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Datasheet for the decision of 18 September 2014

Case Number: T 0840/10 - 3.3.10

Application Number: 04754640.3

Publication Number: 1636153

IPC: C07C2/66, C07C15/073, C07C15/02

Language of the proceedings: EN

Title of invention:
PROCESS FOR THE PRODUCTION OF ALKYLBENZENE WITH ETHANE STRIPPING

Applicant:
Lummus Technology Inc.

Headword:

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

Keyword:
Inventive step - (no) main request
Amendments - added subject-matter (yes) auxiliary requests

Decisions cited:

Catchword:

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Case Number: T 0840/10 - 3.3.10

D E C I S I O N
of Technical Board of Appeal 3.3.10
of 18 September 2014

Appellant: (Applicant)	Lummus Technology Inc. 1515 Broad Street Bloomfield NJ 07003-3096 (US)
Representative:	Brochard, Pascale Osha Liang Sarl 32 Avenue de l'Opéra 75002 Paris (FR)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted on 6 November 2009 refusing European patent application No. 04754640.3 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman	P. Gryczka
Members:	R. Pérez Carlón
	C. Schmidt

Summary of Facts and Submissions

I. The appellant (applicant) lodged an appeal against the decision of the examining division to refuse European patent application No. 04 754 640.3.

II. The documents forming part of the examination proceedings included the following:

D1: US 6,002,057

III. The examining division considered that claim 1 of the then pending main request contained added subject-matter and that none of the processes according to claim 1 of the then pending auxiliary requests 1 to 3 was inventive over document D1, which was the closest prior art.

IV. With the statement setting out the grounds of appeal, the appellant filed a main request and auxiliary requests 1 and 2.

V. The board informed the appellant that it was its preliminary view that all the requests then pending contained added subject-matter, and that it intended to agree with the examining division that the subject-matter claimed was not inventive.

VI. The appellant filed under cover of a letter dated 21 June 2013 a main request and auxiliary requests 1 to 3 replacing the previous requests on file.

VII. The board summoned oral proceedings and informed the appellant that it appeared that all the then pending requests contained added subject-matter.

VIII. The appellant filed under cover of a letter dated 5 February 2014 a main request and auxiliary requests 1 to 3 replacing all previously filed request. Claim 1 of the main request reads as follows:

"*A process for the production of ethylbenzene comprising the steps of:*

a) introducing benzene (F-2) and an ethylene feed (F-1) into a first alkylation reaction zone (110) in the presence of a first alkylation catalyst under first alkylation reaction conditions to produce a first alkylation effluent (118) containing ethylbenzene and a first alkylation overhead stream (111);

b) separating the first alkylation overhead stream (111) into a liquid portion (115) containing benzene and a vapor portion (117) containing unconverted ethylene and ethane;

c) absorbing a major portion of the unconverted ethylene and ethane by contacting the vapor portion (117) of the first alkylation overhead stream with a de-ethanized aromatic substantially ethylene-free lean oil stream (142) containing benzene and ethylbenzene in an absorption zone (130) to produce a rich oil stream (131) containing ethylene and at least some of the ethane;

d) introducing said rich oil stream (131) into a second alkylation reaction zone (140) containing a second alkylation catalyst under second alkylation reaction conditions to produce a first aromatic lean oil stream (141); and,

e) fractionating the first aromatic lean oil stream (141) in a de-ethanizer (190) to produce a de-ethanizer vapor overhead (191) containing a major

portion of the ethane and a liquid bottoms stream (195) containing the de-ethanized aromatic lean oil."

Claim 1 of the first auxiliary request contains, in addition to steps (a) to (e) as in claim 1 of the main request, a step (f) which reads as follows:

"f) feeding at least a portion of the liquid bottoms stream (195) and at least a portion of the benzene (F-2; 165b) as the de-ethanized aromatic substantially ethylene-free lean oil stream (142)."

Claim 1 of the second auxiliary request differs from claim 1 of the main request in that step (d) reads as follows:

"d) introducing said rich oil stream (131) into a second alkylation reaction zone (140) operating adiabatically in a single, liquid phase containing a second alkylation catalyst under second alkylation reaction conditions to produce a first aromatic lean oil stream (141); and,"

Lastly, when compared to claim 1 of the main request, claim 1 of the third auxiliary request includes the feature *"to produce a first aromatic substantially ethylene-free lean oil stream"* in step (d).

