

Veröffentlichung im Amtsblatt	Ja/Nein
Publication in the Official Journal	Yes/No
Publication au Journal Officiel	Oui/Non



Aktenzeichen / Case Number / N<sup>o</sup> du recours : T 38/84

Anmeldenummer / Filing No / N<sup>o</sup> de la demande : 81 200 269.9

Publikations-Nr. / Publication No / N<sup>o</sup> de la publication : 0 036 233

Bezeichnung der Erfindung: "Process for the preparation of benzene-monocarboxylic  
Title of invention: acids"  
Titre de l'invention :

### ENTSCHEIDUNG / DECISION

vom / of / du 5 April 1984

Anmelder/Patentinhaber: STAMICARBON B.V.  
Applicant/Proprietor of the patent:  
Demandeur/Titulaire du brevet :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 52(1), 56

"Inventive Step - Importance of the technical problem  
- Small improvement of commercially used process"

#### Leitsatz / Headnote / Sommaire

The achievement of a numerically small improvement of a process commercially used on a large scale (here enhanced yield of 5%) represents a worthwhile technical problem which must not be disregarded in assessing the inventive step of its solution as claimed.

Europäisches  
Patentamt

Beschwerdekammern

European Patent  
Office

Boards of Appeal

Office européen  
des brevets

Chambres de recours



Case Number: T 38 / 84

**DECISION**  
of the Technical Board of Appeal 3.3.1  
of 5 April 1984

**Appellant:** STAMICARBON B.V.  
Postbus 10  
NL-6160 MC Geleen

**Representative:** Hoogstraten, Willem Cornelis Roeland  
OCTROOBUREAU DSM  
Postbus 9  
NL-6160 MA Geleen

**Decision under appeal:** Decision of Examining Division 003 of the European Patent  
Office dated 21 September 1983 refusing European patent  
application No 81 200 269.9 pursuant to Article 97(1)  
EPC

**Composition of the Board:**

**Chairman:** D. Cadman  
**Member:** K. Jahn  
**Member:** O. Bossung

SUMMARY OF FACTS AND SUBMISSIONS

- I. European Patent Application No. 81 200 269.9 filed on 11 March 1981 and published on 23 September 1981 under publication No. 0 036 223, claiming the priority of the Netherland prior application of 15 March 1980, was refused by decision of the European Patent Office dated 21 September 1983 on the basis of 7 claims of which claim 1 has the following wording:

"Process for the preparation of a benzene-monocarboxylic acid by oxidation of a monoalkyl-benzene compound in the liquid phase with the aid of a gas containing molecular oxygen, in the presence of a catalyst composed of a cobalt and a manganese compound, both of which are soluble in the reaction mixture, characterized in that a manganese: cobalt atomic ratio of between 1 : 500 and 1 : 100,000 is applied."

- II. The refusal was on the grounds that this method was obvious for the man skilled in the art from a combination of FR-A-2 107 340 (1) and EP-A-2 749 (2).

A process for the preparation of benzene-monocarboxylic acids is described in (1) wherein a mixture of a dialkyl-benzene compound and a monoalkyl-benzene compound is oxidized in the liquid phase with the aid of a gas containing molecular oxygen in the presence of a catalyst composed of a cobalt and a manganese compound and wherein a catalyst is used which possesses a manganese to cobalt atomic ratio lying within the range claimed in the present claim 1.

The claimed process only differs from this prior art process in transferring the process steps used in (1) for the transformation of a methyl-group of p-xylene and the methyl-group, directly bonded to the ring, of methyl para-toluate into carboxyl-groups to the oxidation according to the present application wherein the methyl-group of toluene or toluene derivatives, such as halo-, nitro-, cyano-, tertiary alkyl-, alkoxy- and aryloxy-toluenes, is converted to a carboxyl-group.

No reasons can be seen which would prevent the man skilled in the art from using the present starting materials in the known process, especially since the preparation of benzene-monocarboxylic acids by oxidation of such alkyl-benzene derivatives under very similar conditions is already known from (2).

It is true, that on page 9, line 33 of (1) the preferred ratio of Mn : Co is indicated as 1 : 99 to 90 : 10. However, according to page 9, line 32 and according to claim 1 of (1) the possibility of using catalysts having a Mn : Co ratio of 0,1 : 99,9 to 99 : 1 is explicitly indicated, this ratio overlapping with the ratio presently claimed.

The Examining Division cannot view the comparison with the prior art corresponding to (1) given on page 5 of the present application, which shows only a trivial difference in yields between the use of a catalyst having a Mn : Co atomic ratio within a preferred range given in (1) and the use of a catalyst having this ratio outside of the preferred range but lying within the overall range disclosed and claimed in (1), as definitive proof of the presence of the required

inventive step in the present subject-matter as all of the steps of the presently claimed process are obvious in view of the prior art, and particularly in the portion of the range of atomic ratio Mn : Co which overlaps with the range disclosed in (1).

- III. On 9 October 1983 the appellant lodged an appeal against the decision of 21 September 1983, with payment of the fee.

