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12

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Bezeichnung der Erfindung: Process for improving the properties of a
Title of invention: cracking catalyst
Titre de l'invention :

Klassifikation / Classification / Classement : C 10 G 11/02

ENTSCHEIDUNG / DECISION

vom / of / du 28 July 1987

Anmelder / Applicant / Demandeur : PHILLIPS PETROLEUM COMPANY

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence : "Passivation of catalyst"

EPO / EPC / CBE Article 56 EPC

Kennwort / Keyword / Mot clé : "Inventive step (no) - Effect predictably worsened"
"Inventive step (no) - Intermediary effects"
"Technical problem - Redefinition - Unallowability of reversal of desired effect"

Leitsatz / Headnote / Sommaire

- I. It is not acceptable to rely on an effect which has previously been described as undesirable and of no value by the Applicant, to present the same suddenly as possibly representing an advantage from another point of view, and thereby to imply that the technical problem and the considerations for the inventive step should take this reversal into account. While a redefinition of the technical problem is normally permissible and even necessary on the basis of the comparison of achievements with the closest prior art, this should not contradict earlier statements in the application about the general purpose and character of the invention.
- II. Subject-matter falling structurally between two particular embodiments of cited disclosure and displaying, in all relevant respects, effects substantially between those known for the same embodiments, lacks inventive step in the absence of other considerations.

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Chambres de recours



Case Number : T 155/85

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 28 July 1987

Appellant : Phillips Petroleum Company
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Decision under appeal : Decision of Examining Division 029
of the European Patent Office
dated 22.01.85 refusing European
patent application No. 81 102 182.3
pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : P. Lançon

Members : G. Szabo

F. Benussi

Summary of Facts and Submissions

I. European patent application 81 102 182.3 filed on 23 March 1981 and published on 30 September 1981 with publication number 36 661, claiming priority of the prior application on 24 March 1980 was refused by the decision of the Examining Division of the European Patent Office dated 22 January 1985. The decision was based on Claims 1 to 14 filed on 2 May 1983. The main claim was worded as follows:

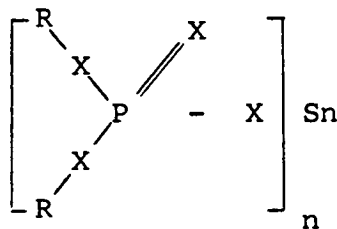
"A process for catalytically cracking hydrocarbons in the absence of antimony utilizing a passivating agent for the passivation of catalyst-contaminating metals, characterized by using a passivating agent containing tin, phosphorus and sulphur".

II. The stated ground for refusal was that the subject-matter of Claim 1 did not involve an inventive step. The cited closest prior art EP-A-1642 (1) described a process using a passivating agent containing tin, phosphorus and sulphur but the use of a combination of these three in the absence of antimony was not specifically disclosed in the Examples. It was also indicated in comparative tests that agents containing tin and sulphur were effective, without antimony and phosphorus. The preferred compounds for tin were stannic 0,0,-dipropyl-dithio-phosphate (identical with di-n-propyl-phosphodithionate of the specification, hereinafter DPPD), and dibutyl tin bis(isooctylmercaptoacetate (hereinafter DBMA). Dibutyltin oxide (DBO) on its own was not the most preferred compound and it was therefore not inventive to choose any of these other agents, in particular DPPA, for the same purpose.

III. The Applicant filed an appeal against the decision on 28 February 1985 with the payment of the fee and submitted a Statement of Grounds on 3 June 1985. New Claims 1 to 13

were also presented at the same time and corrected on 19 November 1985 and 19 December 1985. The new main claim reads as follows:

"A process for increasing the selectivity for gasoline production of a cracking catalyst utilized for catalytically cracking hydrocarbons in the absence of antimony and in the presence of a passivating agent for the passivation of catalyst-contaminating metals, characterized by using a said passivating agent a tin compound of the general formula



wherein each R is the same or different and denotes a hydrocarbyl group having from 1 to 24 carbon atoms, each X is individually oxygen or sulfur wherein at least one X is sulfur, and wherein n is 2 or 4."

