Decision of 4 April 2003

Case Number: T 0727/99 - 3.3.5
Application Number: 91302710.8
Publication Number: 0450860
IPC: B01J 8/22
Language of the proceedings: EN

Title of invention: Method of operating a slurry bubble column

Patentee: ExxonMobil Research and Engineering Company


Headword: Operating a slurry bubble column/EXXONMOBIL

Relevant legal provisions: EPC Art. 56, 83, 123(2)

Keyword: "Amendments - isolated features not disclosed in claimed combination"
"Sufficiency of disclosure - yes, no undue burden"
"Inventive step - yes, prejudice in the art"
"Reformatio in peius - allowed"
"G 0001/99 applied"

Decisions cited: G 0001/99, T 0301/87, G 0009/91, G 0010/91

Catchword: -
DECISION
of the Technical Board of Appeal 3.3.5
of 4 April 2003

Appellant:
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Representative:
-

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Decision under appeal:
Interlocutory decision of the Opposition
Division of the European Patent Office posted
12 May 1999 concerning maintenance of
European patent No. 0450860 in amended form.

Composition of the Board:
Chairman: R. K. Spangenberg
Members: G. J. Wassenaar
          R. T. Menage
Summary of Facts and Submissions

I. The appeal is from the decision of the Opposition Division to maintain European patent No. 0 450 860 in amended form. In the Notice of Opposition the appellant (opponent) has raised objections under Article 100(a) and 100(b) EPC against the claims as granted. During the opposition proceedings objections have been raised under Article 123 EPC against the amended claims. Amended claim 1 as maintained by the Opposition Division reads as follows:

"A method for optimally operating a three phase slurry bubble column for the production of Fischer-Tropsch hydrocarbon synthesis wax in which solid catalyst particles are fluidized in the liquid phase by bubbles of the gas phase, comprising:

(a) injecting the gas phase into said column having a diameter of at least 15 cm at an average gas velocity along the column $U_g$ such that the flow regime is churn turbulent in the substantial absence of slug flow;

(b) fluidizing supported cobalt catalyst particles of average diameter $d_p > 5\mu m$ in the liquid phase to a height $H$ of greater than 3 meters in the column by operating with a solid catalyst particle settling velocity $U_s$ and dispersion coefficient $D$ such that:

$$0.5(U_s-U_L) \leq D/H$$

where

$$U_s = \frac{1}{18} d_p^2 \left( \frac{\rho_g - \rho_l}{\mu} \right) g f(C_p);$$

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and

(c) maintaining plug flow in said column by operating with a gas phase velocity \( U_g \), expanded liquid height \( H \) and dispersion coefficient \( D \) such that:

\[
U_g \geq 0.2 \frac{D}{H}
\]

wherein

\( \rho_s \) = effective density of the particles

\( \rho_1 \) = density of the liquid

\( \mu \) = viscosity of the liquid

\( f(c_p) \) = hindered settling function

\( c_p \) = volume fraction of solids in the slurry (liquid plus solids)

\( U_L \) = liquid velocity along the column

\( H \) = height of the expanded liquid in said reactor

\( g \) = gravitational constant

\( d_p \) = diameter of catalyst particles.

II. In the decision under appeal, inter alia, the following documents were considered:


D17: Catalysis Letters, 7 (1990), "The Shell Middle Distillate Synthesis Process" by J. Eilers et al., pages 253 to 270.


III. In its statement of the grounds of appeal, the appellant maintained that the amended claims extended beyond the content of the application as originally filed, that the subject-matter of the claims lacked an inventive step and that the invention was insufficiently disclosed. Also new documents were cited, of which the following remained relevant for this decision:

D40: Chemierohstoffe aus Kohle, J. Falbe, Georg Thieme Verlag, Stuttgart 1977, Chapter 8, "Fischer-Tropsch-Synthese".


Later in the proceedings a further objection was made under Article 84 EPC and new documents were cited, of which the following remained relevant for this decision:


IV. A third party made observations under Article 115 EPC and cited further documents.

V. The respondent (patentee) refuted the arguments of the appellant and the third party. New evidence was filed as "Attachments A, B, C and D", "Table 1" and "Amended Figures 1 and 2". With its letter dated 20 February 2003 new sets of claims were filed, a main request, three auxiliary requests as alternative A, and four auxiliary requests as alternative B. Claim 1 of the main request was identical to claim 1 as maintained by the Opposition Division. The independent claims of the auxiliary requests according to alternative A comprised further limitations and required, as claim 1 of the main request, that the flow regime was churn turbulent.
The independent claims of the first and second auxiliary requests according to alternative B comprised as limitation that $U_g > 8 \text{ cm/s}$.

