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D E C I S I O N
of 25 April 2005

Case Number: T 0857/02 - 3.3.6

Application Number: 95917740.3

Publication Number: 0711331

IPC: C10G 11/18

Language of the proceedings: EN

Title of invention:

Catalyst, method and apparatus for a particle replacement system for countercurrent feed-packed bed contact

Applicant:

Chevron U.S.A. Inc.

Opponent:

-

Headword:

Catalyst replacement system/CHEVRON

Relevant legal provisions:

EPC Art. 56

Keyword:

"Inventive step - no (main and auxiliary request): claimed subject-matter results from normal experimentation on the basis of commercial catalysts and as suggested in the prior art"

Decisions cited:

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Catchword:

-



Case Number: T 0857/02 - 3.3.6

D E C I S I O N
of the Technical Board of Appeal 3.3.6
of 25 April 2005

Appellant: Chevron U.S.A. Inc.
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 7 March 2002
refusing European application No. 95917740.3
pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: P. Ammendola
Members: G. Dischinger-Höppler
U. J. Tronser

Summary of Facts and Submissions

I. This appeal is from the decision of the Examining Division to refuse the European patent application No. 95 917 740.3 (international publication number WO-A-95/29970) entitled "catalyst, method and apparatus for a particle replacement system for countercurrent feed-packed bed contact".

II. The decision was based on the ground that the claimed subject-matter lacked an inventive step in view of the disclosure of

D1 US-A-5 076 908 in combination with that of

D2 US-A-4 422 960.

The Examining Division held that the method disclosed in D1 differed from the claimed one only in that the size, size distribution and aspect ratio of the catalyst particles to be used were unknown, but no evidence was present to show that these features were essential for the solution of the stated technical problem of limiting the expansion of a plug-flowing packed bed of catalyst particles to less than 10%. Instead it was apparent from D2 that the specific particle diameters set out in the claims were usual in the art.

III. The Applicant (hereinafter Appellant) filed an appeal against this decision. In response to a communication annexed to the summons to attend oral proceedings, wherein the Board raised objections under Article 84, 123(2) and 56 EPC, the Appellant under cover of a

letter dated 4 April 2005 filed amended sets of claims in a new main and auxiliary request which were replaced during oral proceedings held on 25 April 2005 by further amended sets of claims.

IV. The only independent Claim 1 of the main requests reads:

" A method for hydroprocessing in a reactor vessel a hydrocarbon feed stream that is upflowing through a hydroconversion reaction zone having a substantially packed bed of catalyst which is capable of plug flowing comprising the steps of

- (1) disposing catalyst in a reaction zone, said catalyst comprising a plurality of catalytic particulates having
 - (i) a mean diameter ranging from 6 Tyler mesh to 8 Tyler mesh (3.35 mm to 2.36 mm U.S. Standard Sieve Series);
 - (ii) a size distribution such that at least 90% by weight of said catalytic particulates has a diameter ranging from R1 to R2, wherein:
 - (1) R1 has a value of 0.093 inch (2.36 mm),
 - (2) R2 has a value of 0.131 inch (3.32 mm),and
 - (3) the value of the ratio R2/R1 ranges from 1.0 to 1.4,the size distribution being such that a maximum of 2.0% by weight of said catalytic particulates have a diameter less than R1, and such that a maximum of 0.4% by weight of said catalytic particulates have a diameter less than R3, wherein R3 is less than R1 and the value of the ratio R1/R3 is about 1.4;
 - (iii) an aspect ratio of less than 2.0; and

- (iv) a uniform density such that the density of at least 70% by weight of the individual catalyst particles does not vary by more than 10% from the mean density of all particles, the fresh catalyst uniform density ranging from 0.8 g/cc to 1.1 g/cc; and the substantially packed bed of catalyst occupying at least 50% by volume of the reactor vessel
- (2) upflowing through said catalyst of step (1) a hydrocarbon feed stream for hydroprocessing the hydrocarbon feed stream, said upflowing being at a rate of flow such that said substantially packed bed of catalyst expands to less than 10% by length beyond a substantially full axial length of said substantially packed bed of catalyst in a packed bed."