IV. An oral hearing was held on 27 July 1987. The Appellant submitted during the hearing an auxiliary set of claims further limited to the use of DPPD as a passivating agent.

V. The Appellant argued in his submissions and at the oral hearing substantially as follows:

(a) It was important to recognise that the teaching of the cited earlier specification was essentially the discovery of a synergistic effect of the combination of tin with antimony, i.e. a superiority over using either of these metals separately for passivation. There was therefore no good reason to move towards the

use of tin alone. Gasoline yields were consistently better with the combination product than with tin on its own according to the prior art (cf. Fig. 9 of the citation). There was, however, no reason to assume that DPPD in particular would give better results than DBO.

- (b) Any stated preference for DPPD in the cited art (cf. page 11, top paragraph) might be understood in this context, i.e. in respect of compatibility with the agent containing antimony. The closest individual test with tin alone was run 3 in Table VI of (1), using 0.1% DBO. In comparison with the results obtained there, the invention using the same concentration of DPPD achieved gasoline yields increased by 3.5% (cf. also EP-36 661, page 33, run 2 in Table XV and run 2 in Table XVI). This was unexpected since the use of DBO in the citation must be interpreted as the preferred manner of using a tin-containing passivating agent without antimony. Although examples in (1) (Table VIII, page 40, runs C) also use DBMA, this is structurally pointing away in view of the acetate moiety.
- (c) Although great emphasis was laid in earlier submissions on the decisive character of selectivity, the gasoline yield was ultimately most important for the plant engineer. He would, of course, duly consider all aspects of the result and assess the value of the achievement accordingly. Whilst hydrogen and coke formation was normally undesirable in cracking, acceptable levels might include even somewhat increased hydrogen formation if this was coupled with good gasoline yields. After all, hydrogen might be utilised in another plant.
- (d) As to the significance of comparisons, the runs tabulated in (1), Table VIII, pages 40-41 were

individual runs with natural fluctuations, whilst the figures in the application Table XV and XVI on page 33 were the average of many experiments (the same applying to Table VI in (1)). The shown improvements over the prior art were therefore significant and relevant.

- V. The Appellant requests that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 13 filed on 3 June 1985, and corrected by a letter received on 19 December 1985, or as an auxiliary request on the basis of claims submitted at the oral proceedings.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.
2. No formal objections can be raised against the amended claims. Claim 1 in the main request is properly based on the combination of the original main claim with Claim 7 in the same set, whilst the auxiliary set is further limited to the use of DPPD, which was the subject of original Claim 10. The amendment complies with Article 123(2) EPC.
3. The claimed subject-matter relates to the passivation of catalysts for cracking hydrocarbons, and in particular to the use of agents which contain tin in an organic compound having also phosphorus and sulphur atoms. Such kinds of passivating agents were known from (1) which disclosed the incorporation of the same atoms in a wide range of agents, preferably in combination with similar antimony containing

agents for the same purpose. According to the general description, a great variety of sources for these two metals can be used for passivation but those which are soluble in hydrocarbons are naturally preferred and in particular those which contain oxygen, sulphur, nitrogen or phosphorus in the organic moiety (cf. page 10, line 9). For both metals the appropriate stannic bis or antimony tris (0,0-dipropyl phosphorodithioate) compounds are preferred, i.e. DPPD and TPPD (page 9, line 12), respectively, with DBMA as an alternative top preference for tin.