Claim 1 of the third auxiliary request according to alternative B comprised as limitation that the conversion was at least 0.4.

Claim 1 of the fourth auxiliary request according to alternative B differed from claim 1 of the main request in that the requirement of the churn turbulent flow regime was deleted and the catalyst was more narrowly defined, parts (a) and (b) of claim 1 now being worded as follows:

"(a) injecting the gas phase into said column having a diameter of at least 15 cm at an average gas velocity along the column $U_g$ such that the flow regime is in the substantial absence of slug flow;

(b) fluidizing inorganic oxide supported cobalt catalyst particles of average diameter $d_p > 30\mu m$ in the liquid phase to a height $H$ of greater than 3 meters in the column by operating with a solid catalyst particle settling velocity $U_s$ and dispersion coefficient $D$ such that:"

Declarations of Professor Deckwer, dated 11 March 2002, 6 June 2002 and 17 February 2003, a declaration of Dr Eric Herbolzheimer dated 6 February 2003 and a declaration of Professor Bell, dated 19 March 2003, were filed.

During oral proceedings, which took place on 3 and 4 April 2003, the respondent filed a fifth auxiliary request according to alternative B. Claim 1 of this request differed from claim 1 of the fourth auxiliary
request in that part (b) was further limited to read as follows:

"(b) fluidizing inorganic refractory oxide supported cobalt catalyst particles of average diameter $d_p > 30 \mu m$, the oxide consisting of titania or alumina, in the liquid phase to a height $H$ of greater than 3 meters in the column by operating with a solid catalyst particle settling velocity $U_s$ and dispersion coefficient $D$ such that:

VI. The arguments of the appellant, in so far they are relevant for this decision, may be summarized as follows:

Unallowable extension

The claims of all the requests were composed of features which had not been disclosed in the original application in the claimed manner. In particular the use of a column having a diameter of at least 15 cm, a height of the liquid phase of more than 3 metres, the use of supported cobalt catalyst, the use of a churn turbulent flow regime, the use of an average gas velocity $U_g > 8 \text{ cm/s}$ and operating conditions such that a conversion of at least 0.4 was reached, were not disclosed in the claimed combination.

Insufficient disclosure

There was no example of an embodiment of any of the claims. If the dispersion coefficient of the catalyst particles and the gas were the same, as required by all the claims, the gas velocity must have been so low that it was not possible to fluidize the particles to a
height of more than 3 meters. Moreover, the patent in suit did not teach how to build and operate the reactor so that the dispersion requirements were met. Finding the suitable conditions by trial and error was an undue burden.

Clarity

The meaning of the expression "hydrocarbon synthesis wax" in the claims was not clear. During the proceedings the respondent had given this expression different meanings; fractions of C12+, C14+, C19+ and C21+ had been mentioned.

Admissibility of the fourth and fifth auxiliary request, alternative B (reformatio in peius)

By the deletion of the churn turbulent flow regime requirement, claim 1 was broader than the claim as maintained by the Opposition Division. Since the respondent did not appeal, this amounts to a reformatio in peius situation which, in view of the Enlarged Board of Appeal decision G 1/99, was not allowable. The new particle size limitation in the fourth auxiliary request was not suitable to overcome this objection. The further limitation in the fifth auxiliary request to particular catalysts was also not appropriate because they were a selection from a broader range and this selection had not been disclosed in combination with the other features of the claim. A more suitable amendment to replace the unallowable selected feature "churn turbulent flow" would be its replacement by "bubbly or churn turbulent flow". This was based on the application as originally filed and excluded the
transition flow regime shown in D10, page 707, Figure 16.

Inventive step (fourth auxiliary request, alternative B)

The method of claim 1 was essentially a routine scaling up of the method disclosed in example 42 of D26 without surprising effect. The Flory alpha values for Patentee's comparative examples were not substantially different from those calculated for example 42 of D26. The dispersion requirements of claim 1 were obvious desiderata in view of the expected production increase by higher Peclet values in a positive order reaction system. The positive order of the F-T reaction had been published in many documents and was well known in the art, eg D49, D50 and D45.