Claim 1 of the auxiliary request differs therefrom in that step (1) has been worded as follows:

- "(1) disposing catalyst in a reaction zone, said catalyst comprising a plurality of catalytic particulates having
 - (i) porous inorganic oxide support;
 - (ii) one or more catalytic metals and/or additional catalytic additives deposited in and/or on the porous inorganic oxide support;
 - (iii) a uniform size ranging from 6 Tyler mesh to 8 Tyler mesh (3.35 mm to 2.36 mm US Standard Sieve Series);

- (iv) a fines content up to 1.0 percent by weight through 8 Tyler mesh and up to 0.2 percent by weight through 10 Tyler mesh; and
- (v) an attrition up to 0.5 percent by weight through 8 Tyler mesh and up to 0.2 percent by weight through 10 Tyler mesh;
- (vi) a generally uniform spherical shape;
- (vii) a uniform density such that the density of at least 70% by weight of the individual catalyst particles does not vary by more than 10% from the mean density of all particles, the fresh catalyst uniform density ranging from 0.8 g/cc to 1.1 g/cc;
- (viii) a crush strength at least 5 pounds (2.3 kg) force;

the substantially packed bed of catalyst occupying at least 50% by volume of the reactor vessel".

Dependent Claims 2 to 19 or respectively 2 to 12 refer to preferred embodiments of the method of Claim 1 of the main and auxiliary requests.

V. The Appellant, orally and in writing, submitted in essence the following arguments:

- D1 related to a method of hydroprocessing at semi-commercial scale production but did not teach how to select a suitable catalyst. In particular, it did not disclose any specific size and density of the catalyst particles, let alone any values in relation to particle uniformity such as fines content and attrition. Instead a random testing of individual catalysts was proposed.

- In view of D1, the claimed subject-matter solved the technical problem of providing a systematic and practical means of selecting suitable catalysts for applying the method at full scale production.

- The teaching of D2 was irrelevant since it was not concerned with a packed bed reaction system as in D1 but with an ordinary fixed, moving or ebullated bed system with liquid hydrocarbon and gas flowing downwards through the reactor.

- Moreover, the catalyst of D2 was a commercial one with particles of preferably small-size and low density as compared with the catalyst of large particle size and high density used in the claimed method.

- Neither D2 nor D1 gave any hint that the non-preferred particle diameters in D2 would be suitable for carrying out the method of D1 at full scale and that the particle density should be higher.

VI. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 19 submitted in a main request or Claims 1 to 12 submitted in an auxiliary request, both during oral proceedings.

Reasons for the Decision

1. The Board is satisfied that the claims as amended in accordance with the new main and first auxiliary requests comply with the requirements of Article 123(2) EPC since their wording is supported by the application as originally filed. Since the appeal fails for other reasons, there is no need to give further details.
2. As will be apparent from the assessment of inventive step below, the subject-matter claimed in these requests is deemed to be novel in view of the available prior art.
3. *Main request*
 - 3.1 According to the Case Law of the Boards of Appeal of the European Patent Office (see I.D.3.1), a suitable starting point for the assessment of inventive step is normally a prior art document disclosing subject-matter conceived for the same or a similar purpose as the claimed invention.
 - 3.2 In the present case, the state of the art disclosed in D1 qualifies as a starting point for the assessment of inventive step since both, the application in suit and D1 which is mentioned in the application in suit as the relevant prior art document (page 2, lines 15 to 29), relate to a method for on-stream catalyst replacement during hydroprocessing a hydrocarbon feed stream by continuously or intermittently supplying replacement catalyst to a substantially packed bed of catalyst flowing in a plug-like manner downwardly in a reactor vessel whilst being in contact with a hydrocarbon feed

stream having a liquid component and a hydrogen-containing gas component, wherein said stream is up-flowing counter-currently to said bed at a controlled rate. In particular, they relate to a method of economically utilizing space within a hydroprocessing reactor vessel without substantial ebullition of the packed bed if contacted with a hydrocarbon feed moving at maximum space velocity through the vessel in counter-flow to the packed bed (application in suit: page 1, lines 8 to 25, page 3, line 31 to page 4, line 13 and page 7, lines 21 to 28; D1: Claim 1, column 1, lines 9 to 20, column 2, lines 38 to 58 and column 8, lines 36 to 42). This was not disputed by the Appellant.

- 3.3 D1 specifically discloses a method wherein it is essential to select the size, shape and density of the catalyst particles forming said bed in accordance with a maximum rate of flow of the feed stream to avoid ebullition or levitation of said particles and limit bed expansion to less than 10%, (column 7, lines 10 to 22 and column 10, lines 9 to 19).

