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D E C I S I O N
of 20 November 1997

Case Number: T 1078/93 - 3.3.5

Application Number: 90905154.2

Publication Number: 0462216

IPC: B01J 37/00

Language of the proceedings: EN

Title of invention:

Process for improving the physical and catalytic properties of
a fluid cracking catalyst

Applicant:

THIELE KAOLIN COMPANY

Opponent:

-

Headword:

Surfactant/THIELE

Relevant legal provisions:

EPC Art. 84, 54

Keyword:

"Clarity and support (yes), 'acid stable', novelty (yes)"

Decisions cited:

T 0409/91

Catchword:

-



Case Number: T 1078/93 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 20 November 1997

Appellant:
(Opponent)

THIELE KAOLIN COMPANY
Kaolin Road
Sandersville, GA 31082 (US)

Representative:

von Füner, Alexander, Dr.
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Decision under appeal:

Decision of the Examining Division of the
European Patent Office posted 29 June 1993
refusing European patent application
No. 90 905 154.2 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: R. K. Spangenberg
Members: M. M. Eberhard
J. H. van Moer

Summary of Facts and Submissions

- I. European patent application No. 90 905 154.2 was refused by a decision of the examining division. The decision was based on the set of amended claims filed on 14 July 1992.
- II. The grounds for the refusal were that the subject-matter of claims 1 and 2 did not meet the requirements of Article 54 EPC in combination with Article 84 EPC. The examining division held that the expression "acid stable anionic surfactant" had no recognized technical meaning in the field of surfactants. Furthermore, the description contained no teaching which would have allowed the reader to ascertain whether a surfactant was "acid stable". In particular it was neither indicated whether the surfactant was stable to a strong or to a weak acid, nor for how long and at what temperature. In the absence of a clear definition in the description, this ambiguous expression rendered unclear the scope of protection sought.

Regarding novelty, the examining division took the view that D1 (WO 89/01362) described a process as defined in claims 1 and 2, in which the "acid stable anionic surfactant" was **formic acid**. In view of the definition of "surfactant" in D2 (Hawley's Condensed Chemical Dictionary, 11th edition, pages 1111-1112) and of the surface tension of formic acid indicated in D3 (CRC Handbook of Chemistry and Physics, 62nd edition, pages F-35, F38 and F39), there was no doubt that formic acid was an "acid stable anionic surfactant". Although it was used in D1 for peptizing alumina, its inherent properties of decreasing the surface tension of the aqueous slurry containing it implied that it also acted as a surfactant.

- III. The appellants lodged an appeal against this decision. They made reference to ten citations, in particular five commercial brochures, in support of their arguments in the statement of grounds of appeal and also filed additional examples. An auxiliary request was submitted on 10 August 1994. From the said citations, only Van Nostrand Reinhold Encyclopedia of Chemistry, 4th edition, 1984, pages 319, 320, 403, 404, 916 (hereinafter Exhibit 4) is of importance for the present decision.
- IV. In a communication from the board, the appellants were informed of some further possible objections under Article 84 EPC in connection with claim 1 of both requests. The board also made reference to Encyclopedia of Chemical Technology, Kirk-Othmer, 3rd edition, vol. 22, pages 347-361 (hereinafter D6). In reply thereto, the appellants submitted, on 18 November 1997, two sets of amended claims replacing the previous requests.
- V. Oral proceedings were held on 20 November 1997. At the hearing the board informed the appellants that it was questionable whether the amendments of the claims according to the two requests filed on 18 November 1997 were suitable for overcoming the objections raised by the board in respect of Article 84 EPC. In reply, the appellants pointed out in particular that the present invention concerned a process for preparing a fluid cracking catalyst, which process necessarily involved the use of the four catalyst components stated in the description, namely an acid aluminium sulphate silica sol, a clay slurry, a zeolite slurry and an alumina slurry. The appellants submitted a set of eleven claims amended accordingly, as sole request. Claim 1 of the set of amended claims filed at the oral proceedings reads as follows:

"1. A process for preparing a zeolite-containing and sol-based calcinated fluid cracking catalyst with improved physical and catalytic properties, which process comprises adding, prior to drying, at least 0.0198 percent (one pound per 2.5 tons), with respect to the weight of the final catalyst, of an acid stable anionic surfactant to at least one of the catalyst components or to a mixture of said catalyst components, wherein the catalyst components, prior to drying, are an acid aluminum sulfate silica sol, a clay slurry, a zeolite slurry, and an alumina slurry, and drying the combined components."