VII. The respondent's arguments with respect to the above-mentioned objections may be summarized as follows:

The present claims were essentially a combination of the claims as granted. Since objections under Article 100(c) EPC had not been raised in the Notice of Opposition, amendments resulting from such a combination might not be attacked in the appeal proceedings without the consent of the respondent, which was not given in this case. The only feature not present in the granted claims was the churn turbulent flow regime. This feature was, however, clearly disclosed in the application as filed as one of the two acceptable flow regimes out of three possible flow regimes. Moreover, from examples 4 and 5 it was evident that gas velocities above 8 cm/s were preferred, which automatically provided a churn turbulent flow. A
conversion of 0.4 was disclosed in Figure 2 as the minimum conversion for which the advantage of a high Peclet number was shown.

It was evident to the skilled person, and it followed clearly from the description of the patent in suit, that the dispersion coefficient $D$ in part (b) of the claims related to the dispersion of the solid particles and that the dispersion coefficient $D$ in part (c) was the gas dispersion coefficient. Only at low gas velocities could these coefficients be the same. It was evident that the claims, comprising a dependent claim according to which the average gas velocity was up to 25 cm/s, were not limited to only one low gas velocity. Example 7, in which the gas dispersion coefficient had been set equal to the solid dispersion coefficient, was only to demonstrate the operating conditions which had to be chosen in that particular case. For other cases it was not so easy to visualise these operating conditions, but that did not mean that the invention was limited to that particular case. It is true that the exact dispersion coefficients could only be measured in an operating reactor. As pointed out by the appellant, the condition mentioned in part (b) required in fact that the solid concentration at the top of the slurry was at least 14% of that at the bottom. The condition mentioned in part (c) implied a Peclet number above 0.2. It was explained in the patent in suit, and well known in the art, that the Peclet number was very much dependent upon the height/diameter ratio of the reactor and there were standard calculation methods to get a first approximation of this number. The invention did not reside in the development of a new kind of reactor. Standard reactors might be used for the claimed process but the patent taught which kind of
reactors should be chosen and how they should be operated. Knowing these conditions it was no undue burden to select suitable reactors and operating conditions for performing the claimed methods.

The production of wax was disclosed in the original application and in claim 3 as granted without indication of the carbon range. Most workers in the field of F-T synthesis seemed to agree that wax was the C19+ hydrocarbon fraction. Since lack of clarity was not a ground for opposition, the objection that wax was not further specified might not be taken into consideration.

The omission of the churn turbulent feature in the fourth auxiliary request B did not give rise to a reformatic in peius situation because under practical conditions supported cobalt catalysts having a size as required by claim 1 could only be fluidized to the indicated height in a churn turbulent flow regime. Moreover, G 1/99 did not set an absolute bar on the reformatic in peius if a reasonable further limitation to compensate the broadening of a claim by deleting the unallowable feature was not possible. The purpose of the appellant's proposal to replace churn turbulent with bubbly or churn turbulent was not clear. If it intended to exclude a transition flow regime, it rendered the claim unclear, because a transition flow regime has never been recognized as a separate flow regime in the art. If it covered all flow regimes apart from slug flow, then there was no difference with the wording of claim 1 according to the fourth auxiliary request.
D26 did not represent the closest prior art because it only related to laboratory scale experiments and the product comprised only a low amount of wax. The closest prior art was rather D4, disclosing wax production in a slurry bubble column (page 255, Table 6). The processes disclosed in D4 had several drawbacks making them commercially not interesting. The problem underlying the invention was to provide a commercially viable process with a selectivity to wax. This problem was solved as shown by the comparative examples. The large scale and complex nature of the processes made it impossible to make a comparison under equal conditions. It had not been known that the overall reaction kinetics was positive so that it could not be expected that a high Peclet number was advantageous. In D17, a document of the appellant presented at the AIChE Spring National Meeting shortly before the priority date of the patent in suit, a fixed bed reactor was proposed and the use of a slurry bubble column for the production of wax was clearly rejected (page 264).

VIII. The appellant (opponent) requested that the decision under appeal be set aside and that the European patent No. 0 450 860 be revoked.