In the examples of D1 catalysts of different sizes, shapes and densities are tested at liquid/gas flow rates suitable for limiting bed expansion to 10%. It is shown in Table I of D1 that for a given shape (sphere) and relative size (3.2) of the particles, the maximum flow rate for 10% bed expansion decreases as the density decreases. It is thus plausible that substantially the entire volume of the reactor vessel can be filled with a densely packed bed of catalyst by applying the up-flowing liquid/gas feed at a flow rate low enough to keep bed expansion below 10% (column 3,

line 66 to column 4, line 7 and column 18, lines 3 to 55).

As indicated above, particles in the shape of spheres are, amongst others, useful for that purpose (Tables I and II). This corresponds to an aspect ratio of 1.0 within the definition given in the application in suit (page 45, lines 13 to 24).

Commercial catalysts are said to be suitable provided that the catalyst particles are of high uniformity and sufficiently strong to maintain their integrity during movement into and out of the reactor vessel without attrition or breakage (column 17, lines 16 to 21).

However, the sizes mentioned in the examples of D1 are expressed as "relative size" for which no unit is given. Likewise, no unit is given for the particle density and the extent of uniformity and strength of the particles is not defined.

- 3.4 The Board, therefore agrees with the Appellant that the claimed subject-matter differs from the embodiments with spherical catalyst particles of D1 in that the particle diameter and the density have been identified, as well as the particle uniformity in terms of particle size and density distribution.

Thus, the subject-matter of Claim 1 is distinguished from those embodiments in that a catalyst is used wherein the particles have

- (a) a mean diameter between 2.36 and 3.35 mm with at least 90% wt of the particles having a diameter

between R1 and R2 wherein R1 is 2.36 mm and R2 is 3.32 mm, R2/R1 ranges from 1.0 to 1.4, at most 2% wt of the particles have a diameter < R1 and at most 0.4% wt of the particles have a diameter < R3 with R3 < R1 and R1/R3 being about 1.4; and

- (b) a uniform density with at least 70% wt of the particles having a density varying at most 10% from the mean density of all particles and ranging from 0.8 to 1.1 g/cc when fresh.

The Board notes that a clerical error is contained in item (a) since the introduction of the particular values for R1 (2.36 mm) and R2 (3.32 mm) from the description of the application as filed (page 5, lines 31 to 32) inevitably results in the particular ratio R2/R1 of 1.4. It is, thus, evidently impossible either that the ratio R2/R1 covers a range of values or that R1 and R2 are limited to particular values at the same time. However, the error is not essential for the following assessment of inventive step.

- 3.5 The Appellant submitted that the technical problem solved by the claimed subject-matter in view of D1, consisted in a systematic and practical means of selecting suitable catalysts for putting into practice the method of D1 in large scale production but conceded that no evidence is on file showing that this technical problem has actually been solved by the distinguishing features. In particular, nothing indicates that it is important for the solution of the stated technical problem to maintain the particle size and density within the specific ranges defined in Claim 1.

- 3.6 The single example given in the application in suit is not in contradiction to that finding since it only shows that the desired limited bed expansion to less than 10% can be obtained for a particular Ni/Mo catalyst on a porous alumina support having a particular surface area, density, size and size distribution of the particles, and under particular process conditions, including process temperature and pressure, gas/liquid feed composition and ratio, liquid withdrawal rate as well as the rate of catalyst feed and withdrawal (page 51, line 9 to page 55, line 29).
- 3.7 As a consequence, the technical results or effects actually achieved by the claimed invention in comparison with the disclosure of D1 must be considered to consist in the provision of a further method for hydroprocessing using a plug-flowing packed bed of catalyst particles with a bed expansion limited to less than 10%. Therefore, the technical problem solved in view of D1 has to be seen in providing an alternative method.
- 3.8 It remains to be decided whether or not the claimed solution is based on an inventive step in view of the cited prior art.
- 3.9 As stated above under 3.2 and 3.3, D1 aims at suppressing expansion of a plug-flowing packed catalyst bed to less than 10%. It is further stated in D1 that a commercial catalyst can be used, but that the shape, size, density and uniformity of the catalyst particles must be adapted for a pre-selected maximum flow rate of the feed (D1, column 8, lines 36 to 45, column 10, lines 9 to 17, and column 17, lines 16 to 21).

Thus, the skilled person knows from D1 the parameters he has to select and adapt in a commercial catalyst in order to arrive at the desired plug-flowing catalyst bed with limited expansion.