In the Statement of Grounds submitted on 12 January 1984 it was pointed out that in FR-A-2 107 340 (1) a tendency is clearly present towards the use of catalysts with a relatively low Co : Mn atomic ratio. This may be seen from table 1, page 20, in which the best result is obtained in example 4, the yield being 92,6%, at a Co : Mn atomic ratio of 32. Raising this Co : Mn ratio according to example 3, 2 and 1 results in lower yields. A similar tendency may be derived from tables 3, page 21 and 5, page 22. The best yields in these tables are obtained at Co : Mn atomic ratio's of 9 and 19 respectively.

Comparative example 2 (table 1, page 20) of (1), in which an atomic ratio Co : Mn of 2500 is applied, shows a yield of 89,1%. The tendency against higher Co : Mn ratio's as mentioned here above is even more prominent in this comparative example 2.

It is rather doubtful whether the processes of (1) and of the present patent application could be compared. The reason for this is that the variations in the yields in the process of (1) are much greater than in the process according to the present application due to

differences in reactivity of the starting compounds. However, if comparison between the two processes is made, the Co : Mn atomic ratio of comparative example 2 would fit perfectly in the preferred range of the process according to the present application.

Therefore reference (1) teaches away from the present invention, which claims the use of a catalyst with a high Co : Mn ratio.

Moreover, the reimbursement of the appeal fee is requested.

#### REASONS FOR THE DECISION

1. The appeal is in accordance with Articles 106-108 and Rule 64 EPC; it is therefore admissible.
2. In the introductory part of the present specification it is pointed out that in the process according to Dutch patent application No. 7 311 187 benzoic acid is prepared by oxidizing toluene in the substantial absence of a lower fatty acid and/or a halogen compound, and in the presence of a cobalt compound and a manganese compound with a weight ratio between cobalt metal and manganese metal of less than 99.8 : 0.2, which means that the manganese : cobalt atomic ratio must be more than 1 : 466. This process has as a disadvantage that the selectivity of the reaction to form the desired benzene-monocarboxylic acid is not satisfactory. It is said that the aim of the invention is to improve this.

A comparative experiment (A) demonstrates that the oxidation of toluene with air in the liquid phase and in the presence of a soluble catalyst having a ratio of Mn : Co 1 : 67 results in the recovery of 908 moles of benzoic acid for every 1000 of moles/<sup>of</sup>toluene fed to the reactor instead of 913 moles according to the invention which applies for example a ratio Mn : Co 1 : 1000 and 1 : 3300 (examples 1 and 2).

That comparison was dismissed in assessing inventive step, since the difference in yields between the known process and that of the application in suit was considered trivial. In not accepting this comparison, the Examining Division failed to appreciate the problem underlying the present application.

It is true that the improvement is a small one, but even small improvements can mirror a technical problem, particularly where an industrially important process for the production of bulk chemicals is concerned. The catalytic oxidation of toluene is the most important and commonly used process for the manufacture of benzoic acid (cf. Ullmann's Encyklopädie der technischen Chemie 4th edition, Volume 8 1974, page 367 right hand column). This process consumes more than 225 000 metric tons of toluene per year (cf. Kirk-Othmer, Encyclopedia of Chemical Technology 3rd Edition volumn 3, 1978 page 780 the last two paragraphs and page 782 above). According to FR-A-2 196 317(3), which is an equivalent to the above NL-A-, the output of a single plant applying this process can amount to 50 000 tons of benzoic acid per year (see page 4 lines 12 to 15) which is confirmed by KirkOthmer (cf. the list on page 782 below). These figures underline that even a small improvement in the

yield of this process is an economically worthwhile technical problem which must not be disregarded in assessing the inventive step of the improved process.

3. Therefore, according to the evidence before the Board, the technical problem underlying the present application has to be seen in improving the process of catalytic oxidation of toluene in liquid phase with regard to the yield of benzoic acid.

This problem is solved by the applicant in applying a soluble catalyst having a Mn : Co atomic ratio between 1 : 500 and 1 : 100 000.

4. A teaching so defined cannot be gathered from any of the publications before the Board. Therefore, the application in suit is deemed to be novel.

It is therefore to be examined whether the subject matter of the application is obvious in relation to the prior art.

In the first instance, the figures with regard to "selectivity", i.e. the formed benzoic acid and the side products which can be transformed to that acid in relation to the consumed toluene, listed in tables 1 and 2 of (3) are instructive. The "selectivity" which is roughly comparable with the yield of benzoic acid, depends on the atomic ratio Mn : Co and increases starting from both end points, i.e. Mn : Co about 1 : 500 (table 1, 1-b and table 2, 2-b) on the one hand and Mn : Co about 19 : 1 (table 1, 1-i) respectively Mn : Co about 1 : 1,5 on the other (table 2, 2-g), going through a maximum (93,4%, 94%) if a ratio of



about 1 : 19 (table 1, 1-e where 190°C and 10Kg/cm<sup>2</sup> are applied) respectively 1 : 20 is applied (table 2, 2-e, where 160° and 7 Kg/cm<sup>2</sup> are used). From these results a skilled person would conclude that the yield of benzoic acid would continue to decrease if both end points were exceeded.