The respondent (patentee) requested that decision under appeal be set aside and that the patent be maintained according to one of the versions in the following order:

- Main request

- Alternative A; first, second, third auxiliary request

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Alternative B; first, second, third, fourth auxiliary request, all of them as filed on 20 February 2003;

Alternative B; fifth auxiliary request as submitted during oral proceedings.

Reasons for the Decision

1. The independent claims of the main request and the auxiliary requests according to alternative A comprise the condition that the flow regime is churn turbulent. This feature was not present in the claims as originally filed and the claims as granted. This feature has never been presented in the original application as a part of the invention. It is mentioned only once in the original application on page 17 in a sentence stating that if the effective reactor diameter is too small slug flow may occur rather than a dispersion of small gas bubbles typically of bubbly or churn turbulent flow which give better mass transfer performance. The only flow regimes recognized in the prior art as being distinguishable in slurry bubble columns are slug flow, bubbly flow and churn turbulent flow. The transition between these flow regimes is not clearly delineated, but the transition range cannot be regarded as a separate flow regime; see D10, page 707, Figures 15 and 16. Said sentence in the original application, therefore, does not provide more information than to avoid slug flow and to use any of the other flow regimes. There is certainly no disclosure of using the churn turbulent flow regime in combination with F-T synthesis in the presence of supported cobalt catalysts. The only example in the
patent in suit relating to such a synthesis is Example 8, disclosing a Ug of 5 cm/s. Other gas velocities are not disclosed in this context. It is undisputed that at a gas velocity below 7 cm/s a stable churn turbulent flow regime does not exist (D10, page 707, left side column). It is true that Figure 4 of the patent in suit, relating to Example 4, shows that the decay length is higher at higher gas velocities up to about 10 cm/s. The results presented in Fig. 4 have been determined by measuring the solids concentrations of titania and glass particles in F-T wax as the liquid carrier in a non-reactive column. From Figure 4 it cannot be unambiguously derived that for the production of wax in the presence of a supported cobalt catalyst average gas velocities above 8 cm/s, giving rise to churn turbulent flow, should be chosen. Figure 5 shows the dependency of the decay length (D/U) as a function of the solids concentration measured at different gas velocities running from 2 to 18 cm/s in a similar non-reactive system. These gas velocities are just examples for showing the relationship between decay length and solids concentration at different gas velocities. Again it cannot be unambiguously derived therefrom to operate a process for the production of F-T wax with a supported cobalt catalyst at an average gas velocity above 8 cm/s. Thus the combination of features in the independent claims of the main request and the auxiliary requests according to alternative A has no basis in the application as originally filed, so that these claims contain subject-matter which extend beyond the content of the application as filed (Article 123(2) EPC).
2. The independent claims of the first and second auxiliary request according to alternative B comprise the condition that $U_g > 8$ cm/s. This inequality only appears in Example 5 and Figure 5. In Example 5 it is indicated which correlation formula between decay length and particle concentration is reasonable at gas velocities below 4 cm/s, above 8 cm/s and between these velocities. It is not indicated that gas velocities above 8 cm/s are preferred, let alone in combination with the other features of the claims. These claims, therefore, do not fulfil the requirements of Article 123(2) EPC either.

3. Claim 1 of the third auxiliary request according to alternative B comprises the condition that the production of F-T wax is performed at a conversion of at least 0.4. The conversion number 0.4 only appears in Figure 2, a graphical representation of the relationship between the normalized reactor volume and the inverse Peclet number at different conversion rates running from 0.4 to 0.95. The conversion rates are only examples in model calculations for reaction rates that are first order in the concentration of the reactants (Example 2). There is no relationship with the actual production of F-T wax, let alone with the other features of claim 1. Thus, claim 1 of the third auxiliary request according to alternative B is also not allowable under Article 123(2) EPC.

