BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

A		B		C	X

File Number: T 101/91 - 3.4.2

Application No.: 83 112 693.3

Publication No.: 0 111 911

Title of invention: Removal of carbon dioxide from olefin containing streams

Classification: B01D 53/02, C07C 7/12, B01J 20/04

D E C I S I O N
of 1 December 1992

Applicant: Phillips Petroleum Company

Opponent: Rhone-Poulenc Chimie

Headword:

EPC Article 52(1), 56

Keyword: "Inventive step - yes"

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number : T 101/91 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 1 December 1992

Appellant : (Proprietor of the patent)
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Decision under appeal :
Decision of the Opposition Division of the European Patent Office dated 20 September 1990, posted on 7 November 1990 **revoking European patent No.** 0 111 911 **pursuant to Article** 102(1) **EPC.**

Composition of the Board :

Chairman : E. Turrini
Members : C. Black
L.C. Mancini

Summary of Facts and Submissions

I. European patent No. 0 111 911 (application No. 83 112 693.3) was opposed by the present Respondent on the ground that its subject-matter was not novel or did not involve an inventive step having regard to the disclosure in the following documents:

D1: US-A-3 943 226
D2: Journal of catalysis, 67, 1981, pages 362 to 370 "Alkali-promoted alumina catalysts",
D3: Kinetika i Kataliz, Vol. 10, No. 5, 1969, pages 1119 to 1124 and the English translation,
D4: FR-A-2 145 688.

II. The patent was revoked by decision of the Opposition Division. Its decision was based on independent Claim 1 reading as follows:

"A process for removing low concentrations of CO_2 from a gaseous olefin stream containing at least one C_2 to C_4 olefin characterized by passing said gaseous olefin stream containing CO_2 into contact with a regenerable, calcined adsorbent, said regenerable, calcined adsorbent having been made by (a) contacting alumina with an alkali metal compound in an amount of 1-6 weight percent on the adsorbent, calculated as alkali metal oxide, said alkali metal compound being selected from sodium, potassium and lithium compounds which are convertible to the metal oxide on calcination, (b) drying the resulting alkali metal compound-treated alumina, and (c) thereafter calcining the dried treated alumina at a temperature from 200°C to 400°C."

The gist of the Opposition Division's argumentation is that it was known from D4 to remove low concentration of

CO_2 from gaseous olefin streams using alumina, that the particular alumina adsorbent known from D2 did not differ materially from that required by Claim 1 and that it was obvious to combine the teachings of D2 and D4 in this respect and thus arrive at the subject-matter of Claim 1.

III. The appeal lies against this decision. In the course of the appeal proceedings, the Appellant introduced the further documents:

D5: Physical Chemistry (Gucker and Seifert) 1966, pages 652, 653

D6: CRC Handbook of Chemistry and Physics, 67th Edition, 1986-87, pages F63, F64 (not a prior art document.

At the end of oral proceedings, the Appellant (Patentee) requested that the decision under appeal be set aside and a patent granted on the basis of the claims and description handed over at the oral proceedings, of which Claim 1 is as set out in paragraph II above and Claims 2 to 6 are appendant to Claim 1.

The Respondent requested that the appeal be dismissed.

IV. The gist of the Appellant's argumentation is as follows:

D2 does not disclose a purification process; it concerns an activated alumina catalyst and investigates the adsorption of CO and CO_2 on the catalyst. D2 moreover discloses the adsorption of CO_2 present in relatively high concentration from a mixture with Argon, not the low concentration (of the order of ppm) according to the patent in suit. Further said adsorption of CO_2 from a mixture with argon cannot be compared with the purification of C_2 - C_4 olefins as in the patent in suit, because argon does not compete with binding sites on the

adsorbent as do the olefin molecules. In any case the activated alumina disclosed in D2 is different from that required by Claim 1 because it has been prepared using a calcining step at 620°C, not 200 to 400°C as in Claim 1. That the two are different is confirmed by the experimental data submitted with the Affidavit of Dr. M. Bruce Welch accompanying the grounds of appeal. In addition according to D2 no potassium oxides were found on the surface of the calcined catalyst; in the adsorbent according to the patent in suit alkali metal oxide can be detected spectroscopically at the surface. Experiments of the Respondent purporting to disprove this have not necessarily used the same conditions or measuring techniques. Accordingly it is only with hindsight that the subject-matter of Claim 1 can be arrived at from the disclosure in D2.

D1 relates to the removal of strongly acidic impurities from naphtha and D3 to the adsorption of CO on alumina and alkali-treated alumina. These documents are therefore not relevant.

