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File Number: T 291/89 - 3.3.2

Application No.: 81 902 770.7

Publication No.: 0 064 504

Title of invention: Preparation of substantially anhydrous iodine compounds

Classification: C01B7/13

**D E C I S I O N**  
of 14 May 1991

Proprietor of the patent: Eastman Kodak Company (a New Jersey Corporation)

Opponent: Hoechst Aktiengesellschaft, Frankfurt

Headword:

EPC Articles 83 and 56

Keyword: "Inventive step (affirmed) - non-obvious improvement"

**Headnote**



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Boards of Appeal

Chambres de recours

Case Number : T 291/89 - 3.3.2

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.2  
of 14 May 1991

**Appellant :**  
(Opponent)

Hoechst Aktiengesellschaft, Frankfurt  
Werk Knapsack  
Patentabteilung  
W-5030 Hürth-Knapsack  
Germany

**Representative :**

**Respondent :**  
(Proprietor of the patent)

Eastman Kodak Company  
(a New Jersey corporation)  
343 State Street  
Rochester  
New York 14650  
United States of America

**Representative :**

Baron, Paul Alexander Clifford  
Kodak Limited  
Patent Department  
Headstone Drive  
Harrow  
Middlesex HA1 4TY  
Great Britain

**Decision under appeal :**

Decision of the Opposition Division of the  
European Patent Office dated 9 March 1991  
rejecting the opposition filed against European  
patent No. 0 064 504 pursuant to Article 102(2)  
EPC.

**Composition of the Board :**

**Chairman :** Lançon P.A.M.  
**Members :** Eberhard M.M.  
Schulte R.L.J.

## Summary of Facts and Submissions

- I. European patent No. 64504 was granted with three claims on the basis of European patent application 81 902 770.7. Independent Claim 1 reads as follows:

"Process for preparing an anhydrous iodine compound which comprises reacting, under substantially anhydrous conditions, hydrogen with iodine in a non-alcoholic, organic solvent in the presence of a homogeneous rhodium catalyst, there being present no heterocyclic aromatic compound in which at least one heteroatom is a quaternary nitrogen atom and no quaternary organophosphorous compound."

- II. The Appellant (Opponent) filed a notice of opposition requesting revocation of the patent on the grounds of lack of novelty with respect to an earlier European patent application forming part of the state of the art as defined in Article 54(3) and lack of inventive step (Article 56 EPC). In support of this latter ground the Appellant cited, inter alia, the following documents:

- (2) DE-A-2 441 502
- (4) US-A-4 046 807 corresponding to the German patent application (5) DE-A-2 450 965.

- III. The Opposition Division rejected the opposition. The claimed process was regarded as novel over the earlier European application. In its decision the Opposition Division pointed out that the use of hydrogen was neither disclosed nor suggested in (2) and that the process of (4) (or (5)) involved the addition of hydrogen to the carbon oxide reaction gas in order to suppress the formation of

soot and carbon dioxide observed occasionally at reaction temperatures of above 150°C. However, the problem underlying the patent was to find a method for preparing anhydrous iodine compounds in a good yield and at reasonable reaction rates by the direct reaction of hydrogen with iodine. In the Opposition Division's view, documents (2) and (4) were not concerned with this problem and, therefore, could not give an incentive to solve it as set out in Claim 1. In particular, it was not predictable from these documents that good yields at reasonable reaction rates could be achieved by the claimed process.

- IV. The Appellant lodged an appeal against this decision. In his statement of grounds he relied upon a document cited for the first time at the appeal stage, namely (6) US-A-3 848 065, and contended that the process of Claims 1 to 3 of the patent in suit did not involve an inventive step in view of documents (2), (4) and (6) for the following reasons:

According to document (2) the iodide could be added also in form of elementary iodine, carbon monoxide being used in a practically pure form. However, the skilled person concerned with the carbonylation process was aware of the advantages obtained by replacing a part of the carbon monoxide with hydrogen since they were indicated in (4). If only the conversion of iodine to anhydrous iodine compounds was important to the skilled person and not the production of acetic anhydride, then he would have chosen a higher hydrogen partial pressure in view of the teaching of (4). Furthermore, he would have tried to decrease up to nought the content of carbon monoxide in the process of (2) as this gas does not take part in the reaction of formation of iodine compounds. Thereby, he would have arrived at the expected result that the reaction could be carried out in the absence of CO. The skilled person would

have been directly encouraged to perform such experiments in view of (6) which disclosed the reaction of iodine with hydrogen in the presence of a rhodium catalyst.