4. Claim 1 of the fourth auxiliary request according to alternative B is based on claim 3 in combination with claims 4, 7, 8 and 13 as granted with the proviso that the restricted solid particle velocity requirement in granted claim 3 is replaced with the more general expression of granted claim 1. It is, however, directly
apparent that the limited $U_s$ function in granted claim 3 is based on particular assumptions with respect to the densities of the liquid and the catalyst particles and the hindered settling function as explained in Example 8. It is clear to the skilled person that the invention as originally disclosed with respect to the production of F-T wax was not intended to be limited to the exact Figures given in Example 8. The replacement of the expression for $U_s$ in granted claim 3 with the more general expression in granted claim 1 is therefore an allowable amendment. Claim 13 being dependent upon claim 8, claim 8 being dependent upon claim 7, claim 7 being dependent upon claim 4 and claim 4 being dependent upon claim 3, the combination of features according to claim 1 of the fourth auxiliary request follows from a disclosed combination of granted claims. The appellant's objections under Articles 84 and 123 EPC, against claim 1 of the fourth auxiliary request, therefore, do not arise from amendments made during the opposition and appeal proceedings and, in the absence of the consent of the respondent (point VII above), are not taken into consideration by the Board for the reasons set out in T 301/87, OJ EPO 1990, 335, G 9/91 and G 10/91, OJ EPO 1993, 408 and 420.

5. Since the feature of churn turbulent flow, present in claim 1 as maintained by the Opposition Division, has been deleted from claim 1 of the fourth auxiliary request and the respondent has not filed an appeal against the decision of the Opposition Division, the question of reformatio in peius arises. Although reformatio in peius should generally be avoided, there is no absolute bar. According to the Enlarged Board of Appeal decision G 1/99 (OJ EPO 2001, 381) the removal of an inadmissible amendment introduced during the
opposition procedure must be compensated by introducing
one or more originally disclosed features which limit
the scope of the patent as maintained, or if that is
not possible, by one or more originally disclosed
features which extend the scope of the patent as
maintained, but within the limits of Article 123(3)
EPC. Only if the latter is also not possible may the
inadmissible amendment be removed without further
limitation. In the present case claim 1 has been
further limited by the additional requirement that the
catalyst particles have an average diameter larger than
30 µm. According to the respondent this limitation has
the effect that in practice, in order to obtain
sufficient fluidisation of the particles, the gas
velocity must be so high that the flow regime is churn
turbulent and thus no question of reformatio in peius
arises. The Board does not dispute that, if the claimed
method is performed in a large scale commercial
reactor, the gas velocity must be so high that a churn
turbulent flow is unavoidable, but is not convinced
that this flow regime must be present in a medium size
demonstration reactor, which is also covered by
claim 1. An allowable amendment which would limit the
flow regime beyond the limits set in claim 1 as
maintained is not available. The appellant's proposal
to define the flow regime as "churn turbulent flow or
bubbly flow" would not limit the scope of the claim as
granted. Moreover, the scope of the present claim would
not be changed if this definition simply means the
absence of plug flow but would introduce an ambiguity
if it would exclude a not originally defined transition
range. Said proposal is therefore neither suitable nor
"possible" in the sense of G 1/99. Claim 1 of the fifth
auxiliary request also does not solve the problem. It
further limits the claim but introduces a choice of
catalyst carriers from a group of carriers, whereby it is at least questionable if such a choice in combination with the other features of the claim can be unambiguously derived from the application as originally filed. Moreover, also the use of catalysts according to claim 1 of the fifth auxiliary request would not limit the flow regime to churn turbulent flow. In the Board's view, therefore, the limitation in claim 1 of the fourth auxiliary request is the most reasonable limitation to compensate for the *reformatio in peius* caused by the deletion of the feature of churn turbulent flow, objected to under Article 123(2) EPC, and is in conformity with the rules laid down in the order of G 1/99.

6. There was dispute between the parties as to the proper construction of claim 1 in view of the dispersion coefficient D defined in parts (b) and (c). Despite the use of the same character "D" the respondent argued that it was evident that the dispersion coefficient in part (c) is different from the dispersion coefficient in part (b). In the Board's view, the patent in suit clearly indicates that D in the formula of part (b) relates to the dispersion of the solid particles (page 6, lines 11 to 12 and line 27) and D in the formula of part (c) to the gas dispersion (page 6, lines 25 to 26 and page 8, lines 6 to 19). They can have the same value at low gas velocities and such a case is illustrated in Example 7 and Figure 6, but there is no indication that the particle and gas dispersion coefficient should in general be the same. The appellant's argument that it was impossible to fluidize the particles to a height of more than 3 metres if the dispersion coefficients of the catalyst particles and the gas are the same is thus not a reason
for insufficient disclosure but rather an additional reason why in part (c) of claim 1, D must have a different meaning than in part (b).

