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**Datasheet for the decision  
of 13 April 2010**

**Case Number:** T 1484/07 - 3.3.10

**Application Number:** 01915341.0

**Publication Number:** 1263709

**IPC:** C07C 67/38

**Language of the proceedings:** EN

**Title of invention:**

Process for the carbonylation of ethylenically unsaturated compounds

**Patentee:**

Shell Internationale Research Maatschappij B.V.

**Opponent:**

Lucite International UK Limited

**Headword:**

Carbonylation of olefins/SHELL

**Relevant legal provisions:**

EPC Art. 84, 54, 56

**Relevant legal provisions (EPC 1973):**

-

**Keyword:**

"Novelty (yes) - generic disclosure not novelty destroying for specific - example from prior art cannot be modified by combination with description"  
"Inventive step (yes) - non-obvious alternative"

**Decisions cited:**

G 0009/91, T 0332/87, T 0472/88, T 0800/91, T 0068/95,  
T 0420/00

**Catchword:**

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Case Number: T 1484/07 - 3.3.10

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.10  
of 13 April 2010

**Appellant:** Lucite International UK Limited  
(Opponent) Queens Gate  
15-17 Queens Terrace  
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Hampshire S014 3BP (GB)

**Representative:** Walsh, David Patrick  
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15 Clare Road  
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**Respondent:** Shell Internationale Research Maatschappij  
(Patent Proprietor) B.V.  
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NL-2596 HR Den Haag (NL)

**Decision under appeal:** Interlocutory decision of the Opposition  
Division of the European Patent Office posted  
4 July 2007 concerning maintenance of European  
patent No. 1263709 in amended form.

**Composition of the Board:**

**Chairman:** R. Freimuth  
**Members:** J. Mercey  
D. S. Rogers

## Summary of Facts and Submissions

- I. The Appellant (Opponent) lodged an appeal on 4 September 2007 against the interlocutory decision of the Opposition Division posted on 4 July 2007 which found that European patent No. 1 263 709 in amended form met the requirements of the EPC.
- II. Notice of Opposition had been filed by the Appellant requesting revocation of the patent as granted in its entirety on the grounds of lack of novelty and inventive step (Article 100(a) EPC) and insufficient disclosure (Article 100(b) EPC). *Inter alia* the following documents were submitted in opposition proceedings:
- (1) WO-A-01 105 51,
  - (2) WO-A-96 194 34,
  - (3) WO-A-98 427 17 and
  - (5) WO-A-99 218 20.
- III. The Opposition Division held that the subject-matter of the then pending main request was not novel over the disclosure of document (1) and that the amendments leading to the then pending first auxiliary request did not comply with the requirements of Article 84 EPC. It further held that the invention was sufficiently disclosed and that the subject-matter of the second auxiliary request was novel and involved an inventive step, document (3) being considered to represent the closest prior art. In the light of this prior art, the problem to be solved by the invention was seen as the provision of a process for the carbonylation of internally ethylenically unsaturated compounds yielding

products with regioselectivity towards a linear product. None of the cited prior art documents suggested modifying the catalyst ligand and the use of a solvent in order to solve said problem. Claim 1 of the then second auxiliary request read as follows:

"Process for the carbonylation of internally ethylenically unsaturated compounds having from 4 to 20 carbon atoms by reaction with carbon monoxide and a hydroxyl group containing compound in the presence of a catalyst system including:

(a) a source of palladium cations;

(b) a bidentate diphosphine of formula I,



wherein P represents a phosphorus atom;  $R^1$ ,  $R^2$ ,  $R^5$  and  $R^6$  independently represent the same or different optionally substituted organic groups containing a tertiary carbon atom through which the group is linked to the phosphorus atom;  $R^3$  and  $R^4$  independently represent optionally substituted alkylene groups and R represents an optionally substituted aromatic group;

(c) a source of anions derived from an acid having a pKa less than 3, as measured at 18°C in an aqueous solution; carried out in the presence of an aprotic solvent."

IV. The Appellant argued that the subject-matter of claim 1 of the claims as maintained was not clear, because the nature of the species actually carbonylated was unclear, since internally unsaturated olefins could not give a linear product.

The subject-matter was not novel over the disclosures of both documents (1) and (5), Example 19 of document

(1) disclosing all the features of the invention apart from an internal olefin having from 4 to 20 carbon atoms, C<sub>1</sub> to C<sub>4</sub> olefins being however disclosed on page 5 of the description, document (5) being considered to be equally relevant.