5. The two documents cited in the impugned decision do not offer enough material for predicting how the yield of benzoic acid in the oxidation of toluene could be improved. Document (1) which was quoted by the first instance is concerned with the oxidation of a mixture consisting of p-xylene and methyl p-toluate with oxygen in the liquid phase to produce a mixture comprising p-toluenic acid and monomethyl terephthalate. This oxidation is one step in a sequence of reaction steps leading from p-xylene to dimethyl terephthalate (Claim 6).

As far as the oxidation step is concerned, only one component of the mixture due to be oxidised, namely methyl p-toluate, is a monoalkyl-benzene compound in the meaning of the definition according to the present application (cf. page 2 lines 17 to 21). The known process mainly aims at substantially decreasing the amount of coloured and fluorescent impurities (see page 5 lines 15 to 21), a problem which is quite different from that envisaged in the present application.

Nevertheless, this document was considered relevant in assessing obviousness of the present application, since catalysts are disclosed which have the same range of atomic ratio Mn : Co as claimed. However, this approach neglects the technical problem underlying the claimed process and therefore is improper to support the argu-

ment of obviousness. As to the question of yield, table 1 lists up the effective yield of all compounds able to be transformed to dimethyl terephthalate by oxidation and/or esterification (cf. page 12 lines 7 to 16).

Assuming that the yield of monomethyl terephthalate, which is the product of the oxidation of the monoalkylbenzene methyl p-toluate runs parallel to the total yield of oxidation products, the best result is obtained in example 4, the yield being 92,6%, at a Co : Mn atomic ratio of 32. Raising this Co : Mn ratio according to example 3, 2 and 1 results in lower yields. A similar tendency may be derived from tables 3, page 21, and 5, page 22. The best yields in these tables are obtained at Co : Mn atomic ratios of 9 and 19 respectively. Evidently the teaching to carry out the oxidation of the above monoalkylbenzene compound within these atomic ratios fails to render obvious, from the point of view of the problem concerned, the performance of the oxidation of toluene and its derivatives at an atomic ratio Mn : Co beyond 1 : 500.

6. Document (2) discloses a process for the oxidation of a monoalkylbenzene compound including toluene with oxygen in liquid phase in the presence of a catalyst comprising a combination of a cobalt and manganese compound (see Claims 1 and 3, page 2 lines 17 to 25 and page 7 lines 11 to 15). The objective of this process, an improved yield, is achieved by applying a small amount of a lower aliphatic monocarboxylic acid (see Claim 1, page 4 paragraphs 1 and 3). This effect is particularly demonstrated in the oxidation of o-chlorotoluene (tables 1 to 4) and toluene (table 2). The extreme proportions between Mn and Co in the catalysts used in the examples are 1 : 50 (examples 1 to 6, 11,

reference examples 1 to 4) and 3 : 1 (example 12). All examples mirror the change of yield in relation to the amount and concentration of the added aliphatic monocarboxylic acid, and additionally the content of the incorporated reaction accelerator (Br), but not, in the absence of a real comparison, the influence of the atomic ratio within the catalyst itself on the yield. This applies also to examples 11 and 12 where the amount of acetic acid, the reaction temperature, reaction pressure and air flow rate are all the same, the reaction time being similar (4 and 4,5 hours respectively). But since the concentrations of Co and Br are substantially different, the improvement in the yield of benzoic acid (example 11 : 95,5%) cannot be ascribed to the change of the atomic ratio Mn : Co from 3 : 1 (example 12) to 1 : 50 (example 11).

Consequently, the skilled person was not able to draw the conclusion from the prior art that the applicant's problem might be solved by applying a Mn-Co- catalyst having the claimed range of atomic ratio.

7. From the foregoing it follows that the decision under appeal is not supported by the grounds for refusal. However, the patent sought cannot be granted at present, because some questions have yet to be settled. First of all whether the effect demonstrated with catalysts having the atomic ratio Mn : Co of examples 1 and 2 is credibly achieved, i.e. without additional evidence, over the whole range as claimed including the ratio Mn : Co 1 : 100 000. Moreover, it is to be examined whether the improvement in yield demonstrated in the oxidation of toluene extends to the oxidation of its derivatives (cf. page 2 lines 17 to 21). Should this be

the case, the further question arises whether the extent of the improvement qualifies, as in the toluene oxidation, as a technical problem, so that the question of obviousness of its solution has to be assessed from the point of view of this problem. These important questions have not yet been asked by the Examining Division which is in the first place competent for such an enquiry.

8. The appellants have shown no cause for the requested reimbursement of appeal fee. The Board cannot find a substantial procedural violation by reason of which the reimbursement would be equitable.

ORDER

For these reasons,

it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division for further prosecution.
3. The request to reimburse the appeal fee is dismissed.

*J. R. K.*

*D. Cadman*