IX. The arguments of the appellant relevant for the present decision can be summarised as follows:

Document D1, which was the closest prior art, failed to disclose a process involving steps (c), (d) and a fractionating step carried out in a de-ethanizer as

required by step (e). Claim 1 required that the liquid bottoms of step (e) were directly recycled back into step (c), which necessarily implied that said bottoms were ethylene free. Lastly, D1 was not concerned with the alkylation of diluted ethylene feeds and did not disclose how the second alkylation step was integrated with the first. All these features distinguished the claimed invention from D1.

In the light of document D1, the problem underlying the claimed invention was providing a process for the production of ethylbenzene by which substantially complete conversion of ethylene with a reduced overall amount of required catalyst was achieved. Such an effect, which was the result of carrying out the second alkylation step in a liquid phase, could not have been foreseen having regard to the teaching of the prior art. The process subject-matter of claim 1 of the main request was thus inventive.

Step (f) of claim 1 of the first auxiliary request found a basis in paragraphs [28], alone or in combination with figure 2, [29] in combination with figure 2 and [18] of the application as originally filed.

The feature "a second alkylation reaction zone (140) operating adiabatically in a single, liquid phase" in claim 1 of the second auxiliary request found a basis in paragraph [26] of the application as originally filed.

The feature in claim 1 of the third auxiliary request "to produce a first substantially ethylene free lean oil stream (141)" found a basis in paragraphs [26], [27] and [28] of the application as originally filed.

These auxiliary requests did not thus contain added subject-matter.

X. The appellant informed the board that it would not be represented at the oral proceedings which took place on 18 September 2014.

XI. The appellant requested (in writing) that the decision under appeal be set aside and that a patent be granted in one of the following versions (main and auxiliary requests in this order):

- main request,
- first auxiliary request,
- second auxiliary request,
- third auxiliary request,

all requests filed with letter dated 5 February 2014.

XII. At the end of the oral proceedings, the decision of the board was announced.

Reasons for the Decision

1. The appeal is admissible.

Main request, inventive step (Article 56 EPC):

2. Closest prior art:

2.1 The examining division and the appellant considered that document D1 represented the closest prior art and the board sees no reason to differ.

2.2 The appellant did not dispute that document D1 discloses a process for producing ethylbenzene

comprising steps (a) and (b) as required by claim 1 (see column 4, lines 5 to 56).

Document D1 further discloses reacting the remainder of the unreacted ethylene with pure benzene in a second catalytic distillation tower (column 4, lines 20-22 and 49-58, column 10, lines 29-32, column 16, lines 28-31), and thus also discloses a reaction in a second alkylation reaction zone containing a second alkylation catalyst under second alkylation conditions (see step (d) of claim 1) and a fractionating step (step (e)) since these features are implicitly disclosed by a catalytic distillation tower.

2.3 The process of D1 fails to disclose the following features of claim 1:

- The lean oil of D1 is pure benzene instead of a mixture of benzene and ethylbenzene, as required by claim 1.

- D1 is silent about an absorption step (c) of ethylene in the lean oil in an absorption zone to produce a rich oil stream, which is then introduced into a second reaction zone.

- D1 does not disclose the nature of the feeds obtained by fractionating in the second catalytic distillation tower.

2.4 The appellant argued that document D1 failed to disclose any of steps (c) to (e) of claim 1. However, by disclosing that the unreacted ethylene is recovered by reaction with pure benzene in a catalytic distillation tower, D1 discloses a second alkylation step with an aromatic lean oil. Furthermore, a

catalytic distillation tower discloses, implicitly, a fractionating step.

2.5 The appellant argued that D1 failed to disclose that the separation step (e) was carried out in a de-ethanizer, which should be considered an additional distinguishing feature.

However, the catalytic distillation tower of D1, is a de-ethanizer in the sense of claim 1, i.e. a device capable of removing ethane.

2.6 The appellant argued that the wording of claim 1, according to which the liquid bottoms of step (e) contained "the" de-ethanized aromatic lean oil, necessarily implied that this compound was directly recycled back into step (c) and this could only be possible if it was ethylene free. That the de-ethanized aromatic lean oil was ethylene-free and that it was directly recycled back into step(c) were thus further distinguishing features with respect to D1.

However, claim 1 does not require that the liquid bottoms of step (e) is identical to the lean oil introduced as (142) in step (c), since the use of the wording "comprising" does not exclude the presence of additional separation or purification steps between those required by claim 1. Furthermore, the appellant already acknowledged that the lean oil in stream (142) has a different composition than the lean oil obtained in step (e) (statement of grounds of appeal, page 8, first full paragraph and paragraph [29] of the application).