The Appellant further submitted that the claimed subject-matter was not inventive over document (5) either alone or in combination with document (3). Document (5) disclosed the carbonylation of olefins of formula C<sub>n</sub>H<sub>2n</sub> with a high turnover number, the only difference to the claimed process being the express use for internally unsaturated olefins. The skilled person would, however, have applied the teaching of document (5) to internal olefins with the expectation of achieving a high turnover number, selectivity to the linear product being merely a bonus effect. With a letter dated 9 April 2010, the Appellant submitted three further documents to counter the Respondent's arguments based on higher esters not forming azeotropes (see point V below) that the skilled person would not have taken document (5) into account. Furthermore, the skilled person would have expected that by applying the reaction of document (5) to an internally unsaturated olefin, linear products would be obtained, since it was known that internal olefins isomerised to give the terminal olefin and that terminal olefins were more reactive than internal olefins. The Appellant filed documents (12) to (14):

- (12) Applied Homogeneous Catalysis with Organometallic Compounds, VCH, Vol. 2, 1996, pages 980 to 986,
- (13) Journal of Molecular Catalysis 94 (1994), pages 7 to 17 and

(14) Journal of Molecular Catalysis A: Chemical 123,  
(1997), pages 91 to 101

to support this argument. Furthermore, document (3) taught the carbonylation of internally unsaturated olefins giving high linearity of products using catalyst ligands having a covalent bridging group,  $R^2$ , said group embracing the bridging group R of the ligands in contested claim 1. With its letter of 14 November 2007, the Appellant also submitted experimental data, namely amended Annexes 2 and 3 which had originally been filed on 22 May 2007 before the Opposition Division. At the time it was first filed, the purpose of Annex 3 was to demonstrate lack of sufficiency of disclosure under Article 100(b) EPC. Annex 2, now having been corrected, showed that not all aprotic solvents resulted in improvements *vis-à-vis* the process of document (2) and Annex 3, now having been amended, showed that when in the catalyst ligand of formula (I) in claim 1 of the claims as maintained the group R was methyl-substituted, or the groups  $R^3$  or  $R^4$  were ethylene or substituted methylene, little or no carbonylation took place, such that the process could not be inventive over the whole scope of the claims.

V. The Respondent (Proprietor of the patent) submitted that the feature objected to under Article 84 EPC, namely "internally unsaturated" emanated from granted claim 10, and was thus not open to objection in the opposition proceedings. In any case, it was evident that the skilled person would clearly understand what was meant thereby.

The claimed subject-matter was novel over both of documents (1) and (5), since neither of these documents disclosed the carbonylation of internally unsaturated olefins having from 4 to 20 carbon atoms, only ethylene being exemplified therein.

With regard to inventive step, the Respondent submitted that since document (5) related to a process for the carbonylation of an olefin and an alkyl alcohol to form an alkyl alkanoate, characterised in that the ratio of alkyl alcohol to alkyl alkanoate was greater than the ratio of alkyl alcohol to alkyl alkanoate in an azeotropic mixture thereof, it was evident that the skilled person would only apply this teaching if the alkyl alcohol and alkyl alkanoate formed an azeotrope. Since, however, no azeotropes would appear to be known for methyl or ethyl pentanoates, the teaching of document (5) extended only to the carbonylation of ethylene. Even if the skilled person would have consulted said document, it did not suggest the carbonylation of internally unsaturated C<sub>4</sub>-C<sub>20</sub> olefins, let alone with selectivity to the linear product. With regard to the Appellant's objections based on document (3), the Respondent concurred with the argumentation and conclusions in the contested decision in this respect. The claimed subject-matter was thus inventive. In response to the experimental data provided by the Appellant, the Respondent filed document (15):

(15) WO-A-2007/119079

said patent application emanating from the Appellant itself and teaching that substituted aromatic diphosphines were indeed active carbonylation catalysts

for *inter alia* internally unsaturated olefins in the presence of a wide range of aprotic solvents.

- VI. The Appellant requested that the decision under appeal be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed.

- VII. Oral proceedings were held on 13 April 2010 in the absence of both the Respondent and the Appellant, who, after having been duly summoned, informed the Board that they would not attend. At the end of the oral proceedings, the decision of the Board was announced.

### **Reasons for the Decision**

1. The appeal is admissible.
2. *Amendments (Article 123(2) and (3) EPC)*

Claim 1 is based on original claim 1, together with original claim 10 (which corresponds to granted claim 10) and page 7, line 31 of the application as filed. The amendments made to the claims as maintained by the Opposition Division were not objected to by the Appellant, nor does the Board see any reason to question their allowability under Article 123(2) and (3) EPC of its own motion.