- V. During the oral proceedings held on 14 May 1991, the Appellant raised an objection of insufficiency of disclosure against the process as defined in Claim 1 of the patent. In this respect he stressed that in the absence of the disclaimed compounds the reaction of iodine with hydrogen could lead to the desired result only if small cations such as Li cations were present in the reaction medium. However, Claim 1 did not mention the presence of a lithium salt as promoter so that the desired product could not be obtained.

As regards inventive step, the Appellant referred to the following passages of (2): page 5, lines 15-23; page 7, lines 8-9; page 8, second paragraph; page 11, lines 8-16; page 13, lines 1-4 and 15; page 15, lines 5-18; example 12 which, in his opinion, suggested that an alkyl iodide could be formed under anhydrous conditions by addition of iodine and contact with a homogeneous Rh catalyst in the presence of a promoter such as lithium acetate. He argued that in case of halogen loss the skilled person could have determined the iodine amount which was necessary to obtain the alkyl iodide instead of the acetic anhydride. The Appellant further stressed that elementary iodine, hydrogen and a promoter were present in the process of (4). In his view it could be derived from this document that acetic anhydride and an anhydrous alkyl iodide was formed as soon as the water was consumed.

- VI. The Respondent's arguments can be summarised as follows:

The ground of insufficiency of disclosure was put forward for the first time during the oral proceedings so that the

Respondent did not have the opportunity to deal with this matter before or to ask instructions thereupon. Example 5 of the patent showed anyway that the desired product could be obtained with a very good yield even in the absence of lithium iodide.

In the process of document (2) the alkyl iodide was regenerated and was thus neither consumed nor exhausted. The net reaction was, therefore, one between a carboxylic acid ester or ether and carbon monoxide. Therefore, there was no production of hydrogen iodide, alkyl iodide or lithium iodide except where the small amount required for the working of the process was formed in situ. Even in such cases no hydrogen was used and the reaction was quite different from the present invention. There was nothing in (2) which would have suggested that iodine compounds could be produced instead of acid anhydrides nor that hydrogen could be substituted for CO. A fortiori, there was no suggestion that in such a case a Rh catalyst would be preferred over other noble metals for the production of iodine compounds.

In the process of (4) carbon monoxide was one of the major reactants not hydrogen. Furthermore, the only advantage given in (4) in connection with the presence of hydrogen was the soot removal. From a comparison of example 13 with the other examples it could be concluded that the worst possible yield of acetic anhydride was obtained in the presence of hydrogen. Even if an alkyl iodide was formed in the carbonylation reactions of (2) or (4), the yield thereof was very low and, therefore, these documents could not suggest the claimed solution to the problem of increasing the yield of the desired iodine compounds.

VII. The Appellant requests that the decision under appeal be set aside and the patent be revoked.

The Respondent requests that the appeal be dismissed.

### Reasons for the Decision

1. The appeal is admissible.
2. The Appellant's objection that the process as defined in Claim 1 does not lead to the desired products (cf. point V above) was submitted for the first time during the oral proceedings before the Board, i.e. at a very late stage of the proceedings. When questioned by the Board about the reasons for this tardiness the Appellant indicated that it was found only about one week before oral proceedings that in the absence of the disclaimed compounds the active complex of the rhodium catalyst can be formed only if small cations such as lithium cations are present in the reaction medium.

Although these arguments were presented as an objection concerning sufficiency of disclosure (Article 100(b)), they raise in fact the question whether Claim 1 recites all the features necessary for the obtention of the desired products, i.e. whether this claim meets the requirements of Article 84 EPC. However objections based upon Article 84 do not constitute a ground of opposition.

In view of the Appellant's strong reliance on this point during the oral proceedings, the Board has examined these belatedly submitted arguments on its own motion.

First of all the Board observes that, on the one hand, the Appellant's allegation was not supported by any evidence and, on the other hand, it was contested by the Respondent's representative. In this context the latter

pointed out that a good yield was obtained in example 5, although the reaction was carried out in the absence of lithium iodide and that it was not clear how far the lithium acetate used as hydrogen iodide "acceptor" was dissociated in the reaction medium of example 5. The Board further notes that, although LiI or Li acetate is present in the reaction medium of the examples, it is not derivable from the patent that the presence of one of these compounds or of lithium cations is necessary for the solution of the problem. Thus, according to column 3, lines 18-24, the ionic rhodium species of the catalyst may be formed by using iodine compounds such as LiI, CH<sub>3</sub>I, HI or iodine. Moreover according to column 2, line 57 to column 3, line 4 the hydrogen iodide "acceptor" may be methyl acetate instead of lithium acetate and its initial inclusion in the reaction medium is only preferred. Finally, as stressed by the Respondent's representative, owing to the fact that this argument was relied upon for the first time during the oral proceedings the Respondent did not have the opportunity to make tests in order to verify the Appellant's allegation and to take position upon this matter.