VI. The appellants argued inter alia that they could not find a single reference suggesting that formic acid was a surfactant or that the function of "peptizing" alumina was equivalent to the function of a surfactant. The definition given in D2, when read in its entirety, did not suggest that formic acid was a surfactant. The alternative definition in the second phrase of D2 had to be read in conjunction with the first definition, otherwise all the compounds of D3 that reduced the surface tension of water would be surfactants, which would be inconsistent with other scientific references. The additional citations demonstrated that formic acid failed to function as a surfactant. It was not a surfactant because, on the one hand, it was too soluble in water and did not concentrate at the surface of a liquid, and, on the other hand, it did not have a hydrophobic portion with a long hydrocarbon chain of 10 or more carbon atoms and, therefore could not lower the interfacial tension.

As regards clarity of the expression "acid stable anionic surfactant", the appellants argued that this expression was an accepted term in the industry and had a recognized meaning in the field of surfactants. This expression had been accepted by the US and Australian

patent offices and was commonly used in the industry's promotional literature, as shown by the cited commercial brochures. The terms "acid stable" characterised the relative chemical stability of the surfactant. The latter had to be stable in the acidic medium in which it was used. The acidic composition, pH, residence time, and temperature of the catalyst components were known by the catalyst's producer. The latter was, thus, able to select a commercially available acid stable anionic surfactant suitable for performing the invention on the basis of the available commercial brochures.

- VII. The appellants requested that the decision under appeal be set aside and that the case be remitted to the first instance for further prosecution on the basis of the amended set of claims filed at the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.
2. The amended claims 1 to 11 filed at the oral proceedings meet the requirements of Article 123(2) EPC. Claim 1 is based on the combination of original claim 1 with features disclosed in original claims 1 and 3 and in the description as filed: see in particular page 4, last paragraph, and page 6, last paragraph, in connection with the lower amount of surfactant and the use of an anionic surfactant respectively. The addition of the acid stable surfactant to at least one of the four components recited in claim 1 or to a mixture thereof is disclosed in the paragraph bridging pages 9 and 10 of the original description read in connection with page 9 (2nd to 5th paragraphs). The use of a kaolin-type clay

slurry according to claim 2 is supported by the statement on page 9, lines 13-14, of the description. Claims 3 to 11 correspond to claims 3 and 5 to 12 as originally filed.

3. *Clarity and support: Article 84 EPC*

3.1 Regarding the expression "acid stable" anionic surfactants, it should be noted that the description does contain information about the pH and the temperature of the acidic components and of the acidic component mixture to which the surfactant is added. In particular, the pH values of the acid alum silica sol (pH of about 2.8 to 3.5) are indicated on page 9 of the description and the examples disclose the pH values and temperatures of the said sol, as well as the pH values and temperatures of the component mixture to be fed to the spray-dryer (pH of from 3.12 to 3.33). The pH of the zeolite slurry is also mentioned on page 9. In the board's judgment, in view of the information given in the description, in particular as to the pH and the temperature of the acidic alum silica sol and of the acidic medium resulting from the mixture of the catalyst components, the skilled person can readily understand what is meant by "acid stable" anionic surfactant. The board agrees with the appellants' submissions at the oral proceedings that it is clear to the skilled person that the surfactant must be stable in the acidic medium resulting from the mixture of the catalyst components, in particular under the preferred conditions of pH and temperatures indicated in the description. Furthermore, in the case where the surfactant is added to an acidic catalyst component, in particular to the acid aluminium sulfate silica sol, the anionic surfactant must also be stable to this acidic sol at the preferred pH and temperatures indicated in the description. Therefore, the expression

"acid stable anionic surfactant" used in connection with the process of preparation of the fluid cracking catalyst as defined in claim 1 is considered to meet the requirement of clarity.