3. *Sufficiency of Disclosure (Article 100(b) EPC)*

The appealed decision found the invention to be sufficiently disclosed (cf. point III supra).

Sufficiency of disclosure was no longer contested during the appeal proceedings, nor does the Board see any reason to take a different view to the Opposition Division. Hence, it is unnecessary to go into more detail in this respect.

4. *Clarity (Article 84 EPC)*

4.1 Claim 1 was amended during opposition proceedings *inter alia* by specifying that the ethylenically unsaturated compounds to be carbonylated are "internally" unsaturated, said amendment resulting from a combination of claim 1 with dependent claim 10 as granted. The Appellant submitted that as a result thereof the subject-matter of claim 1 did not fulfil the requirements of Article 84 EPC, because the nature of the species actually carbonylated was unclear, since internally unsaturated olefins could not give rise to a linear product. The Respondent argued that the amended claims should not be open to an objection under Article 84 EPC, since the amendment resulted from a combination with dependent claim 10 as granted.

4.2 However, according to the jurisprudence of the Boards of Appeal the Board has the power and the duty to examine whether the patent as amended satisfies all the requirements of the EPC, as long as the objections **arise** out of the amendments made thereto. Thus, an amendment directly giving rise to an alleged ambiguity under Article 84 EPC should be dealt with by the Board under the power given by Article 101(3) EPC 2000 (Article 102(3) EPC 1973) (see G 9/91, OJ EPO 1993, 408, point 19 of the reasons), since in this context the term "arise" is to be construed broadly, including any

case where the amendment clearly brings into focus an ambiguity that may have existed all along (see decision T 472/88, point 2 of the reasons, not published in OJ EPO). This principle applies also when a feature of a dependent granted claim is incorporated into an independent claim thereby making visible that ambiguity in the amended independent claim (see decision T 420/00, point 3.6.3 of the reasons, not published in OJ EPO). Thus, in the present case, the examination of claim 1 for clarity is limited to identifying those unclarities concealed in claim 10 that become apparent once the feature of claim 10 is incorporated into independent claim 1.

- 4.3 However, the Appellant has not submitted any arguments as to why the introduction of the feature "internally" from granted claim 10 into claim 1 generates ambiguity which was previously concealed in claim 10. In any case, the Board holds that the subject-matter of claim 1 is clear, since the features required for defining a process, namely the educts and physical steps to be performed, are clearly defined. Which species actually reacts is a technical insight into the reaction which is, however, not part of the subject-matter claimed. Thus, claim 1 fulfils the requirements of Article 84 EPC.

5. *Novelty*

- 5.1 The Appellant has challenged the novelty of the claimed invention with regard to both documents (1) and (5), document (1) being comprised in the state of the art at least according to Article 54(3) EPC, since it has a

priority date of 4 August 1999, the patent in suit claiming an earliest priority of 14 March 2000.

- 5.2 Document (1) discloses in Example 19 a process for the carbonylation of ethylene. The Appellant based its objection of lack of novelty in the oral proceedings before the Opposition Division on a combination of this example with the general part of the description, arguing that Example 19 could be modified by replacing ethylene with a C<sub>4</sub>-olefin disclosed on page 5, line 26 of the application as filed.

In this context, the Board firstly notes that according to the established case law of the Boards of Appeal regarding the examination of novelty, the teaching of a document is not confined to the detailed information given in the examples, but embraces the whole disclosure of that document (see decision T 332/87, point 2.2 of the reasons, not published in OJ EPO). Nevertheless, the general principle consistently applied by the Boards of Appeal for concluding lack of novelty is that there must be a direct and unambiguous disclosure in the state of the art which inevitably leads the skilled person to subject-matter falling within the scope of what is claimed.

In the present case, there is no specific disclosure in document (1) to combine Example 19 with this particular part of the description and thus the skilled reader of document (1) does not have any indication to select a particular olefin from the generic disclosure of the description, which also indicates other olefins to be equally suitable, and to combine it with the particular process of Example 19, this example disclosing nothing

more than the reaction of the particular combination of reactants employed therein.

Thus, the process of claim 1 is not directly and unambiguously disclosed in document (1).

5.3 Document (5) discloses in claim 1 a process for the carbonylation of alkenes of formula  $C_nH_{2n}$ .

However, although comprised within the general disclosure of document (5), there is no specific disclosure of an internally ethylenically unsaturated compound having from 4 to 20 carbon atoms, the only olefin being disclosed in said document being the  $C_2$ -olefin, ethylene.

The subject-matter of claim 1 is therefore not anticipated by document (5).

