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DECISION
of 28 April 2004

Case Number: T 0074/01 - 3.3.6

Application Number: 92870074.9

Publication Number: 0571701

IPC: C10G 69/12

Language of the proceedings: EN

Title of invention:
Process for the alkylation of aromatics

Applicant:
ATOFINA Research

Opponent:
-

Headword:
Alkylated aromatics/ATOFINA

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive Step - no"

Decisions cited:
-

Catchword:
-

Case Number: T 0074/01 - 3.3.6

DECISION
of the Technical Board of Appeal 3.3.6
of 28 April 2004

Appellant:
ATOFINA Research
Zone Industrielle C
B-7181 Seneffe (Feluy) (BE)

Representative:

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 5 September 2000 refusing European application No. 92870074.9 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: G. N. C. Raths
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 European patent application No. 92 870 074.9 relating to a process for the alkylation of aromatics, on the ground that the subject-matter of the then pending claims lacked an inventive step in view of document

D1 EP-A-0 439 632, in combination with document

D3 GB-A-2 053 959.

In its decision, the Examining Division held that, starting from D1 and facing the technical problem of catalyst deactivation, it was merely a matter of the ordinary skill of an artisan to identify the components which are responsible for the deactivation and to neutralise these components by hydrogenation as was well-known from D3.

II. An appeal was filed against this decision.

III. In its provisional opinion expressed in the communication dated 20 June 2003, the Board of Appeal addressed problems under Articles 123(2), 84, 83 and 56 EPC and referred, *inter alia*, to D1, D3 and

D4 DE-A-1 493 365,

also cited during the examination procedure.

IV. Under cover of the letter dated 1 September 2003, the Appellant (Applicant) filed new claims in a main and in an auxiliary request.

V. In the course of the oral proceedings before the Board of Appeal, held on 28 April 2004, the Appellant filed an amended set of eight claims in a new single request, Claim 1 reading:

"1. In a process for the at least partial liquid phase alkylation and/or transalkylation of diluted aromatic compounds, the steps comprising:

(a) subjecting a diluted aromatic hydrocarbon feedstock consisting of a catalytic reformate containing less than 70 mole% of aromatic compounds and containing at least 1 mole% C_5-C_7 olefins to a selective hydrogenation of the C_5-C_7 olefins in the presence of a palladium on alumina catalyst;

(b) supplying the feedstock resulting from step (a) to a reaction zone containing a zeolite-type aromatic alkylation catalyst;

(c) supplying an alkylation agent comprising C_2 to C_4 olefins to said reaction zone;

(d) operating said reaction zone in at least partial liquid phase at a temperature comprised between 38 and 300°C, a pressure comprised between 0.3 and 7 MPa, a WHSV in terms of grams of aromatic hydrocarbons and olefin per gram of catalyst per hour comprised between 0.5 and 50 hr^{-1} and a molar ratio of aromatics to olefins of at least 3:1 to cause alkylation of said aromatic compounds by said alkylation agent in the presence of said catalyst; and

(e) recovering alkylated aromatic compounds from said reaction zone."

Dependent Claims 2 to 8 relate to specific embodiments of the process of Claim 1.

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

- The closest prior art was represented by D1 which related to the alkylation of pure benzene with pure olefins over a zeolite-type catalyst.

- The subject-matter differed from D1 in that the aromatic feedstock, i.e. the catalytic reformate, as well as the olefinic stream used as alkylation agent, e.g. an FCC off-gas stream were extremely diluted. The benefit of the application in suit over D1 consisted in the direct transformation of two industrial by-products, without any additional purification, into a valuable product at high conversion rate.

- This benefit was achieved by the finding that it was necessary to selectively hydrogenate the C_5-C_7 olefins contained in the catalytic reformate before the alkylation in order to prevent catalyst deactivation.

- At the time of the invention it was not obvious to identify the C_5-C_7 olefins as potential source of problem in the alkylation of diluted aromatic feedstock since D1 dealt only with the alkylation of pure aromatics and D4 was not comparable due to the different kind of catalyst used and was ambiguous in its teaching since it simultaneously claimed to purify the aromatic feedstock from olefins and to react it with C_2 to C_{16} olefins.

VII. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the following points:

Claims 1 to 8 as submitted at the oral proceedings.

VIII. At the end of the oral proceedings the Chairman announced the decision of the Board.

Reasons for the Decision

1. *Amendments (Articles 84 and 123 EPC), sufficiency of disclosure (Article 83 EPC) and novelty (Article 54 EPC)*

1.1 The Board is satisfied that the claims filed during the oral proceedings meet the requirements of Articles 84 and 123(2) EPC and that the claimed subject-matter is novel over the cited prior art (Article 54 EPC).