3.2 According to the description, the preferred acid stable anionic surfactants are the fluorohydrocarbon surfactants, and the examples are accordingly performed with two acid stable anionic fluorohydrocarbon surfactants, namely those designated Zonyl TBS and FC-95 by the Dupont Company and the 3M Company respectively. Although it is indicated in the description that any acid stable surfactant will give satisfactory results, the description does not mention any example of acid stable anionic surfactant other than the said anionic fluorohydrocarbon surfactants. In these circumstances, the question arises whether claim 1, which embraces the addition of acid stable anionic surfactants other than the said fluorohydrocarbon surfactants, meets the requirement of support by the description set out in Article 84 EPC. Therefore, the question has to be examined whether or not the skilled person would be able to find out, without undue burden, which anionic surfactants other than the fluorohydrocarbon surfactants are stable in the acidic medium used in the claimed process (see in this respect T 409/91, OJ EPO 1994, 653).

D6, which illustrates the common general knowledge in the field of anionic surfactants before the priority date, discloses the different groups of known anionic surfactants as well as numerous commercial surfactants with their trade names and their manufacturers (see pages 347-361). Acid stability or alkaline stability of these surfactants is also dealt with in this document. It is indicated at page 349 in connection with surfactants containing a sulfonate group attached to an alkyl, aryl, or alkylaryl hydrophobe that the salts of

sulfonic acid "are relatively unaffected by pH" and are stable to oxidation and hydrolysis. According to page 355, 3rd paragraph, the surfactant Dowfax 2A1, a sulfonate with ether linkages, is characterised "by high tolerance for strong acids and bases". Concerning the surfactants with a sulfate group, it is disclosed on the same page (4th paragraph) that "attachment of the sulfate group to a carbon group of the hydrophobe through the C-O-S linkage limits hydrolytic stability, particularly under acidic conditions". According to the last paragraph of page 355, "sensitivity to hydrolysis in hot alkaline and acidic media is one of the disadvantages of alcohol sulfates". On page 357, it is stated in connection with sulfated alcohol ethoxylates that "at very high alkalinity and in acid media, the sulfate linkage is susceptible to hydrolysis, particularly at high temperatures". Phosphate esters are dealt with on page 359 where it is stated that "in contrast to sulfonates and sulfates, the resistance of alkyl phosphate esters to acids and hard water is poor" (see penultimate paragraph). D6 does not mention specific acids, or ranges of concentrations or pH values; however, the board is satisfied that, in view of this teaching, the skilled person would be able to determine without undue burden, by some routine experimentation, which of the anionic surfactants known from D6 as being stable in acidic conditions are stable in the acidic medium involved in the present process, in particular at the pH and temperatures values indicated in the description. Therefore, the board considers that, on the basis of the information given in the description and of the common general knowledge in the field of surfactants, the skilled person would be able to perform the claimed process without undue burden also in the case of acid stable anionic surfactants other than the fluorohydrocarbon surfactants.

It follows from the above that claim 1 meets the requirements of Article 84 EPC.

4. Turning to the issue of novelty, the essential reason given by the examining division for lack of novelty over D1 was that formic acid was an acid stable anionic surfactant, taking into account the definition given in D2 for the term "surface-active agent". Although the subject-matter of claim 1 filed at the oral proceedings might be novel over the process of D1 even if formic acid were an acid stable anionic surfactant, the board deems it appropriate to examine the question whether or not formic acid is a surfactant in view of its importance for the still outstanding examination of inventive step.