## 6. *Inventive step (Article 56 EPC)*

6.1 According to the established jurisprudence of the Boards of Appeal it is necessary, in order to assess inventive step, to establish the closest state of the art, to determine in the light thereof the technical problem which the invention addresses and successfully solves, and to examine the obviousness of the claimed solution to this problem in view of the state of the art. This "problem-solution approach" ensures assessing inventive step on an objective basis and avoids an *ex post facto* analysis.

6.2 The present invention is directed to a carbonylation process with high regioselectivity towards a linear

product (see paragraph [0007] of the patent in suit). A similar process is disclosed in document (3), cited in paragraph [0004] of the patent in suit, which discloses in claims 11 to 14 a process for the carbonylation of *inter alia* internal ethylenically unsaturated olefins having 2 to 22 carbon atoms by reaction with carbon monoxide and a coreactant which may be an alcohol in the presence of, for example, sulphonic acids (see page 5, line 20) and a catalyst obtainable by combining *inter alia* a palladium cation with a diphosphine of formula  $R^1>P-R^2-PR^3R^4$  according to claim 1, wherein  $R^2$  is a covalent bridging group and  $R^1$  is a bivalent radical that together with the phosphorus atom to which it is attached is an optionally substituted 2-phosphatrimethylcyclo[3.3.1.1{3,7}]decyl group and  $R^3$  and  $R^4$  are univalent radicals of up to 20 atoms or jointly form a bivalent radical of up to 20 atoms. Said process may be considered to be carried out in the presence of an aprotic solvent, since, as submitted by the Appellant, the starting olefin would act as such a solvent. Examples 6 and 7 of document (3) illustrate said process for an internal- $C_{14}$  olefin and methyl 3-pentenoate to give 78% and 84% selectivity into linear methyl esters, respectively, using 1,3- $P,P'$ -di(2-phospho-1,3,5,7-tetramethyl-6,9,10-trioxatrimethylcyclo[3.3.1.1{3.7}-decyl)propane (DPA3) as the diphosphine ligand.

- 6.2.1 Where the patent in suit indicates a particular piece of prior art as the starting point for determining the problem underlying the patent in suit, in the present case document (3) in paragraph [0004] of the patent specification, then the Board should adopt this as the starting point for the purpose of a problem-solution

analysis unless it turns out that there is closer state of the art of greater technical relevance (see e.g. decisions T 800/91, point 6 of the reasons; T 68/95, point 5.1 of the reasons, neither published in OJ EPO).

The Appellant also addressed document (5) as closest prior art. However, the disclosure of document (5) does not address the problem of product linearity, internal olefins not being explicitly addressed therein, ethylene being the only olefin specifically taught (see point 5.3 above). In contrast, document (3) (see page 1, lines 18 to 19, page 7, lines 16 to 18 and Examples 6 and 7) specifically addresses problems associated with internal unsaturation and teaches that the catalyst disclosed therein is particularly advantageous in the conversion of internal olefins

Thus, the Board considers, in agreement with the Respondent and the Opposition Division, that in the present case the process of document (3) represents the closest state of the art and, hence, takes it as the starting point when assessing inventive step.

6.3 In view of this state of the art, the problem underlying the patent in suit, may be regarded as the provision of an alternative process for the carbonylation of internally ethylenically unsaturated compounds yielding products whilst maintaining a high regioselectivity to a linear product.

6.4 As a solution to this problem, the patent in suit proposes the process using specific diphosphine ligands as defined in claim 1, wherein the covalent linking group R is a 1,2-aryl group and  $R^1$ ,  $R^2$ ,  $R^5$  and  $R^6$

independently represent the same or different optionally substituted organic groups containing a tertiary carbon atom through which the group is linked to the phosphorus atom.

- 6.5 In view of the results given in Table I, and in particular, in Table III, of the specification of the patent in suit, which show high product linearity, it is credible that the problem underlying the patent in suit has been successfully solved.
- 6.5.1 The Appellant submitted that the problem had not, however, been solved over the whole scope of the claims, since in view of the experimental data in Annex 3 (see point IV above), the process of claim 1 lacked inventive step insofar as in the ligands of formula (I), the group R was substituted or the groups R<sup>3</sup> or R<sup>4</sup> were other than methylene.
- 6.5.2 However, Annex 3 was originally filed during opposition proceedings to demonstrate lack of sufficiency of disclosure under Article 100(b) EPC, said ground for opposition no longer being at stake in the appeal proceedings (see point 3 above). As such, these experimental data do not comprise a comparison with the closest prior art, namely the process according to document (3), the experiments having been designed for a different purpose. Therefore, these data are not suitable for supporting an objection under Article 56 EPC.