4. The specific disclosure in (1) includes many comparative tests with tin alone as DBO or DBMA, with antimony alone as TPPD, or the two in combination. The purpose was to demonstrate the superiority of the combination in increasing direct gasoline output and to decrease the formation of hydrogen and coke. The results show that at the best, with a total amount of 0.1% antimony and 0.01% tin, the gasoline yield topped 61% (with a 69.8% conversion, 87.6% selectivity, 385 units of hydrogen and 7.2% coke). Against that stands the demonstrated best result with about the same total amount of passivator in the form of 0.1% DBO, i.e. tin alone, showing a yield of only 54.1% (with 61.4% conversion but 88.6% selectivity, and 578 units of hydrogen and 7.2% coke (cf. averages of many runs in Table VI, page 33). The apparent drop in what might be called a "first crop" gasoline yield is somewhat less dramatic in real terms of an ultimate total yield if it is taken into consideration that a lower conversion with tin allows, together with better selectivity, relatively greater additional crops on recycling. This is in view of the increased unconverted material which is available, the ultimate inevitable loss being consequently less (cf. arguments of Appellants in their Statement of Grounds, page 2). This illustrates the importance of not considering yield or selectivity on its own.

5. The technical problem in respect of this state of the art was to provide good gasoline yields, preferably without unacceptably high amounts of hydrogen and coke. The claimed solution of the problem involves tin alone as a passivating agent, in the form of DPPD or analogue compounds. It appears from the tabulated results in the specification (cf. run 2, Table XV, page 33) that DPPD provides a 57.6% gasoline yield, with 64.7% conversion, 89% selectivity, and 502 units of H₂ and 7.7% coke. It appears that the results fall between those achieved by the cited art with an antimony/tin mixture, and tin alone, except of the slightly increased selectivity (0.4%) and worse coke production than before. The claimed subject-matter has therefore achieved the main goal, the effect required by the stated technical problem, except in respect of the additional aim to reduce coke production. In view of the higher conversion rate, the catching up of the ultimate yield with the top yield of the prior art will not be as fast as with DBO, but it is clear that the efficacy of the most preferred process for producing gasoline in the present application falls between the relevant two best results in the cited state of the art.
6. Although the preferred agent DPPD in the present application was expressly mentioned in (1), it was not specifically exemplified in comparative tests. Such use can therefore be considered as novel.
7. As to the inventive step, it is very relevant in the present case that the cited disclosure represents a range of possibilities beyond what is claimed in document (1) as the invention. Indeed, the quantitative comparisons with results representing variations in parameters allow interpolations of situations which are not specifically tested but fall in respect of their conditions within the

ranges presented. This is possible since there are recognisable trends within the results which are beyond the fluctuations of individual results and can be seen in tabulated averages and the best fit curves shown in the drawings.

8. Although the results with DBO are most relevant for comparisons because the concentration of the passivator is identical with that shown in Example XII, Table XV, run 2 of the present application, this cannot be taken as necessarily the best use of tin alone on the basis of (1) as a total disclosure. The exemplification with DBO cannot therefore mean that other agents for tin would have been expected to perform even worse under similar conditions. It is manifest that DBMA has also been tested (cf. Table VIII). Although the individual results with a very low concentration (0.011%) are lagging behind those with DBO at a ten times higher concentration, the selectivity figures were already outstanding in some individual results (cf. about 91% and 89% (calculated) in 5C and 6C). It is more relevant though that this agent overcomes the so-called "negative contribution" of DBO in comparison with blank tests i.e. the apparent drop in conversion and yield, and if one also takes into consideration that the test conditions behind Table VIII were substantially below the optimum of 0.1% total metal content or more, also suggested by Table VI, then the possibility of obtaining even better results with DBMA than with DBO could not have been dismissed.
9. The expected superiority of DBMA over DBO was later on clearly confirmed in the present specification which demonstrated that at a comparable catalyst/oil ratio, DBMA was capable of achieving even at that low 0.011%

concentration a 67% yield at the level of 83.8% selectivity (cf. Table XIII, Run 6). It was, however, already conceivable from (1) that agents like DBMA or even more DPPA, which retained at least the sulphur atom, or both the phosphorus and the sulphur atoms, would represent conditions in between tin alone, without such additional atoms, and tin and antimony with both of such atoms. In view of the declared express preference for DPPD in addition to DBMA, it would have been obvious to expect the possibility of an improvement with the agent over DBO. The outcome of such interpolation was, as expected, essentially in between those with DBO, and DBO with TPPD.

