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Datasheet for the decision of 19 January 2011

Case Number:	т 0867/08 - 3.3.06
Application Number:	00202032.9
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Language of the proceedings:	EN

Title of invention: Improved fischer-tropsch process

Patentee: ENI S.p.A., et al

Opponent: Sasol Technology Ltd. ExxonMobil Research and Engineering Company

Headword: Fischer-Tropsch process/ENI

Relevant legal provisions:

Relevant legal provisions (EPC 1973): EPC Art. 56

Keyword: "Inventive step (all request) no: obvious desiderata"

Decisions cited:

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Beschwerdekammern

Boards of Appeal

Chambres de recours

Case Number: T 0867/08 - 3.3.06

DECISION of the Technical Board of Appeal 3.3.06 of 19 January 2011

Appellant:	ENI S.p.A.
(Patent Proprietor)	Piazzale E. Mattei, 1 I-00144 Rome (IT)
Representative:	Coletti, Raimondo Barzanò & Zanardo Milano S.p.A. Via Borgonuovo, 10 I-20121 Milano (IT)
Respondent: (Opponent)	Sasol Technology Ltd. PO Box 1 Klasie Havenga Road Sasolburg 9570, Republic of South Africa (ZA)
Representative:	Copsey, Timothy Graham Kilburn & Strode LLP 20 Red Lion Street London WC1R 4PJ (GB)
(Opponent)	ExxonMobil Research and Engineering Company 1545 Route 22 East P.O. Box 900 Annandale NJ 08801-0900
Representative:	von Kameke, Allard Uexküll & Stolberg Patentanwälte Beselerstrasse 4 D-22607 Hamburg (DE)
Decision under appeal:	Decision of the Opposition Division of the European Patent Office posted 14 April 2008 revoking European patent No. 1061115 pursuant to Article 102(1) EPC 1973.

Composition of the Board:

Chairman:	P	-P. Bracke
Members:	G.	Dischinger-Höppler
	J.	Geschwind

Summary of Facts and Submissions

- I. This appeal is from the decision of the Opposition Division to revoke the European patent No. 1 061 115 relating to an improved Fischer-Tropsch process.
- II. Two notices of opposition had been filed against the granted patent, wherein the Opponents sought revocation of the patent on the grounds of, inter alia, Article 100(a) EPC for lack of inventive step (Article 56 EPC). The oppositions were based, amongst others, on document

D4 EP-A-0 450 860.

III. The decision under appeal was based on the granted claims as the main request and amended claims according to an auxiliary request.

Claim 1 of the main request reads:

"1. A process for optimizing the production of heavy hydrocarbons according to the Fischer-Tropsch process and the relative separation of the above hydrocarbons, starting from mixtures of reagent gases, essentially consisting of CO and H_2 , optionally diluted with CO₂, in the presence of supported catalysts, which comprises:

(a) feeding the reagent gases into a reactor, preferably from the bottom, so as to obtain a good dispersion of the solid in the liquid phase, in this at least way partially transforming the reagent gases into heavy hydrocarbons, the gas flow-rates being such as to operate under heterogeneous or churn-turbulent flow conditions; (b) at least partially recovering the heavy hydrocarbons formed in step (a) by their external or internal separation from the catalyst particles;

the above process being characterized in that in step (a) the reaction takes place:

(1) in the presence of solid particles so that the particle Reynolds' number (Re_p) is greater than 0.1, wherein $Re_p = d_p \cdot v \cdot \rho_1 / \mu$ wherein d_p is the average particle diameter, v is the relative velocity between particle and liquid, ρ_1 is the density of the liquid, μ is the viscosity of the liquid; (2) maintaining the solid particles suspended at a height H, with such U_s , U_1 and U_g values as to have a Bodenstein number $Bo_s \leq 1$."

- IV. In its decision, the Opposition Division revoked the patent for lack of inventive step of the subject-matter of Claim 1 of both requests inter alia in view of the disclosure of document D4 as the closest prior art.
- V. This decision was appealed by the patent Proprietor, now Appellant, who filed amended sets of claims in two auxiliary requests in a letter dated 12 January 2011.

Claim 1 of both auxiliary requests differs from that of the main request in that now " Re_p is from 0.2 to 25" and " $Bo_s \leq 0.4$ ". In addition to that the following feature has been added at the end of Claim 1 of the second auxiliary request: "and further characterized in that

- 2 -

the solid catalytic particles consist of cobalt supported on alumina".