Furthermore, these experiments show that under specific operating conditions and with a specific olefin as starting material, two specific ligands result in no

product at all, the Appellant concluding herefrom that the process was not inventive for all processes according to claim 1 wherein the group R was substituted or the groups R<sup>3</sup> or R<sup>4</sup> were other than methylene. However, such bidentate diphosphine ligands wherein the corresponding aromatic group, R, is substituted by a wide range of substituents, including methyl, are disclosed in document (15) (see pages 103 to 111), said patent application having been filed by the Appellant itself, and are described therein as being suitable for the carbonylation of internally unsaturated olefins with a high selectivity towards the linear product (see page 112, lines 28 to 29, 34 to 35 and 39), the Appellant not having contested said teaching. Due to these inconsistencies between the Appellant's findings in document (15) and Annex 3, the Board holds that it is not plausible that such ligands would not result in any product at all, with the consequence that the results of Annex 3 do not convince the Board.

With regard to the experimental data in Annex 2, this allegedly shows that not all aprotic solvents result in an improvement *vis-à-vis* the process of document (2). However, firstly, the use of an aprotic solvent does not form part of the characterising feature of the solution to the problem underlying the present invention (see point 6.4 above) and, secondly, any comparison with the process of document (2) is irrelevant in the present assessment of inventive step, since said document does not represent the closest prior art (see point 6.2 above).

- 6.5.3 Thus, neither the data of Annex 2 or 3 convincingly show that the problem underlying the present invention has not been solved.
- 6.6 Finally, it remains to be decided whether or not the proposed solution to this problem is obvious in view of the cited prior art, the Appellant arguing that the claimed subject-matter was not inventive exclusively on the basis of the combination of the teachings of documents (5) and (3).
- 6.6.1 However, document (5) does not address the problem of product linearity, which is not surprising, because internal olefins are not explicitly addressed therein, nor are any olefins other than ethylene specifically taught, such that the problem of product linearity does not arise (see point 6.2.1 above). Thus, document (5) provides no incentive for the skilled person to substitute the 2-phospha-tricyclo[3.3.1.1{3,7}]decyl ligand of document (3) with the ligands of formula (I) according to present claim 1 when seeking an alternative process for the carbonylation of internally ethylenically unsaturated compounds yielding products whilst maintaining a high regioselectivity to a linear product, i.e. for solving the problem underlying the invention.
- 6.6.2 Hence, the Respondent's argument that the skilled person would not even have considered document (5) at all when seeking a solution to the problem underlying the patent in suit, in view of its teaching being restricted to the case when the alkyl alcohol and alkyl alkanoate formed an azeotrope, is irrelevant, as are the documents (12) to (14) filed by the Appellant with

a view to counteracting this argument of the Respondent (see point IV above), since the claimed process is in any event inventive over the combination of the teachings of documents (5) and (3).

6.7 For the following reasons the Board cannot accept the Appellant's arguments designed to show a lack of inventive step.

6.7.1 The Appellant submitted that internally unsaturated olefins would isomerise in the carbonylation reaction such that both internally and terminally unsaturated olefins would exist in equilibrium in the reaction mixture, terminal olefins being more reactive, such that when starting from an internally unsaturated olefin, the skilled person knew that linear products were likely, documents (12) to (14) being cited in support of this argument.

However, regardless of how internally unsaturated olefins isomerise in such a reaction, there is no teaching in any of the cited documents that the particular ligands of document (5) would result in the high selectivity to the linear product as achieved in document (3) and in the Examples of the patent in suit, the only specific olefin described in document (5) being ethylene, wherein the question of product linearity does not arise.

6.7.2 With regard to the Appellant's submission that the high selectivity was a mere bonus effect, a high turnover number for the reaction already being taught by document (5), the Board holds that a high turnover number does not automatically lead to high selectivity

as required for a "bonus effect". Indeed, the Appellant itself does not in fact allege that the one leads automatically to the other, but rather that selectivity was only secondary to turnover number, a low selectivity being possibly still viable when the turnover number was high. However, the objective problem underlying the patent in suit comprises maintaining a high selectivity (see point 6.3 above), the Appellant's arguments being thus unpersuasive.

6.8 For these reasons, the Board concludes that the method for the carbonylation of internally ethylenically unsaturated compounds according to claim 1 and dependent claims 2 to 9 involves an inventive step within the meaning of Articles 52(1) and 56 EPC.

## **Order**

### **For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

D. Magliano

R. Freimuth