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File Number: T 791/90 - 3.3.1

Application No.: 84 902 837.8

Publication No.: 0 149 664

Title of invention: A COMPOSITION AND A METHOD OF CAPTURING SULPHUR

Classification: C10L 1/32

D E C I S I O N
of 30 June 1992

Proprietor of the patent: AB CARBOGEL, et al

Opponent: SNAMPROGETTI S.p.A., Milano c/o ENIRICERCHÉ

Headword: Capturing sulphur/CARBOGEL

EPC Article 56

Keyword: "Inventive step (yes), non-obvious combination"



Case Number : T 791/90 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 30 June 1992

Appellant :
(Opponent)

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Decision under appeal :

Decision of Opposition Division of the European
Patent Office dated 14 August 1990 rejecting the
opposition filed against European patent
No. 0 149 664 pursuant to Article 102(2) EPC.

Composition of the Board :

Chairman : K.J.A. Jahn
Members : R.K. Spangenberg
J.-C. Saisset

Summary of Facts and Submissions

- I. The appeal was filed on 10 October 1990 and the prescribed fee was paid on 11 October 1990. It lies from the decision of the Opposition Division of the EPO of 14 August 1990 rejecting the opposition against European patent No. 0 149 664, granted in response to European patent application No. 84 902 837.8 filed on 11 July 1984 as International Patent application No. PCT/SE84/00259. The patent specification contained six claims; independent Claim 1 reads as follows:

"A method of capturing sulfur emanating from burning carbonaceous fuel particles, upon burning of an aqueous slurry of said carbonaceous fuel particles, prior to the point at which gaseous sulfur oxides are formed, characterised by the steps of:

- (a) providing an aqueous carbonaceous fuel composition slurry which comprises 60-80% by weight of carbonaceous fuel particles with an ash content of below about 5% by weight, on a dry basis, 0.05-2.0% by weight of a flow-enhancing chemical which includes a non-ionic dispersant, and a liquid carrier phase consisting essentially of water, and
- (b) mixing into said aqueous slurry a sulfur-capturing substance, having a particle size when present in solid form of less than 10 micrometer, which includes compounds of a metal with a higher affinity for sulfur than Fe and which is selected from the group consisting of hydroxides, oxides, and carbonates of calcium, magnesium, and manganese, in an amount of 0.1-5% by weight of the compound calculated as pure metal upon total solid fuel weight, thereby to provide uniform and intimate contact of said sulfur-

capturing substance with said carbonaceous fuel particles in said slurry."

Independent Claim 4 related to an aqueous carbonaceous fuel composition comprising the constituents specified under (a) and (b) in Claim 1.

The decision under appeal referred to the following documents:

E1 = US-A-4 282 006

E2 = US-A-3 941 552

E3 = EP-A-0 010 289

E4 = Combustion-Fossil Power Systems-1981-3rd edition, publ. Combustion Eng., Windsor, USA, pp. 17/53-54,

E5 = Proceedings of the 1982 Joint Symposium on Stationary combustion NOx Control - vol. 1 - Utility boiler applications - Electric Power Research Institute.

According to the Opposition Division the technical teaching of the patent in suit satisfied the requirement of Article 83 EPC. Further, the Opposition Division concluded that the subject-matter of the patent in suit was novel and inventive. The Opposition Division considered that document E2 represented the closest state of the art and that the other documents, in particular E1, did not suggest the solution to the problem of modifying the slurry of E2 in such a manner that a pipeline-pumpable slurry is produced which contains a higher amount of particulate carbonaceous material than the slurry of E2. In particular, two essential features, namely the fact that the carbonaceous fuel particles have an ash content

of below about 5% by weight, and that the sulfur capturing substance has a particle size of less than 10 micrometer, were not disclosed in either of the relevant documents E1 and E2.

II. A Statement of Grounds of Appeal was submitted on 11 December, 1990. The Appellant cited a further document:

E6 = The application of dry additives on reducing sulfur dioxide emission for brown-coal fired boilers - K. Hein, W. Glaser - Technol. Use Lignite 1981 - pp. 362-378 - published 1982.

In the Statement of Grounds of Appeal and in subsequent letters, the Appellant argued that the ash content of the carbonaceous fuel particles was not relevant to the solution of the stated technical problem and that the suitable particle size of the sulfur capturing substance could be derived from E4, E5 and E6. The objections against the sufficiency of the disclosure were maintained, based on fresh experimental evidence (amended examples N1 - N4 and new examples N5 - N12).