- 3.10 Catalysts for use in hydroprocessing of heavy hydrocarbon oils having a mean particle diameter within the claimed range of 2.36 and 3.35 mm are known in the art. This is apparent from D2 which discloses for that purpose catalysts having an average particle diameter of between 0.6 to 3.0 mm (column 16, lines 47 to 49).
- 3.11 The Board concludes, therefore, that a person skilled in the art looking for an alternative to the method disclosed in D1, would consider known hydro-processing catalysts of different particle size such as those disclosed in D2 and adapt by common serial investigation the other parameters (shape, density and uniformity) in conformity with the flow rate allowable to keep bed expansion below 10% as taught in D1. In doing so, he would come across those catalysts which have the claimed size and density, the selection of which is arbitrary and one of the options which are obvious to select for a skilled person in order to provide an alternative to the method of D1.
- 3.12 The Appellant's argument that the process of D2 was not comparable with that of D1 and, therefore, irrelevant is based on the fact that the hydrogen treating apparatus used in the examples of D1 is designed for feeding the gas/liquid reactants to the top of a reactor vessel containing a fixed bed of catalyst so

that the feed is flowing downwardly in the reaction vessel (column 47, lines 40 to 68).

However, D2 is not restricted to this particular flow system. Instead, it is indicated that the method of hydroprocessing heavy hydrocarbon oils of D2 may be carried out in any ordinary flow system, including moving bed systems and upward flow of the gas/liquid feed (column 34, lines 13 to 22). In this respect, the claimed process does not, therefore, differ from what is disclosed in D2.

3.13 The Appellant referred to the examples of D2 and submitted that the catalysts preferred therein were of small particle size and low density so that a combination of the catalysts of D2 with the method of D1 would not result in the claimed subject-matter.

It is indeed apparent from Tables 3-1 and 3-2 of D2 that the catalysts used in the examples have a mean particle diameter of only 1.0 and 1.2 mm and a catalyst density of 0.41 to 0.74 g/cm³, both being outside the claimed ranges. However, the examples in D2 are run in a fixed bed system with down-flowing feed (3.11 above) where ebullation can hardly occur (see D1, column 2, lines 38 to 45).

Therefore, D2 does not contain any prejudice against considering catalysts having particle sizes in the upper part of the disclosed range covering 0.6 to 3 mm if ebullation of the bed is an issue as in the system of D1 where the feed is up-flowing. The same applies even more so to the particle densities in the selected range of between 0.8 and 1.1 g/cm³ since it is shown in

D1 that bed expansion at a given maximum flow rate decreases with increasing density of otherwise identical catalyst particles. This was not disputed by the Appellant. It has to be noted in this respect that density is not mentioned in any other part of D2 than in Tables 3-1 and 3-2 and the density values are neither identified as particle density nor mentioned as critical.

- 3.14 Therefore, the Board does not see any reason why a skilled person should not follow the teaching in D1 to try commercial catalysts and adapt the shape, density and uniformity of the particles in accordance with a flow rate suitable to limit bed expansion to 10%. In doing so on the basis of the catalysts disclosed in D2 with particle sizes ranging from 0.6 to 3 mm, he would in an obvious manner arrive at the claimed subject-matter.

For these reasons, the Board concludes that the subject-matter of Claim 1 is not based on an inventive step as required by Article 52(1) EPC in combination with Article 56 EPC.

4. *Auxiliary request*

- 4.1 Claim 1 differs from that of the main request by specifying that the catalyst particles used in the claimed method have

- (a) a porous inorganic oxide support,
- (b) one or more catalytic metals deposited in or on the porous inorganic oxide support,

- (c) a particular fines content,
- (d) a particular attrition and
- (e) a particular value of crush strength (2.3 kg).

4.2 The Appellant submitted in essence that it was its intention to restrict the claimed subject-matter to the only example contained in the application in suit and that it was important for the claimed method that the particles are uniform both in size and density in order to avoid particle segregation.

The Board notes that none of the additional features is suitable to support the presence of inventive step since they concern catalyst properties which are either also present in the catalysts of the cited prior art (see D1, column 1, lines 51 to 57 for features (a) and (b); D2, column 10, lines 62 to 68 for features (a) and (b) and Tables 3-1 and 3-2 for feature (e)) or design options for which no particular advantage is apparent and which are obvious to be selected (features (c) to (e)) in view of the teaching in D1 that the catalyst particles should have a high degree of uniformity and be sufficiently strong to maintain their integrity during movement into and out of the reactor vessel without attrition or breakage (column 17, lines 16 to 21).

4.3 For these reasons, the Board finds that the subject-matter of Claim 1 of the auxiliary request is also not based on an inventive step and does not comply with the requirements of Articles 52(1) and 56 EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

G. Rauh

P. Ammendola