- VI. Upon requests made by all parties, oral proceedings before the Board of Appeal were held on 19 January 2011.
- VII. The Appellant, orally and in writing, made in essence the following submissions:

- The technical problem solved by the invention consisted in the finding of process conditions where it was possible to optimize synthesis and separation.

- Document D4 offered specific operative conditions defined in three equations as a solution to the problem of optimum synthesis without using downcomers as in other prior art. Therefore, document D4 was the closest prior art. However, document D4 did not address the problem of optimized separation. Instead it was indicated in Example 8 of this document that particles with diameters greater than 100 µm cannot be effectively fluidized.

- By contrast, as was illustrated in Example 7 of the patent in suit, it was possible to use larger particles, e.g. of a diameter of 200 μ m with Re_p being much greater than 0.1 and yet to obtain a good dispersion in the solid phase so that Bo_s < 0.4 by suitably dimensioning the reactor. As this was not suggested in the prior art, the criteria for an inventive step were fulfilled for the subject-matter claimed in both, the main and first auxiliary requests. - Concerning the second auxiliary request, it was argued that document D4 did not suggest alumina as support for the catalyst.

VIII. The Respondents, orally and in writing, submitted the following arguments:

- Neither did the claimed process exclude the use of particle sizes of 100 µm or less nor was the teaching of document D4 restricted to Reynolds' numbers of 0.1 or less. The claimed process instead consisted only in the replacement of the known Stokes' law correlation by another known correlation for describing the settling velocity of the catalyst particles.

- The same arguments applied to the subject-matter of the auxiliary requests since the added features were also known from document D4. Moreover, those features were either only arbitrary selections of the limiting values or usual in the art.

IX. The Appellant requests that the decision under appeal be set aside and the patent be maintained as granted or, alternatively, in amended form on the basis of the claims according to the first or second auxiliary request filed with letter dated 12 January 2011.

The Respondents requested that the appeal be dismissed.

Reasons for the Decision

 The patent in suit relates to an improved process for the Fischer-Tropsch (FT) reaction which essentially

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consists in a first reaction phase and a second separation phase. It is explained in the patent in suit that in the FT process the flow rates of the fluids should be such as to guarantee a turbulent flow regime in the column and a practically homogeneous suspension of the solid in the whole reaction volume.

Further, it is indicated in the patent that the solid particles should have dimensions which are sufficiently large so as to enable them to be easily separated from the liquid products, but sufficiently small for being easily fluidized. While the average particle diameter in the FT process may vary from 1 to 200 μ m, dimension of less than 10 μ m would make the separation very expensive (paragraphs 1 to 8 of the patent).

- 2. Document D4 forms the starting point in the patent in suit and is referred to as disclosing that an average particle diameter of less than 51 µm was required. This particle diameter, however, would create drawbacks in the separation phase (paragraphs 14 and 15 of the patent).
- 3. Document D4 actually discloses a method for optimal operation of a slurry bubble reactor for the FT synthesis of hydrocarbons from synthesis gas in the presence of supported cobalt catalyst particles. The liquid FT products are separated from the solid catalyst by filtration or decantation (document D4, Claim 2 in combination with page 2, lines 5 to 14, page 3, lines 47 to 48, page 5, lines 10 and 11, page 6, lines 5 to 6 and lines 41 to 42 and page 11, lines 50 to 52).

- 5 -

Further, it is taught in document D4 that the gas flow in the reactor should be such as to provide bubbly or churn-turbulent flow rather than slug flow with an appropriate velocity so that the solid phase is fluidized while still maintaining plug flow in the gas phase in order to give better mass transfer performance during reaction (page 4, lines 33 to 35, page 6, lines 43 to 50).

According to document D4 (Claim 1), this is achieved by operating with a solid particle settling velocity (U_s) , a dispersion coefficient (D) and a gas velocity (U_g) such that the following equations are respected:

 $0.5 (U_{s} - U_{L}) \leq D/H;$

 $U_{s} = 1/18 \cdot d_{p}^{2} \cdot (\rho_{s} - \rho_{1}) / \mu \cdot g \cdot f(C_{p});$ and

 $U_q \geq 2 D/H$

wherein

ρ_{s}	=	effective density of the particles
ρ	=	density of the liquid
μ	=	viscosity of the liquid
$f(C_p)$	=	hindered settling function
	=	volume fraction of solids in the slurry
$U_{\rm L}$	=	liquid velocity along the column
Н	=	height of the expanded liquid in said reactor
g	=	gravitational constant
dp	=	diameter of particles.