III. The Respondent protested against the late introduction of document E6 and examples N5 - N12. Further, he argued that the compound used as a dispersant in Examples N1 to N5 and in N8 to N12 was an anionic polymer, whereas the patent in suit prescribed a nonionic dispersant. It was alleged that N6 and N7 did not contain a dispersant, and that the expression "no fluid mixture" as used in these examples was not clear. It was submitted that the possibility of successfully repeating the working examples of the patent in suit had never been questioned, and that, therefore, the disclosure was in accordance with Article 83 EPC.

IV. The Appellant requests that the decision under appeal be set aside and the patent revoked.

The Respondent requests that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.
2. Document E6 and Examples N6 to 12 have been taken into consideration. They were filed together with the Grounds of Appeal as an immediate response to objections raised in the decision under appeal against the date of availability to the public of the subject-matter of document E5 and to the Comparative Examples N1 to N4. Therefore, in the Board's judgment, this evidence was not filed late.
3. In support of the alleged insufficiency of disclosure, the Appellant has filed 12 examples. However, the four examples submitted during the opposition proceedings were performed without a dispersant, i.e. contrary to the express requirement of Claims 1 and 4. It is true that Examples 1 to 5 of the patent specification also do not indicate the presence of a non-ionic dispersant. Nevertheless, in the Board's judgment, they must be seen in the context of the description, in particular column 6, lines 17 to 31. Thus it is implied that the coal/water slurry mentioned in the first line of Example 1 already contained a sufficient amount of a non-ionic dispersant. This has been confirmed by the Respondent during the pre-grant proceedings, see the letter received on 20 February 1987, page 2, second paragraph. In contrast, in the Appellant's Examples N1 to N5 and N8 to N12 filed with the statement of grounds of appeal, an anionic flow-enhancing compound was used (see the Appellant's letter received on

2 July 1991, page 1, last paragraph). For the additive used in Examples N6 and N7 no chemical structure was submitted by the Appellant, however, it was confirmed that a nonionic polymer was used. However, not all nonionic polymers are automatically dispersants and the data submitted are not sufficient to establish whether this compound was in fact a dispersant. Thus, these examples do not show that the information in the patent specification as a whole is not sufficient for the skilled person to carry out the method of Claim 1.

Moreover, even an incidental failure of a specific nonionic dispersant would not render the disclosure as a whole insufficient since the description would guide the person skilled in the art towards attaining the desired effect (see also e.g. T 14/83 OJ EPO 1984, 105).

The Board is therefore satisfied that no ground of opposition according to Article 100(b) EPC prejudices the maintenance of the patent.

4. After examination of the cited documents, the Board has reached the conclusion that the claimed subject-matter is novel. Since novelty is not in dispute, it is not necessary to give detailed reasons for this finding.
5. The sole remaining issue to be dealt with is whether the subject-matter of the claims involves an inventive step.
 - 5.1 In the Board's judgment, the closest state of the art is represented by document E1, which relates to a high-solids coal-water slurry which is suitable for long distance pipeline conveyance and which is especially suitable for

direct burning in a furnace (column 1, lines 22 to 30). A stable coal-water slurry is disclosed in Claims 4 and 12, comprising at least about 60 weight percent of coal, at least about 20 weight percent of water and dispersing agent, in which the coal must fulfill specific particle size requirements, and in which the dispersing agent is present at from 0.05 to 2 weight percent based on the dry weight of the coal. The coal particles of this slurry have a low ash content, e.g. less than 5% by weight (see column 1, line 35 and column 28, line 65 to column 29, line 3).

The dispersing agents used are preferably surfactants, which can be anionic, cationic or even in some cases nonionic (see column 29, lines 60 to 64).

- 5.2 In the light of this closest state of the art the technical problem to be solved by the subject-matter of the patent in suit can be seen in modifying this slurry, so that the sulfur emanating from burning particulates is captured prior to the formation of gaseous sulfur oxides (see the patent specification, column 2, lines 21 to 23), and, at the same time, maintaining sufficient pumpability of the slurry (patent specification, column 6, lines 17 to 20).
- 5.3 The patent proposes to solve this problem essentially by incorporating into the slurry a sulfur capturing substance, having a particle size when present in solid form of less than 10 μm , selected from hydroxides, oxides, and carbonates of calcium, magnesium, and manganese, in an amount of 0.1 - 5 weight-% of the compound calculated as pure metal upon total solid fuel weight, together with 0.05 - 2.0 weight-% of a flow-enhancing chemical which includes a non-ionic dispersant.