- 4.1 D2 does indeed disclose that a surface-active agent (surfactant) is "any compound that reduces surface tension when dissolved in water or water solutions". However, this definition is not the one commonly used in the art. This is in particular apparent from "Römpps Chemie-Lexikon", 7th edition, 1981, vol. 2, page 1546, where it is pointed out under the heading "Grenzflächenaktive Stoffe" that although polar solvents like alcohols, ethers, etc. are surface-active, the term surface-active agent is in general reserved for those compounds containing a lipophilic and a hydrophilic group. The said definition in D2 is obviously broader than the commonly accepted definitions which are indicated in other chemical dictionaries or encyclopedias, and in standard textbooks or handbooks relating to surfactants and illustrating the common general knowledge in this field before the priority date. According to these documents, a surfactant is a compound which, when dissolved in a liquid, reduces the surface tension or interfacial tension by preferred adsorption, ie by concentrating, at the liquid/vapour surface or other interfaces, and

the molecule of a surfactant contains groups of opposing solubility tendencies, ie at least one hydrophilic (lipophobic) group and at least one lipophilic (hydrophobic) group. Owing to this amphiphilic character, the concentration of a surfactant solute at a phase interface is greater than its concentration in the bulk of the solution: see, for example, Exhibit 4, page 319, left-hand column; Encyclopedia of Chemical Technology, 3rd edition, vol. 22, 1984, pages 332 to 333; Tensid-Taschenbuch, 2nd edition, 1981, pages 2 to 3; Surfactants and Interfacial Phenomena, Milton J. Rosen, 1978, pages 1 to 3; Römpps Chemie-Lexikon, 8th edition, 1988, vol. 6, pages 4153 to 4155. Taking into account that this definition based on the essential structural characteristics and behaviour of surfactants is consistently disclosed in several documents, in particular in the literature concerning surfactants, and corresponds to what the skilled person normally understands under the term "surfactant", the board considers that it also applies to the present case. This definition clearly rules out formic acid being considered as a surfactant or as functioning as a surfactant. Formic acid indeed contains a hydrophilic group -COO^- , however it does not contain both a hydrophilic and a hydrophobic group and, as pointed out by the appellants, it does not concentrate at the surface in an aqueous solution.

- 4.2 D1 discloses a process for preparing a fluid cracking catalyst having improved attrition resistance, wherein colloidal silica is mixed with a catalyst ingredient mixture comprising a zeolite and a kaolin clay, and with sufficient water to form a first slurry, which is mixed with an acid dispersed alumina to form a second slurry. The latter is mixed with sufficient water to form a sprayable formulation, which is spray-dried and the spray-dried particles are then calcined. Formic

acid is used for dispersing the alumina. The colloidal silica is preferably selected from the group consisting of Nyacol 9950, Nyacol 2050, Nyacol 2034 DI, Nyacol 2040 NH4 and Nyacol 830 (see claims 1, 3 and 7, pages 20-23; sentence bridging pages 25 and 26; examples).

As formic acid cannot be considered as a surfactant and D1 does not disclose the addition of an acid stable anionic surfactant to at least one of the catalyst components or to their mixture before spray-drying, the process according to claim 1 differs from the process of D1 at least by this feature. Therefore, the subject-matter of claim 1 is new over the disclosure of D1.

4.3 Novelty of the claimed process with respect to the other documents cited in the search report was not disputed by the examining division. Furthermore, additional features have been incorporated into the amended claim 1 filed at the oral proceedings compared to claim 1 of 14 July 1992. The board has also examined the said documents and has come to the conclusion that the process according to claim 1 also meets the requirement of novelty with regard thereto.

5. Taking into account, on the one hand, that inventive step of the amended claim 1 containing the present combination of features has not been examined by the examining division, and, on the other hand, that additional examples have been submitted at the appeal stage, the board finds it appropriate to remit the present case to the first instance for further prosecution.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance for further prosecution on the basis of the amended set of claims filed at the oral proceedings.

The Registrar:


E. Gorgmaier

The Chairman:


R. Spangenberg

REB

JM