7. Rule 27(1)(e) EPC requires the use of examples where appropriate. The absence of an example of the invention as now claimed is thus no reason for insufficient disclosure if the description as a whole enables the skilled person to perform the invention. The only features which might cause problems in this respect are the dispersion coefficients. As already indicated by the appellant it is not necessary to determine the exact value of the particle dispersion coefficient since the formula in part (b) simply means that the concentration of particles at the top of the liquid in the column is at least 14% of the concentration of the particles at the bottom of the column. There is no evidence that this condition cannot be fulfilled within the constraints of claim 1 according to the fourth auxiliary request.

How to measure the gas dispersion coefficient is explained in detail in the patent in suit (Example 3) and amounts to the determination of the well known Peclet number. Although the exact Peclet number can only be determined in an already existing reactor, the skilled person knows that a column with a high length/diameter ratio has a high Peclet number. Model calculations, such as the Towell and Ackermann correlation, are available to get at least a first impression of the Peclet number of a reactor at a certain gas velocity. There is no evidence that on the basis of the prior knowledge about Peclet numbers and the information given in the patent in suit it would have been an undue burden to build a demonstration
reactor having a Peclet number above 0.2. In the absence of convincing evidence that the process according to claim 1 of the fourth auxiliary request cannot be performed by a team of skilled persons the Board must conclude that the invention as now claimed fulfils the requirements of Articles 83 and 100(b) EPC.

8. None of the cited documents discloses in combination all the features of claim 1 of the fourth auxiliary request (hereinafter simply referred to as claim 1), so that the method according to claim 1 is novel. The novelty of the method of claim 1 has, in fact, not been disputed. The inventive step in the method according to claim 1 has been disputed by the appellant on the basis of D26 as the closest prior art document. D26 discloses F-T processes with a supported cobalt catalyst. The reactor can be chosen from various types well known in the art, for example fixed bed, fluidized bed, ebulliating bed or slurry (column 6, lines 21 to 31). For fluidized bed or slurry reactors the catalyst should be in finely divided form. A typical size analysis is given from which it follows that the average particle size is above 30 μm (column 7, line 52 to column 8, line 4). The experimental work, disclosed in 42 examples, is mainly done with catalysts in a fixed bed. Only in Example 42 an experiment on laboratory scale is performed with a slurry bed. To this end a cobalt catalyst supported on alumina was mixed with a liquid (Synfluid) and loaded into a 1 in. ID by 3 ft. long slurry reactor and a mixture of CO, H₂ and N₂ was fed to the reactor. During the experiment an unstable period was observed due to problems with the temperature control which caused a temporary collapse of the slurry bed; see Example 42 and Figure 3. A gas chromatogram of a sample of the middle distillate and
heavier liquid products is shown in Figure 4. It is indicated that the product is predominantly normal paraffins with a typical Schultz-Flory distribution, showing that the catalyst is promoting the F-T reaction. The Flory-alpha number has not been disclosed. The appellant has tried to calculate the Flory-alpha number on the basis of Figure 4 and came to the conclusion that it must have been about 0.86. The respondent rejected the appellant's calculation and submitted that the carbon distribution shown in Figure 4 is not in agreement with a typical Schultz-Flory distribution. In the Board's opinion the Figures presented by the appellant are not reliable because the baseline of the graph is not visible and, more importantly, the sampling is obscure. With respect to wax production it can only be derived with certainty from Figure 4 that some wax (C21+) has been produced in the experiment according to Example 42. The CO-conversion is also not indicated. For a similar supported cobalt catalyst in a fixed bed reactor a CO conversion of 33% has been disclosed (column 14, Table VIII, Example 8).

9. The appellant submitted that starting from Example 42 of D26 it was obvious to scale up the process, whereby a skilled person, in view of the common general knowledge that the F-T reaction is a first order reaction and that a high Peclet value promotes a first order reaction, would arrive at a process according to claim 1. In the absence of essential product and process information such as Flory alpha number and CO-conversion in the laboratory experiment of Example 42 of D26, however, the Board cannot consider this example as a suitable starting point for assessing the inventive step of an industrial process for producing
wax. The Board rather considers that D4, disclosing an industrial F-T process for producing gasoline and a laboratory scale F-T process for producing wax, represents the closest prior art.