Moreover, document D4 explicitly states that making the catalyst particles smaller improves the fluidization

but increases the difficulty in separating them from the liquid product stream (page 6, lines 41 to 42).

Hence, document D4 addresses the same technical problem as the patent in suit (point 1 above) and qualifies, therefore, as a suitable starting point for the assessment of inventive step.

4. By contrast, the claimed process requires that the reaction takes place such that the Bodenstein number is \leq 1 and that the Reynolds' number of the particles $\text{Re}_{p} > 0.1$

wherein $Re_p = d_p \cdot \nu \cdot \rho_1 / \mu \text{ with}$ $\nu = relative \text{ velocity between particle and liquid}$

instead of respecting the equations given in document D4.

5. Since, the Bodenstein number of the solid Bo_s corresponds to $(U_s - U_L) \cdot H/D$, the above term $0.5(U_s - U_L)$ $\leq D/H$ in the first equation of Claim 1 of document D4 is equivalent to $Bo_s \leq 2$ (see paragraph 18 of the patent). The Board notes that the adaptation of the Bodenstein number implies proper selection of inter alia the gas velocity U_g which is also defined in third equation of Claim 1 of document D4 (see Claim 1 of both the patent and document D4).

> The Appellant argued that the use of the term $U_s = 1/18 \cdot d_p^2 \cdot (\rho_s - \rho_1)/\mu \cdot g \cdot f(C_p)$, i.e. the second equation in Claim 1 of document D4, required as a precondition that the Stokes' law applied and that,

therefore, the Reynolds' number of the particles had to be $Re_p < 0.1$. On the basis of example 8, the maximum diameter of the catalyst particles useful in the process of document D4 was thus below 51 µm.

- 8 -

6. However, according to the description of the preferred embodiment particle diameters of up to 200 μ m can be used in the process of document D4 (page 5, lines 42 to 45). This is corroborated by Figure 7 which illustrates the acceptable operating range found in Example 8 and shows optimal performance also for catalyst particles having diameters above 51 μ m and even above 100 μ m. For such particle sizes the particle Reynolds' number is clearly greater than 0.1 since, as was correctly shown by the Appellant, the condition Re_p < 0.1 ceases to apply for particle diameters above 51 μ m under the operating conditions used in Example 8 of document D4.

> Thus, despite the fact that for the process claimed in document D4 the Stokes' law might apply and despite the statement that the allowable range of particle sizes that can be effectively separated from the liquid medium underlies practical restrictions (page 11, lines 45 to 52), the disclosure of document D4 is not limited to the use of catalyst particles having diameters below 51 µm and a Reynolds' number of 0.1 or less.

In Example 8 of document D4, no specific Bodenstein number is disclosed. It is only stated that particles with diameters above 100 μ m cannot be effectively fluidized without a backmixing debit on the kinetic driving force (page 11, lines 49 to 50). Hence, the claimed process differs from the embodiments with Reynolds' numbers greater than 0.1 illustrated in Example 8 and Figure 7 of document D4 only in that the Bodenstein number of the solid $Bo_s \leq 1$.

7. According to the patent in suit, the technical problem solved by the claimed method in view of the prior art disclosed in document D4 consists in an optimized operation in both the reaction phase and separation phase without substantially varying the activity of the catalyst (paragraph 16).

> According to the Appellant it was apparent from Example 7 of the patent in suit that the above technical problem actually has been solved by the claimed process.

8. Example 7 of the patent relates to calculations concerning the effect of geometry of the bubble column reactor on the dispersion degree of the solid phase. It is calculated that it should be possible to use catalyst particles having a particle diameter d_p of 200 µm and a Reynolds number Re_p of greater than 0.1 (here 8.9) and still to have a low Bodenstein number Bo_s of 1 or less (here 0.26) to guarantee proper fluidisation of the particles.

> These calculations have not been verified by experimentation. Apart from the fact that theory and praxis very often differ from each other significantly, the calculations in Example 7 of the patent only apply for the specific conditions used. Assuming that in FT processes the properties of the wax are always the same, specific values have been selected at least for the parameters H, U_{q} , ρ_{s} , the average concentration of the

solid $C_{\rm p}$ and the diameter of the reactor $D_{\rm c}$ in order to obtain those results.