D4 should be considered as the nearest prior art. It discloses the purification of propylene using alumina as adsorbent but not the particular alkali metal treated alumina required by Claim 1. Impurities listed, which may be present in small amounts, include oxygen, water, sulphur, CO, CO_2, organic and inorganic compounds. There is no specific reference to CO_2 removal; the tenor of the disclosure is the removal of organic compounds by a reactive process, as evidenced by the appearance of a brownish coating on the adsorbent apparently ascribable to oligomers of the organic compounds. D4 moreover teaches purification in the liquid phase, a comparative example demonstrating that gas phase purification is less efficient. D4 therefore does not disclose or suggest the

subject-matter of the claim, nor is there any reason for the skilled person to combine its teaching with that of D2. The adsorption of gases on solid adsorbents depends to a great extent on their critical temperatures (D5). Whereas the critical temperatures of argon and CO_2 are very different those of ethylene and CO_2 are close to each other (D6). Therefore CO_2 should separate easily from argon and if the adsorption capacity of CO_2 is improved, that of argon will be little affected. However it is to be expected that a modification of alumina will affect the adsorption of CO_2 and ethylene in a comparable manner, and the fact that D2 discloses that treatment of alumina with potassium improves the adsorption of CO_2 from argon would not suggest to the skilled person the application of the treated alumina to separate CO_2 from olefins. In any case D4 discloses a treatment for impurities down to less that 2 ppm and the skilled person is not prompted to look for improved adsorption capacity.

The Respondent's counter-arguments may be summarised as follows:

From D4 it is known to purify propylene using a mineral adsorbent, in particular alumina. CO_2 is mentioned as an impurity to be removed. According to D4 loading capacity is increased for the same degree of purification by treatment in the liquid phase as compared with the gaseous phase (as in comparative example). Therefore D4 can be read as disclosing the removal of CO_2 in low concentration (down to less than 2 ppm) from propylene in the gaseous phase using alumina as adsorbent, though not the activated alumina required by Claim 1.

Since the capacity of an adsorbent to adsorb a gas depends on the partial pressure of the latter and is therefore very low at low concentrations, the problem to be solved

can be seen as increasing the capacity of the adsorbent for the said gas.

D2 provides a solution in that it discloses that by treating alumina with a potassium compound in the manner required by Claim 1, the adsorption capacity for alumina is increased. The calcining temperatures of the two adsorbents are indeed different, but the patent in suit does not show any advantage of the required calcination temperature range (200°-400°C) over calcination temperatures outside this range. The Appellant's contention that the adsorbent required by Claim 1 differs from that of D2 in having alkali metal oxides at its surface is not valid. The Respondent prepared an adsorbent according to the patent in suit and this did not show the said oxides. In any case D2 merely says that potassium oxide was not detected, not that it was not present. Further the experimental results accompanying the Affidavit compare the adsorbent according to the patent with that of D2 in terms of parameters which are not disclosed in the patent, let alone considered to be significant. As regards the relation between adsorption and critical temperature, this is not so clear cut as the Appellant makes out, as can be seen by inspecting Table 22.2 of D5, for example the values for oxygen and hydrogen.

Reasons for the Decision

1. The appeal is admissible.

2. As compared with Claim 1 as originally filed, Claim 1 now under consideration has been restricted by incorporating the features of original Claim 8 (preferred calcination

temperature of 200°-400°C), page 1, lines 8 to 11 (low concentrations of CO_2), and page 2, lines 29, 30 (1 to 6 weight percent of alkali metal compound). The further additional feature of drying before calcination is implicit in the description and is disclosed in Example I. The claim has also been restricted compared with that of the granted patent, and the description amended only to conform to the claim, so that the requirements of Article 123(2) and (3) EPC are met.

3. Novelty of the claimed subject-matter is not an issue in dispute between the parties, and will in any case be apparent from the discussion of inventive step.

4. For the investigation of inventive step, D4 is clearly the most appropriate prior art, because it discloses a process for removing low concentrations of impurities from olefins using mineral adsorbents, e.g. alumina. D1, relating to the removal of strongly acidic impurities, e.g. hydrogen halides, from naphtha hydrocarbons is more remote from the subject-matter of Claim 1 and can be left out of consideration. D2 and D3 relate to studies of the adsorption of carbon monoxide and carbon dioxide on alumina rather than to purification processes. Of these D3, relating only to the adsorption of carbon monoxide, is less pertinent and can also be left out of consideration. For completeness, reference is made to D5 and D6 cited by the Appellant wherein D6 is not state of the art within the meaning of Article 54(2) EPC. These are excerpts from standard reference books and their late introduction into the proceedings is accepted.