For these reasons, the argument of the appellant that claim 1 requires that the liquid bottoms of step (e) of

claim 1 is directly recycled into step (c), with the additional consequence that the liquid bottoms of step (e) is a de-ethanized aromatic lean oil does not hold.

2.7 The appellant argued that document D1 was directed to the alkylation of diluted benzene feeds, whereas the claimed invention concerned the alkylation of diluted ethylene feeds, and that should represent an additional distinguishing feature vis-à-vis the prior art.

However, claim 1 relates, in general, to the production of ethylbenzene from ethylene and benzene without any limitation of their concentration and is not thus restricted to dilute ethylene feeds.

In addition, D1 discloses a process using ethylene from a fluid catalytic cracking unit (col. 15, l. 64) which is also an embodiment of the preferred diluted feeds according to the claimed invention (page 6, line 9).

For these reasons, the level of dilution of the ethylene feed used as starting material cannot be considered a distinguishing feature with respect to D1.

2.8 The appellant further argued that document D1 failed to disclose any details on how the second alkylation was integrated with the first, and this should represent a further distinguishing feature vis-à-vis the prior art.

However, claim 1 only requires that the alkylation is carried out, following an absorption step (c), "in a second alkylation reaction zone" containing "a second alkylation catalyst" and "under second alkylation conditions". These features are not more precise than the disclosure of D1 that a second alkylation is carried out in a second catalytic distillation tower

(see point 2.2 above).

2.9 The appellant argued that the optional second alkylation step in document D1 was merely suggested as a side remark and could not represent a reasonable starting point for evaluating inventive step.

However, this argument is not supported by the disclosure of D1 which is not limited to what could be considered as the core of the invention described therein, but also includes all possible embodiments which this document puts at the disposition of the skilled reader, including the optional alkylation step.

3. Technical problem underlying the invention:

The technical problem as defined in the application as originally filed (see page 3, lines 11-13 of the description and point III.1 of the grounds of appeal) is to provide a process for the production of ethylbenzene by which substantially complete conversion of ethylene with a reduced overall amount of required catalyst is achieved.

4. Solution:

The solution proposed by claim 1 of the main request is a process characterised in that it requires a lean oil comprising ethylbenzene, an absorption step of ethylene in the lean oil in an absorption zone and a fractionating step set to lead to specific feeds.

5. Success:

There is no direct comparison on file which could show that the alleged advantage in terms of substantially

complete conversion of ethylene with a reduced overall amount of catalyst compared to the process of the closest prior art D1 had been achieved by the distinguishing features of the invention.

The question nevertheless arises whether even without a comparison with the prior art such an improvement should be recognised, since the alleged effects could be inherently the result of the features distinguishing the claimed process from the closest prior art.

In this respect, the appellant argued that step (c) of mixing the reactants before contacting with the catalyst bed in step (d) led to of a liquid alkylation medium and thus the second alkylation step (d) was necessarily a liquid phase reaction. Said liquid phase reaction was responsible of the substantially complete conversion of ethylene in the second alkylation reaction zone, under mild conditions and with a reduced amount of catalyst so that the problem underlying the claimed invention was solved by the features of claim 1.

The description of the application discloses an improved conversion of 99,9% in connection with a process having a second reactor which operates adiabatically in a single, liquid phase over a fixed bed of loose catalyst of a specific type.

However, claim 1 only requires that the rich oil stream obtained in the absorption step is introduced into the second alkylation reaction zone, but this does not necessarily restrict the alkylation step to a process in liquid phase since claim 1 does not exclude for example further heating the mixture or reducing the pressure in the alkylation reaction zone so that it

also contemplates carrying out the second alkylation reaction in vapour phase.

The appellant itself acknowledged that the combination of two catalytic distillation towers, which is an embodiment of claim 1, could not lead to a conversion higher than 90% (see page 8, lines 1-3 of the statement setting out the grounds of appeal).

The board concludes thus that the alleged advantage in terms of substantially complete conversion over a reduced amount of catalyst has not been achieved by every process according to claim 1 and, hence, that the afore mentioned technical problem is not credibly solved over the whole breadth of the subject-matter of said claim.