1.2 Concerning sufficiency of disclosure, the Board has some reservations as to whether the application in suit discloses how to selectively hydrogenate only the C_5-C_7 olefins in embodiments covered by Claim 1 where the catalytic reformate may contain higher and lower olefins, e.g. in the presence of C_4 olefins as in the medium reformate used in Example 1 of the application in suit (Table 1, left-hand column). In the absence of any evidence in this respect, the term "selectively" is, therefore, interpreted to indicate that only olefinic but no aromatic compounds are hydrogenated (see e.g. D3, page 1, lines 55 to 59).

However, no further comments on these matters are necessary in the present case, since the appeal fails on the ground of inventive step.

2. *Inventive Step*

2.1 The application in suit relates to the general technical problem of providing a process for preparing alkylated aromatic compounds from an aromatic hydrocarbon feedstock and an olefinic stream under at least partial liquid phase conditions and in the presence of a zeolite-type alkylation catalyst (page 2, lines 1 to 29 of the application as published; in the following, references will be constantly made to the application as published).

In the application in suit, two known processes for preparing alkylated aromatic compounds are identified, namely

- a process where benzene and diluted ethylene are reacted in the vapour phase over a zeolite-type catalyst (page 2, lines 17 to 22), and

- a process, known from D1, where benzene is alkylated with pure C_2-C_4 olefins in at least partial liquid phase over a zeolite-type catalyst (page 2, lines 23 to 29).

2.2 The Board agrees with the Appellant that D1 is a suitable starting point for the assessment of inventive step since it is concerned with the same type of process as the application in suit, comprising alkylation in at least partial liquid phase.

This document discloses the alkylation of a pure aromatic hydrocarbon stream, such as benzene, with C_2-C_4 olefins over the zeolite-type catalyst at a temperature of 100 to 600°F (i.e. about 38 to 316°C), at a pressure of 50 to 1000 psig (i.e. about 0.35 to 7 MPa) while maintaining at least a partial liquid phase, at a WHSV in terms of grams of aromatic hydrocarbon and olefin per gram of catalyst per hour of 0.5 to 50 and at a molar ratio of aromatics to olefins of preferably at least 4:1 (page 4, lines 33 to 38 and page 4, line 56 to page 5, line 7).

Contrary to the opinion of the Appellant, D1 further suggests to use the olefins in diluted form, e.g. with paraffins, small amounts of water or nitrogen compounds (page 4, lines 38 to 42).

The process of Claim 1 differs therefore from that disclosed in D1 only in that - instead of using pure aromatic hydrocarbon - a diluted aromatic hydrocarbon feedstock is used, namely a catalytic reformate which contains less than 70 mole% of aromatic compounds and at least 1 mole% of C_5-C_7 olefins and that the olefins are hydrogenated in the presence of a palladium on alumina catalyst prior to the alkylation reaction.

2.3 According to the application in suit, it would be advantageous if there existed a process for alkylating the benzene present in light reformate with a diluted stream of C_2-C_4 olefins, such as FCC offgas which is normally used as fuel gas (page 2, lines 5 to 10 in combination with lines 13 to 16).

The Board agrees that it is certainly beneficial if, as explained by the Appellant, a valuable product can be derived directly from industrial by-products without the need for purification. Therefore, the technical problem to be solved in view of D1 may be seen in providing a process for preparing C_2-C_4 alkylated aromatic compounds from a hydrocarbon feedstock containing aromatic compounds without any need for isolating or purifying the aromatic compounds in advance.

2.4 According to Claim 1 this problem is solved by using a catalytic reformate as the feedstock and subjecting it to a selective hydrogenation of the C_5-C_7 olefins contained in an amount of at least 1 mole%.

2.5 The Appellant argued that it was the merit of the application in suit to identify those components contained in the reaction mixture which are responsible for the deactivation of the catalyst and the low conversion into the desired products.

2.6 It is apparent from Example 1 and Example 2 of the application in suit, the latter being a comparative example, that the amount of C_2-C_4 alkylated aromatics is heavily reduced if the reaction mixture contains C_5 and C_6 olefins in addition to the C_2-C_4 olefins during alkylation. It is shown that the selectivity for C_2-C_4 alkylated aromatic compounds is below 70% in the process of Example 2, where the reaction mixture contains 1.22 wt% of C_2-C_4 olefins and 1.15 wt% of C_5 and C_6 olefins, as compared with more than 98% in the process of Example 1 where the reaction mixture contains only C_2-C_4 olefins in an amount of 1.76 wt% (Tables I to IV).

