lines 3 to 4; page 877, 4th paragraph).

These documents cannot, therefore, *per se* suggest that supercritical carbon dioxide is able to remove phthalates and PAH impurities from vulcanised nitrile rubber much more efficiently, i.e. much faster and to considerably lower residual impurities, than conventional fluorcarbon type solvents (Freon(R) P11; cf. point 7.4 supra) (cf. D2, page 368, lines 2 to 6 of the penultimate paragraph; D3, sentence bridging pages 875 and 876; page 877, fifth paragraph).

8.3.2 Furthermore, insofar as D2 refers to the contact of **elastomeric** materials with supercritical fluids, when such materials are used for seals and O-rings in supercritical-fluid processes, it stresses that these materials undergo degradation by swelling and subsequent blistering. In this respect D2 also discloses that plasticizing agents may be extracted from such seals and thereby cause embrittlement (cf.

page 387, penultimate paragraph).

8.3.3 Similarly D4 points at the danger that "Some compressed gases, among them carbon dioxide, can produce large, pressure-dependent swelling of elastomers; and during pressure releases, regardless of gas type, there is a risk of the absorbed gas blowing up the elastomer" (cf. page 55, left hand column, lines 5 to 9 from bottom).

8.3.4 The sole information concerning vulcanised elastomeric materials, which is contained in documents D2 and D4, thus, warns that these materials may be damaged by swelling with and subsequent decompression of supercritical fluids.

8.3.5 Considering the situation outlined in points 8.3.1 to 8.3.4 supra the skilled person could not expect that it would be possible, by treating vulcanised nitrile rubbers with supercritical carbon dioxide under specific conditions,

(i) to remove phthalate and PAH impurities to lower levels than those achieved by conventional solid-liquid extraction

(ii) without damage and, thus, without impairing of their mechanical and physical properties.

8.3.6 The above conclusion is not affected by the disclosure of D1, which *inter alia* relates to the extraction of impurities from rubber particles with supercritical carbon dioxide, because according to this citation the rubber is not vulcanised (cf. point 3.1 supra). Any swelling and blowing up, which might also occur according to this document, would, therefore, have no

impact on the properties of the final article, which is prepared from the cleansed rubber particles, because voids and ruptures in the raw rubber material will not persist after moulding and vulcanisation.

8.4 The subject-matter of Claim 1 thus complies with the requirements of Article 56 EPC.

8.5 By virtue of their appendance to Claim 1 the same conclusion applies to Claims 2 and 3.

9. Apart from any necessary consequential amendment of the description the application in suit is thus in line with the requirements of the EPC.

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 set of Claims 1 to 3 submitted during oral proceedings, after any necessary consequential amendment of the description.

The Registrar: The Chairman:

E. Görgmaier C. Gérardin