6. According to the case law of the Boards of Appeal, alleged but unsupported advantages cannot be taken into consideration in determining the problem underlying the invention (see e.g. decision T 20/81, OJ EPO 1982, 217, Reasons 3, last paragraph). As the alleged improvement in terms of substantially complete conversion over a reduced overall amount of required catalyst lacks the required support, the technical problem as defined above needs reformulation.

7. Reformulation of the technical problem underlying the claimed invention:

Therefore, in view of the teaching of D1, the problem underlying the claimed invention can only be defined as providing an alternative process for the production of ethylbenzene by alkylation of ethylene with benzene.

8. Solution:

The solution proposed by claim 1 of the main request is a process characterised in that it requires a lean oil comprising ethylbenzene, an absorption step of ethylene in the lean oil in an absorption zone, and a fractionating step set to lead to specific feeds.

9. Success:

The board has no doubts that the technical problem as defined in point 7. above has been solved by the process subject-matter of claim 1.

10. Finally, it remains to be decided whether or not the proposed solution to the objective problem underlying the patent in suit is obvious in view of the state of the art:

10.1 Claim 1 relates to a process characterised in that it requires a lean oil comprising ethylbenzene, an absorption step of ethylene in the lean oil in an absorption zone, and a fractionating step set to lead to specific feeds.

10.2 Document D1 discloses a process in which benzene is used as lean oil for absorbing unreacted ethylene and ethane. This mixture is further reacted in a second alkylation step to obtain additional ethylbenzene.

On column 5, lines 22-32, D1 teaches that zeolite beta, which is among the preferred catalyst according to the claimed invention (page 12, line 9), limits the extent of multiple alkylation, i.e. reaction of ethylbenzene with further ethylene to yield polyethylbenzene, which is difficult to separate from the former.

Document D1 further discloses that zeolite beta is a suitable transalkylation catalyst (col. 10, lines 52-53), so that any polyethylbenzene which would be obtained by polyalkylation would react with further benzene to form additional ethylbenzene.

From these passages of D1, the skilled person would extract the information that, at least as long as zeolite beta is used as catalyst, a mixture of benzene and ethylbenzene is a suitable lean oil for absorbing the unconverted ethylene and ethane, since during the subsequent alkylation step zeolite beta minimises any further alkylation of ethylbenzene and promotes the transalkylation of any polyalkylated product which could be formed.

For these reasons, the board concludes that the skilled person would consider a lean oil comprising ethylbenzene as a suitable alternative to the lean oil of D1.

10.3 Document D1 does not explicitly disclose a step of absorbing ethylene in lean oil in an absorption zone before the second alkylation step. D1 only discloses that a second alkylation step is advantageous since it allows recovery of the unreacted ethylene.

D1 provides, nevertheless, a detailed information on the first alkylation step, which is carried out with the same reagents as the second alkylation (ethylene and benzene) and in the same type of reactor (a catalytic distillation tower). In the context of this first alkylation step, D1 discloses introducing the reagents in a catalytic distillation column (col. 7, line 48) below the catalyst bed, thereby allowing

absorption of the ethylene in the lean oil before contacting with the catalyst bed (col. 8, lines 1-3).

Although D1 lacks detailed information with regard to the second alkylation step, the skilled person, trying to obtain a further process to that of D1, would seek the missing information in the details disclosed in the same document with respect to the first alkylation step and assume that the second alkylation step is carried out under the same conditions as the first.

The appellant has alleged that the paragraph on col. 1, lines 1-3 was immediately followed by an alternative embodiment, in which the mixing was not carried out in an absorption zone, and that D1 did not disclose any preference towards any of the two, so that the skilled person had no indication on which one could be suitable for carrying out the second alkylation step.

However, document D1 presents both embodiments as equivalent alternatives, and choosing one of two equally suitable alternatives cannot be considered inventive.

10.4 For the same reason, although D1 is silent about any fractionating step after the second alkylation, the skilled person will consider using the information provided in D1 with respect to the first alkylation step, according to which an overhead vapour containing inter alia ethane is separated from a bottom product (column 16, lines 19-27) which contains ethylbenzene and some benzene (column 16, line 27) which is a de-ethanized lean oil in the sense of claim 1.

10.5 The board concludes for these reasons that the process of claim 1 of the main request is not inventive over

the disclosure of document D1.

10.6 The appellant argued that the effect of the invention was superior than a mere aggregation of two catalytic distillation towers, as proposed by D1 and was, therefore, inventive.

However, for the reasons explained under point 5. above, it has not been credibly proven that any alleged improvement could be obtained over the whole breadth of the claim, precisely since the features of claim 1 do not exclude a process comprising two sequential catalytic distillation towers. These argument is thus unconvincing.