Examples 1 and 2 of the application in suit, in particular, show that in the presence of C_5 and C_6 olefins considerable amounts of C_5 and C_6 alkylated products are obtained at the expense of the desired C_2-C_4 alkylated aromatic compounds (Table IV).

It is, therefore, credible that the above stated technical problem in view of D1 is actually solved by the claimed process.

2.7 It remains to be assessed whether, in view of the cited prior art documents, it was obvious for someone skilled in the art to solve this problem by the means claimed.

2.8 The Appellant argued that those skilled in the art would have expected the process of D1 to be also feasible with catalytic reformate instead of pure benzene since there was no reason to assume that the zeolite-type catalyst would not be selective with respect to C_2-C_4 alkylation.

However, a process for the alkylation of a diluted aromatic feedstock is already known from D4 (page 1, line 8 to page 2, line 12), where it is emphasized that the feedstock must be free of olefins before the alkylation even though a mixture of C_2-C_{16} olefins, pure or diluted with paraffins, is later on used for alkylation. It is recommended to remove the olefins via hydrogenation (page 2, third and fourth paragraph, page 3, second and third paragraph, Example 1 and Claims 1 to 3).

The hydrogenation catalyst is not mentioned in D4, but it is known from D3 that palladium on alumina is a suitable catalyst for selective hydrogenation of olefins in the presence of aromatic hydrocarbons, e.g. in a catalytic reformate (page 1, lines 20 and page 2, lines 6 to 15, 24 to 26 and 43).

2.9 The Appellant argued that D4 related to the old alkylation technology using a Friedel-Crafts catalyst which is environmentally harmful and no longer allowed to use.

Therefore, a totally new development on the basis of zeolite-type catalyst, e.g. represented in D1, has taken place in the art. A skilled person would not have considered D4 at all in order to develop the new technology, in particular since the reactions over zeolite-type catalysts and Friedel-Crafts catalysts were not comparable.

The Board is not convinced by this argument since, as eventually agreed by the Appellant, it is common general knowledge of those skilled in the art that olefins are a reactive group of compounds and suitable for catalytic alkylation of aromatics. The skilled person has, therefore, to expect that in the presence of a mixture of different olefins, not only specific olefins might undergo reaction unless there is a reason to assume that the particular catalyst used is selective with respect to those specific olefins.

However, no evidence has been submitted in this respect. Therefore, a person skilled in the art has no reason to assume that zeolite-type catalysts would be more selective for the desired C_2-C_4 alkylation reaction than the Friedel-Crafts catalyst. Rather on the contrary, the skilled person would derive from the use of pure benzene according to D1 that undesired reactions are not excluded if diluted benzene was used.

2.10 Finally, the Appellant argued that the teaching in D4 to purify the aromatic feedstock from olefins on the one hand and to react it with C_2-C_{16} olefins on the other hand was ambiguous. D4 did not, therefore, teach away from the presence of C_5-C_7 olefins in the aromatic feedstock to be used for alkylation.

In fact, D4 teaches that C_2-C_{16} olefins are suitable as alkylation agents in the presence of a Friedel-Crafts catalyst (Claim 1). However, it is shown in Example 7, aiming at the production of propylbenzene, that benzene is lost for the desired product due its reaction with higher olefins, if these are not to be removed beforehand from the aromatic hydrocarbon feedstock.

Therefore, the correct understanding of the teaching of D4 is, in the Board’s opinion, that selective alkylation with specific olefins is only possible if any undesired olefins contained in the diluted feedstock are removed in advance in order to prevent their reaction with the aromatic compounds.

2.11 The Board has not overlooked that the amounts of aromatic compounds (less than 70 mole%) and C_5-C_7 olefins (at least 1 mole%) contained in the catalytic reformate according to Claim 1 of the application in suit are not specifically mentioned in the prior art. However, no particular effects due to these amounts are derivable from the application in suit or the prior art. Nor has the Appellant provided evidence or arguments in this regard. Therefore, no inventive merit can be attributed to these features.

2.12 For these reasons, the Board concludes that it was obvious for the skilled person to use in the process of D1 a diluted aromatic hydrocarbon feedstock such as a catalytic reformate and to selectively hydrogenate the olefins contained therein prior to the alkylation, as suggested in D4, over the hydrogenation catalyst specifically disclosed in D3 for that purpose, in order to provide in view of D1 a process for preparing C_2-C_4 alkylated aromatic compounds from an impure aromatic hydrocarbon feedstock without any need for isolating the aromatic compounds.

Consequently, the subject-matter of Claim 1 is not based on an inventive step and does not meet 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 G. Raths