10. The argument that at least the 89% selectivity (Table XV, Run 2) was better than the best result in this respect with the two extremes, cannot change the impression. The marginal improvement (0.4%) in this respect would not quite compensate for the loss of direct yield, since the increased necessity for recirculation is an additional, undesirable burden. The particular figure of 89% was not an outstanding maximum either, as individually better results with DBMA were already indicated in the cited art, i.e. 89.2 and 90.5% (Table VIII, runs 5C and 6C).

The further worsening of the formation of coke is clearly also contrary to what the plant engineer would consider a satisfactory alternative. Whilst the figures are based on an average of a number of runs, the statistical significance of 0.4% increase of selectivity is still unknown (cf. for instance the different averages of apparently identical sets of experiments in (1), Table VI, runs 12 and 13). It is correct to say that even small improvements in yield or other industrial characteristics could mean a very relevant improvement in large scale production (cf. T 38/84, "Oxidation of toluene", OJ 8/1984, 368) but the improvement must be significant and therefore

above margins of error and normal fluctuations in the field in consequence of other parameters. This is not the case here.

11. It is acceptable to recognise a less ambitious effect than what might have been promised by the applicant, if this is what is in fact achieved by the invention (cf. T 184/82, "Poly(p-methylstyrene articles", OJ 6/1984, 261, at 264 and Headnote). Thus the attainment of good gasoline yields without an unacceptable amount of hydrogen and coke by-products, has turned out to be a realistic goal in the light of document (1). An absence of upsetting the balance of inevitable by-products, as the Appellant suggested, was therefore the real complement of the basic requirement for good gasoline output, and not any longer the originally suggested "dramatic" increase of selectivity "in addition to decreasing hydrogen and coke production" (cf. page 3, lines 14-19 of the application.

12. It is, however, not acceptable in the view of the Board, to rely on an effect which has previously been described as undesirable and of no value by the Applicant, to present the same suddenly as possibly representing an advantage from some other point of view and thereby to imply that the technical problem and the considerations for the inventive step should take this reversal into account. Whilst a redefinition of the technical problem in respect of a particular state of the art is normally permissible and even necessary on the basis of the comparison of achievements with the closest prior art this should not contradict earlier statements in the application about the general purpose and character of the invention. The argument that an increased production of hydrogen, as a by-product, could be something desirable in a manufacturing unit using hydrogen for other purposes, must therefore be

dismissed either as contrary to the original disclosure and contributing no worthwhile effect in the given context, if exceeding levels of acceptability, or as irrelevant, if within limits of toleration.

13. The argument that there was no interest, incentive or good reason for the skilled person to move away from the high performance optimum of using tin and antimony together towards tin alone, in view of evidence that such move would be retrograde or disadvantageous, cannot render the same move non-obvious provided the outcome involves no unexpected solution of a proper technical problem. There can be no invention in merely worsening the prior art, especially if such consequence is substantially foreseeable (cf. also T 119/82, "Gelation" OJ 5/1987, 217 at 227 and Headnote II) even if some aspect of the results may not be accurately predictable. As to the closest art below the level of claimed performance in the present application, i.e. the exemplified passivation with tin alone shown in (1), there was no more significant improvement in this respect than what could be gathered from the data available, as it was explained above. The subject-matter could therefore be considered as obvious with respect to closest arts in both directions, independently. In addition, however, the selection of an intermediate position within a well charted field, i.e. the choice of one of the agents expressly recommended there, has only brought about substantially intermediate and therefore predictable results, fully within the range of effects already available or within the natural fluctuation of numerical values therefor. More particularly, subject-matter falling structurally between two particular embodiments of a cited disclosure and displaying, in all

relevant respects, effects substantially between those known for the same embodiments, lacks inventive step in the absence of other considerations.

14. For the reasons above, the use of DPPD must be considered as obvious, and no inventive step can therefore be attributed to Claims 1 in the main and auxiliary requests.

Order

For these reasons it is decided that:

the appeal is dismissed.

The Registrar:

The Chairman:

F.Klein

P.Lançon