10.7 Since the process of claim 1 of the main request is not inventive, this request is not allowable.

First auxiliary request, amendments:

11. Claim 1 of the first auxiliary request has been amended by adding the feature "feeding at least a portion of the liquid bottoms stream (159) and at least a portion of the benzene (F-2; 165b) as the de-ethanized aromatic substantially ethylene-free lean oil steam (142)".

11.1 With regard to the support of this amendment in the application as originally filed, the appellant argued that according to paragraph [28], a portion 196 of the de-ethanizer bottoms was cycled back to the alkylator 110 (page 13, lines 17-21), and this necessarily implied that the remainder had to be supplied to line 142. This passage provided thus a basis for the amendment of claim 1.

However, this passage fails to disclose any feeding of

the remainder of the de-ethanizer bottoms either to line 142 or to any other point of the plant.

11.2 The appellant has also relied on figure 2 in combination with the passage of paragraph [28] on page 13, lines 17-21 as a basis for the afore-mentioned feature of claim 1.

However, figure 2 fails to disclose features of claim 1 such as the first alkylation column 110, and discloses additional features such as the vent stripper 150, which are not required by claim 1. For this reason, figure 2 cannot provide a basis for the subject-matter of claim 1 of the first auxiliary request.

The appellant argued that one skilled in the art would immediately recognise from figure 2 that some features were merely optional. However, the appellant has not provided any evidence in this respect, and figure 2 does not contain any indication which could show the skilled reader which parts of the process are essential and which ones could be suppressed or regarded as merely optional.

11.3 The appellant also relied on paragraph [29] in combination with figure 2, which discloses (page 14, lines 1-5) that a portion of the benzene is fed into the lean oil as a basis for the features of claim 1.

However, either this passage or figure 2 includes features required by amended claim 1, such as that the benzene comes from feeds F-2 or 165 b. In addition, both passages require additional features not which have not been specified in claim 1 such as the vent stripper 150, the condenser 192 or the cooler 189. For these reasons, paragraph [29] in combination with

figure 2 cannot provide a basis for step (e) of claim 1.

11.4 Lastly, the appellant saw a basis for the feature "feeding a portion of the benzene (F-2)... as lean oil stream (142)" in paragraph [18].

However, said paragraph merely discloses that the fresh benzene feed (F-2) can be fed to "other places in the process that are benzene rich" which is different from feeding said fresh benzene to (142).

11.5 For these reasons, claim 1 includes subject-matter which had not been disclosed in the application as originally filed (Article 123(2) EPC) with the consequence that the first auxiliary request is not allowable.

Second auxiliary request, amendments:

12. Step (d) of claim 1 of the second auxiliary request contains the feature "a second alkylation reaction zone (140) operating adiabatically in a single, liquid phase".

As a basis for this feature, the appellant relied on paragraph [26] of the application as originally filed.

However, this passage discloses an alkylator (140) containing a fixed bed of loose catalyst which operates adiabatically in a single, liquid phase, whereas claim 1 is not limited by such compulsory additional features.

For this reason, claim 1 of the second auxiliary request contravenes the requirements of Article 123(2)

EPC with the consequence that this request is not allowable.

Third auxiliary request, amendments:

13. Claim 1 of the third auxiliary request contains the feature "to produce a first substantially ethylene free lean oil stream (141)". As a basis, the appellant cited paragraphs [26], [27] and [30] of the application as originally filed.

Paragraph [26] discloses a complete conversion of ethylene in connection with a particular type of reactor operation, a specific type of catalyst and defined reaction conditions, which are not features of claim 1 of the third auxiliary request.

Paragraph [27] recites that stream (141) contains benzene, ethylbenzene, ethane and methane; but it fails to disclose that the stream (141) does not contain anything else apart from these components, let alone that it is substantially ethylene-free.

Paragraph [30] discloses an overall ethylene conversion of about 99.9%, and that "very little ethylene is vented from the vent scrubber 150". At least some ethylene remains thus unreacted in stream (141) and needs to be vented, so that this paragraph cannot provide a basis for the feature "to produce a first substantially ethylene free lean oil" required by claim 1.

For these reasons claim 1 of the third auxiliary request does not find a basis in the application as originally filed as required by Article 123(2) EPC with the consequence that this request is not allowable.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

C. Rodríguez Rodríguez P. Gryczka

Decision electronically authenticated