10. D4 discloses a laboratory scale F-T synthesis for the production of wax in a slurry bubble column of 5 cm diameter with a precipitated high alkali iron catalyst. The result was a wax rich product at a CO conversion of >90% (pages 252 to 255 and Table 6). The respondent calculated for the run presented in Table 6 a Flory alpha number >90 and a wax yield >75% (Table 1 of comparative examples filed with the respondent's letter dated 27 January 1999). These results were not contested by the appellant. Starting from the process according to Table 6 of D4, the problem underlying the invention could be regarded as being the provision of a large scale F-T process for producing wax in large quantities at a high yield. The respondent proposed to solve that problem by the combination of process conditions set out in claim 1. According to the comparative examples 2 and 3 of 27 January 1999, F-T synthesis was performed in agreement with present claim 1 in reactors having a diameter of 120 cm and reactor volumes of 11760 and 21390 litre respectively, whereby wax was produced with a yield >75% at a CO- conversion of 80%. The reactions had a Flory alpha number >0.92. The Board is therefore satisfied that the method according to present claim 1 actually solves the above-mentioned problem.

11. Apart from the larger reactor dimensions, the method according to claim 1 differs from the process according to Table 6 of D4 essentially in the use of supported cobalt catalysts with an average particle size > 30 μm.
in combination with a gas dispersion corresponding to a Peclet number ≥ 0.2. Supported cobalt catalysts were known in the art of F-T synthesis at the priority date of the patent in suit but have not generally been used in slurry bubble columns. According to D4, supported catalysts are less suitable for this purpose (page 242). According to D17, a document presented only one month before the priority date of the patent in suit, slurry reactors could be used for wax synthesis, but have not been employed on a commercial scale because the continuous separation and regeneration of the catalyst may be difficult, and erosion of both catalyst and plant may cause problems. Other drawbacks of slurry-bed reactors mentioned in D17 are the need to overcome gas-liquid mass transfer and back mixing, as well as the long lead time and costs associated with the scaling-up of reactors of this type. For these reasons a tubular fixed-bed reactor has been chosen for the F-T reaction for heavy paraffin synthesis (wax); see page 264. Thus at the priority date of the patent in suit there was clearly no incentive for workers in the art to build a large slurry bubble column for F-T wax synthesis. The only prior art document on file disclosing supported cobalt catalyst for F-T synthesis in a slurry-bed reactor is Example 42 of D26, discussed above. For the reasons given, the Board cannot accept this example as a disclosure of a process for the production of wax. For large supported catalyst particles the skilled person would expect problems with the adequate fluidization of these particles. Considering the problems met during the run according to Example 42 and the circumstance that inert gas is added, which enhances fluidization, D26 rather leads the skilled person to doubt about the suitability of supported catalysts in a large scale slurry bed
reactor. If there are already problems with the fluidization of supported catalysts in a laboratory scale reactor the skilled person will hesitate to use them in a large scale reactor. The Board does not dispute the appellant's allegation that fluidization problems encountered for larger particles, such as supported cobalt particles, could be overcome by choosing light weight supports so that the difference in density between the particles and the liquid in which they are dispersed is low. The prior art, however, does not contain any pointer to this possibility. Therefore, this argument is not suitable to demonstrate that a skilled person would have considered using supported cobalt particles, which generally have a much higher density than the wax in which they are dispersed; see Example 8 of the patent in suit. Moreover, light weight catalyst carriers probably do not have the required mechanical strength. For unsupported catalysts disintegration is not a problem and even regarded advantageous but that is not the case for supported catalysts; see D4, page 242.