Hence, while Example 7 might show a solution to the technical problem of optimizing synthesis and separation, this solution is not present in Claim 1 of the patent in suit since none of the selected values of the parameters which determine the results in Example 7 have been specified in Claim 1.

It is apparent from the patent in suit that other values of the parameters H, U_g , ρ_s , C_p and D_c do not necessarily present an improvement over the process of document D4 (see e.g. Figures 1 and 4 and Example 7).

9. According to the Appellant, the statement in Example 8 of document D4 indicating that catalyst particles greater than 100 µm in diameter cannot be effectively fluidized (point 6 above) amounted to a prejudice against the using of catalyst particles greater than 100 µm in diameter.

However, the Board is not convinced by this argument since that statement in Example 8 of document D4 is relevant only for the specific catalyst material of relatively high density (2.7 g/cm³) which has been used in the example under specific conditions ($U_g = 5$ cm/sec, $f(C_p) = 1$).

As is correctly indicated in the patent in suit, the Reynolds' number of the catalyst particles in FT processes is virtually dependent only on the density and dimensions (average diameter) of the particles. Further, it is stated that the expert in the field who knows the density of the catalytic particles, can obtain particles with an average diameter such that particle Reynolds' number Re_p is greater than 0.1, preferably from 0.2 to 25 (paragraph 31).

Hence, it is clear that the allowable particle diameter depends on the density of catalyst material used, the gas velocity U_g and the function $f(C_p)$ which varies between 1 and 0 (see document D4, page 11, lines 27 to 30).

- 10. Claim 1 thus covers embodiments for which no effect with respect to the ease of separation of the catalyst particles from the liquid has been shown when compared with the process of document D4.
- 11. In view of the disclosure in Example 8 of document D4, the technical problem actually solved by the means claimed, namely by selecting the Bodenstein number of the solid so that $Bo_s \leq 1$ (see point 6 above), might be seen in an improvement of the FT synthesis in the reaction phase.
- 12. It remains to be decided whether, in view of the prior art, it was obvious for someone skilled in the art to solve the above stated technical problem of improving the FT synthesis by the means claimed, namely by selecting H, U_s, U₁ and U_g so that the Bodenstein number fulfils the constraint $Bo_s \leq 1$
- 13. Keeping in mind, as was submitted by the Respondents, that the Bodenstein number just defines how uniformly the catalyst particles are fluidized, with $Bo_s = 0$ for an ideal backmixing reactor, it is obvious for a

skilled person to conduct the FT process under conditions where the Bodenstein number is as low as possible. This is also reflected in document D4, where a Bodenstein number of 2 is nothing else than the upper limit, while lower Bodenstein numbers are clearly preferred since that document also aims at high productivity which means that the catalyst distribution should be as uniform as possible (page 6, lines 32 to 33 and 43 to 50). Excellent reactor performance is disclosed if $H = D/U_s-U_L$) or, in other words, $Bo_s = 1$ (page 6, lines 19 to 21).

Document D4 even discloses how to obtain suitable Bodenstein numbers since it refers to the effect of the reactor geometry on the settling behaviour of the particles, hence the extent of backmixing or the Bodenstein number. Nothing else is illustrated in Example 7 of the patent in suit which was essentially relied on by the Appellant when discussing inventive step (document D4, page 2, lines 52 to 53 and page 3, lines 28 to 32).

Hence, the feature of selecting H, U_s , U_1 and U_g so that the Bodenstein number fulfils the constraint $Bo_s \leq 1$ amounts to an obvious desideratum, in particular if the actually selected values are not indicated. On such a feature an inventive step cannot be based.

14. For these reasons, the Board finds that the subjectmatter of Claim 1 of the Respondent's main request does not comply with the requirements of Articles 52(1) and 56 EPC. 15. The same applies to Claim 1 of the first auxiliary request which differs from Claim 1 of the main request only insofar as the values for Rep and Bos are more restricted to indicate the obvious desiderata of still larger particle sizes and more uniform fluidisation.

> It also applies to Claim 1 of the second auxiliary request since the feature of using an alumina supported cobalt catalyst is also suggested in document D4 (page 5, lines 10 to 11).

16. Consequently, the subject-matter of Claim 1 of none of the Appellant's requests complies with the requirements of Articles 52(1) and 56 EPC, so that the appeal has to be dismissed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

C. Vodz

P.-P. Bracke