5. The introduction to D4 indicates that it is known to dry gases using e.g. alumina, and also to remove organic impurities from C_2-C_4 olefins using mineral adsorbents. D4 goes on to refer to prior art in which, using this

procedure for ethylene, a pressure of 35 kg/cm^2 was necessary to achieve the unsatisfactory purification capacity of less than 0.5 kg olefin per litre adsorbent. Working at more convenient lower pressures gave even worse results. To overcome this disadvantage D4 proposes carrying out the purification, specifically of propylene, in the liquid state. Impurities stated to be present in the propylene to be purified are oxygen, water, carbon monoxide, carbon dioxide, acetylene, propadiene, sulphur and methyl acetylene (page 2, lines 13,14 and page 3, lines 32,33), in amounts of less than 10 ppm, even less that 2 ppm (page 2, line 15) possibly even less that 1 ppm (page 3, line 32). Of these only the unsaturated compounds are specifically mentioned as regards impurities actually removed, in that a brownish colour developing on the adsorbent is attributed to oligomerisation of the said compounds. In a comparative example in which the purification is carried out in the gaseous phase as in the prior art process, operating at the same specific purification capacity as in the liquid phase purification resulted in a less pure product as measured by the ash content of the polymerised purified propylene. Only by operating at a lower throughput could the same purification be achieved.

6. Since D4 already discloses a process for removing low concentrations of impurities from C_2-C_4 olefins, the problem underlying the patent in suit is accordingly not that stated on page 2, lines 16,17, but rather that of improving the process disclosed in the comparative example in a different manner from that claimed in D4. The question to be answered therefore is whether it is obvious for the average skilled person to take the steps necessary to bridge the gap between the disclosure in D4 and the subject-matter of Claim 1.

7. In the first place it is by no means certain that the disclosure in D4 would set him off on the path leading to the subject-matter of Claim 1, because D4 only mentions carbon dioxide incidentally as an impurity present in the olefin to be purified, and gives no clear indication that the disclosed process is intended to remove it, or that it is in fact removed. Moreover in view of the stated disadvantages of the gaseous phase process (D4: page 1, lines 11 to 20), which are overcome by operating in the liquid phase, there is no incentive for him to abandon the liquid phase process and revert to the less advantageous one, and it is more likely that he would attempt to modify the liquid phase process.

Nevertheless, assuming in favour of the Respondent that he would seek to improve on the gaseous phase process and apply it to the removal of small quantities of carbon dioxide from an olefin gas stream, he still has, in order to come to the subject-matter of Claim 1, at least to use the particular alumina adsorbent required by Claim 1. It is true that D2 discloses an alumina catalyst which has been treated with an alkali metal (potassium) compound in a manner very similar to the adsorbent of the patent in suit, as a result of which the adsorption of carbon dioxide is increased (page 365, left column, lines 10 to 13). However, as argued by the Appellant, and in contrast to the view of the Opposition Division (page 4 of the decision), D2 does not disclose a purification reaction, but a study on chemisorption and oxygen exchange of carbon monoxide and carbon dioxide on potassium-promoted alumina catalysts. Further in the disclosed experimental methods, the adsorption of carbon dioxide present in relatively high concentration in argon (partial pressures of 3 Torr and 10 Torr respectively, whereby 1 Torr = 133,3 Pa) is discussed, which gives no clear incentive to use the said

catalyst to remove very small amounts of carbon dioxide from olefins. Without going into the Appellant's argument concerning competition for binding sites on the adsorbent, it is noted that the description in the patent in suit (page 4, lines 63 to 65 and Table I) provides evidence that the nature of the adsorption of CO_2 from ethylene cannot be predicted on the basis of its adsorption from an inert gas. Moreover the adsorbent required by Claim 1 differs from that disclosed in D2 in that in its preparation a calcination temperature of 200 to 400°C is used as opposed to 620°C. That the adsorbent as a result is different is shown by the results of calcining experiments contained in the affidavit accompanying the grounds of appeal, even though the differences are indicated in terms of parameters not disclosed in the patent in suit.

The Respondent and the Opposition Division have argued that the patent in suit contains no data demonstrating the advantage of calcination at 200 to 400°C as compared with lower or higher temperatures. However in the application as originally filed, the range of 200 to 400°C was stated to be a preferred range within a broader range of 100 to 700°C and the Board accepts that this preference relates to the result sought to be achieved.

Accordingly even if the average skilled person started from the disclosure in D4, he would not be led to combine its teaching with that of D2, and even if he did, he would not arrive at the subject-matter of Claim 1.

8. In view of the foregoing it is not necessary to go into the question of a difference between the adsorbent required by Claim 1 and the catalyst disclosed in D2 based on the presence or absence of alkali metal oxide at the surface of the alumina.

9. The subject-matter of Claim 1 is therefore seen as involving an inventive step and, Claims 2 to 6 being dependent on Claim 1, are allowable for the same reason. The requirements of Article 52(1) and 56 EPC are accordingly met.

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 with the order to grant a patent on the basis of the documents filed at oral proceedings.

The Registrar: The Chairman:

P. Martorana E. Turrini