12. For laboratory scale F-T slurry synthesis, which is generally performed in a long, relatively narrow tube, the Peclet number of the reaction is generally high. For a large scale reactor, in which fluidization is much more of a problem, it is not self evident to chose a reactor and process conditions to reduce the back mixing to such an extent that the Peclet number is higher than 0.2. It is undisputed that the conversion in a reaction with a positive order reaction rate is improved by minimizing the back mixing, ie at a high Peclet number or, in other words, essentially plug flow. It is also undisputed that on a micro scale the F-T reaction has a positive order reaction rate; see
D40, paragraph 8.1.6.1, pages 233 to 234, and D50, pages 34 to 35. The appellant further drew attention to D49, page 23, Table 3, to show that for cobalt catalysts a pressure increase improves the conversion, which implied a positive order reaction rate. Apart from the fact that this document is not prior art, it is to be observed that the Figures in said table relate to calculations based on proposed rate expressions and not on experimental data. The table rather demonstrates how much the theoretical conversion is dependent upon the reaction rate equations used. According to some equations there is hardly any improvement by increasing the pressure from 20 to 50 bar, others predict a remarkable improvement. It cannot be derived from D49 that a skilled person would have known at the priority date of the patent in suit which equation best fits for reactions involving supported cobalt catalysts in a slurry bubble reactor.

D50 discloses kinetic studies during slurry screening tests for several supported cobalt catalysts in laboratory reactors. It was concluded that reactions performed at high H₂/CO feed ratios and high space velocities were best described by a first order rate equation (page 34). It is, however, doubtful whether the results found in D50 for laboratory scale experiments are also valid for a larger scale reactor. D45, published after D50, describing the scaling up of F-T processes using supported cobalt catalysts shortly before the priority date of the patent in suit, neither discloses the order of the reaction rate nor any advantage of a high Peclet number. The only slurry bubble reactor shown in some detail in Fig.4 is unlikely to be used under process conditions involving a Peclet number above 0.2. From Figure 14, showing the conversion as a function of the reactor size based on
simulation modelling, it follows that the conversion is independent of the effective reactor diameter at conversions below 0.9 and that higher theoretical conversions can be obtained at a lower expanded slurry height if the effective reactor diameter is smaller. Interpreting Figure 14 in the context of the patent in suit it can be derived from Figure 14 that a higher Peclet number will probably have a small positive effect on the productivity, but it is doubtful whether, before the publication of the patent in suit, the skilled person would have drawn that conclusion. Anyhow the possible positive effect of a high Peclet number suggested by Figure 14 is so small that it is unlikely that the skilled person would have considered a high Peclet number as an essential feature for a large scale F-T reactor. These observations are supported by Professor Deckwer's declaration of 6 June 2002. The appellant made further reference to D42 disclosing that for a large scale F-T process the reactor should have a height up to about 20 metres and a diameter of 2.5 metres (page 5, right hand column). The Board does not dispute that in a process using such a reactor the conditions might imply a Peclet number above 0.2. The reactor disclosed in D42, however, was designed for a process with unsupported catalyst particles. Process conditions developed for unsupported catalyst particles cannot simply be transferred to a process using larger supported catalyst particles because of their different fluidization characteristics. The Board further considers that even if the skilled person was aware of the relevance of the Peclet number for the F-T synthesis he would have balanced the advantages and disadvantages of a high Peclet number. In the Board's opinion there is insufficient evidence that the possible increase of conversion by a high Peclet number
could have been so substantial that it would outweigh the disadvantages, such as higher construction costs for a higher column and reduced fluidization of the catalyst. Moreover, before the priority date of the patent in suit it was not apparent to the skilled person that it was even possible to fluidize supported cobalt catalyst particles with an average particle size above 30 μm to a degree as required by part (b) of claim 1 at a Peclet number of at least 0.2. The appellant's allegation that by the routine further development of the process as outlined on page 6 of D45 the skilled person would have come to the solution as now claimed has not been substantiated and seems to be based on hindsight. The other citations on file, not discussed during the oral proceedings, do not provide any further argument with respect to inventive step. The Board, therefore, holds that it was not obvious to a person skilled in the art trying to solve the above-mentioned problem to use a supported cobalt catalyst with an average particle size > 30 μm in combination with a slurry bubble column operating with a Peclet number of at least 0.2.

13. For these reasons the Board holds that the subject-matter according to claim 1 of the fourth auxiliary request involves an inventive step. Claims 2 to 8 of this request are sub-claims dependent upon claim 1. The inventive step of their subject-matter follows from this dependency.

The description is not yet in agreement with the amended claims and should be adapted.
Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the department of first instance with the order to maintain the patent with the claims 1 to 8 according to the fourth auxiliary request Alternative B, description and, if necessary, the Figures to be adapted.

The Registrar: 

[Signature]

N. Maslin

The Chairman:

[Signature]

R. Spangenberg

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