- 5.4 The Board is satisfied, having regard to Examples 1 to 5 of the patent in suit, that the present technical problem has been effectively solved.
- 5.5 No incentive to solve this problem in the way suggested by the disputed patent can be derived from document E1. Although it is true that in this document the problem of desulfuration is addressed, the solution offered therein consists in no more than conventional de-ashing, involving ball milling and NaOH treatment (see e.g. column 6, lines 47 to 49 and Example 4). It may also be immediately apparent to the skilled person that the preferred inorganic dispersants used in that document, in particular NaOH and K_2CO_3 (see column 6, lines 41 to 46) are sulfur capturing compounds. Nevertheless, this document is, in the first place, concerned with the improvement in pumpability of highly concentrated coal slurries. In this respect, it is expressly stated that the use of higher valence cations such as Ca, Al and Mg ions increases the tendency of the slurry to flocculate and therefore impairs its pumpability (see column 27, lines 4 to 14 and 50 to 58). This is, in the Board's judgment, more of a warning against the use of alkaline earth compositions, rather than an encouragement to do so. In addition, the document states that the nature of the surfactant depends on the surface chemistry of the coal particles to be used and may be anionic, cationic or even nonionic (see column 29, lines 53 to 64). However, no specific rules for selecting a particular surfactant for a particular coal are given and for all the coals specifically mentioned in the document the recommended surfactant is anionic (see for example Claim 13 and column 29, line 64 to column 31, line 17). Consequently, there is no hint in this document to the solution of the present problem, consisting essentially in combining a sulfur capturing substance selected from hydroxides, oxides, and carbonates of

calcium, magnesium, and manganese with a nonionic surfactant.

- 5.6 It is common ground among the parties that, e.g. from documents E2 and E6, the principle of removing sulfur from coal-based fuels by capturing the sulfur, with e.g. calcium carbonate, at the moment of the burning in order to avoid contamination of the off-gas with sulfur oxides, already belonged to the state of the art.

Thus, in document E2 a method is described for reducing sulfur oxide emissions in the combustion of a fuel comprising dispersing into the fuel finely pulverised lime or limestone (Claim 3). The amount of lime should normally be in excess over the stoichiometric amount based on the sulfur content of the coal but not more than twice the stoichiometric amount (column 4, lines 13-25). Coals which can be typically used contain from 1 to 2% of sulfur (column 5, lines 11 to 13). However, the fuel to which the method is applied is a dispersion of finely divided coal having a particle size of less than 100 μm , water and oil (see Claim 1). Therefore this document would not suggest to the person skilled in the art a solution to the technical problem of capturing sulfur from a pumpable coal-water slurry of high coal content that did not contain oil without adversely affecting the pumpability of such a slurry.

Document E6 relates in essence to an improved method for sulfur oxide retention during combustion of Rhinish brown coals. It discloses that the presence of additives such as $\text{Ca}(\text{OH})_2$, CaCO_3 , CaO and dolomite reduces the SO_2 -emission, i.e. the SO_2 -content of the off-gas. It also discloses that for industrial combustion a high specific surface area of the additive is a vital boundary condition (page 364, second paragraph). The mixture which is burned

does not contain water but solely coal and dry additives (page 368, second paragraph and page 375, last sentence). Thus no hint to the solution of the present technical problem can be derived from this document either.

5.7 Documents E3 and E4 do not even mention the problem of removing sulfur at the moment of burning. Document E3 describes a chemical treatment of coal for removing sulfur present in a specific form, namely in the form of pyritic sulfur (see Claim 1). The coal slurry used has a solids concentration in the range of between about 4 and 40% by weight of coal (page 5, lines 3 to 5), i.e. it cannot be burnt directly. Document E4 relates to the removal of sulfur dioxide from the off-gas of coal burning plants by the use of lime or lime stone. Therefore, the information contained in these documents could not help the person skilled in the art in solving the present technical problem.

5.8 Thus, since the combined use of a sulfur capturing compound selected from hydroxides, oxides, and carbonates of calcium, magnesium, and manganese and a nonionic surfactant for solving the present problem was not obvious, it is not necessary to investigate whether or not the remaining features of Claim 1, i.e. the selected particle size and the amount of sulfur capturing substance, would further contribute to the inventive step.

5.9 The same conclusion would be reached if, as in the decision under appeal, document E2 is considered as the closest state of the art. In this case the technical problem can be seen in providing an alternative to the pumpable slurry containing a sulfur capturing substance disclosed in that document. It is immediately apparent from the summary of the relevant content of documents E1

to E6 that they do not provide any suggestion that this technical problem would be solved by the essential features of Claim 1, in particular by omitting the oil and employing a nonionic surfactant.

5.10 Since essentially the same considerations apply to the composition of Claim 4, this composition also involves an inventive step. The dependent Claims 2 and 3 as well as 5 and 6 derive their patentability from that of the respective independent claims.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

E. Görgmaier

K. Jahn