Under these circumstances and in the absence of evidence provided by the Appellant who has the onus of proof, the patent proprietor is given the benefit of the doubt (cf. decision T 219/83, OJ EPO 1986, 211).

As regards sufficiency of disclosure the Board wishes to point out that this question has to be judged not merely on the basis of claims but also on the basis of the description: cf. decision T 14/83, OJ EPO 1984, 105. In the present case the Appellant did not contest that the nine examples of the patent are reproducible and lead to the desired iodine compounds with the desired high yield.



Furthermore, the Board has no reason to doubt that the process as described in the examples is sufficiently disclosed for it to be carried out by a skilled person.

Under these circumstances the requirements of sufficiency of disclosure and of Article 84 are considered to be met.

3. The Appellant further sought to introduce document (6) into the proceedings for the first time at the appeal stage. Although this document was mentioned in the search report, it was neither cited in the notice of opposition nor in the course of the opposition procedure. According to decision T 198/88 (OJ EPO 1991, 254) a document does not automatically form part of the opposition or appeal proceedings if it is merely cited and acknowledged in the contested patent since the opposition procedure is not part of the grant procedure. The same applies obviously to a document which is merely cited in the search report.

After examination of document (6) of its own motion the Board has reached the conclusion that even if it were taken into consideration the outcome of the decision would be the same. Therefore, document (6) had to be disregarded pursuant to Article 114(2).

4. The patent in suit relates to a process for preparing an anhydrous iodine compound by the reaction of hydrogen with iodine in the presence of a noble metal-containing catalyst under anhydrous conditions.

As indicated in the patent in suit it was known to produce anhydrous hydrogen iodide by the direct reaction of iodine vapour with hydrogen over a platinum catalyst at elevated temperatures. After examination of the documents acknowledged in the description, the Board agrees that this known process represents the closest prior art.

Although this method provides HI of high purity, the reaction is slow, conversion is not complete and yields of the desired product are consequently low.

In the light of this prior art, the problem underlying the patent can be seen in providing a method for preparing anhydrous iodine compounds such as hydrogen iodide, methyl iodide or lithium iodide in good yield and at reasonable reaction rates by the direct reaction of hydrogen with iodine in the presence of the noble metal-containing catalyst.

According to Claim 1, it is proposed to solve this problem by carrying out the reaction in a non-alcoholic, organic solvent in the presence of a homogeneous rhodium catalyst, the presence of specific compounds stated in Claim 1 being excluded.

In view of the production rate of hydrogen iodide indicated in examples 1 and 2 of the patent and of the high yields of lithium iodide and methyl iodide stated in table 1 (i.e. 86.4-90%), it appears to the Board that this problem has been plausibly solved by using a homogeneous rhodium catalyst under the claimed conditions.

5. None of the cited documents discloses the production of anhydrous iodine compounds by reacting iodine with hydrogen in the presence of a homogeneous rhodium catalyst. Therefore, the claimed process is novel. This not being in dispute there is no need to give further details.
6. It still remains to examine whether the claimed process involves an inventive step with regard to the teaching of the cited documents.

6.1 Document (2) relates to the manufacture of anhydrides of monocarboxylic acids such as acetic anhydride by carbonylation. A carboxylic acid ester, for example, methyl acetate, or an ether is reacted with an acyl iodide under anhydrous conditions to form the anhydride. The acyl iodide is itself formed in a first reaction step by carbonylation of an alkyl or aryl iodide with carbon monoxide in the presence of a noble metal catalyst (cf. Claims 1-3; page 4, last paragraph; page 5, lines 20-23). The alkyl iodide is regenerated in the second reaction step, separated from the acetic anhydride and recycled (cf. page 6, lines 1-4; page 7, lines 8-9). In a preferred embodiment the two reaction steps are combined, the alkyl iodide and the ester or ether being then introduced into the same reaction zone (cf. page 8, lines 8-18). Document (2) further teaches that the alkyl iodide may also be formed in situ by feeding the necessary iodine amount in the form of another organic iodide, hydrogen iodide, metal iodides, alkali iodides or even elementary iodine (cf. page 8, lines 18-26). Lists of appropriate solvents, promoters and noble metal containing catalysts, a.o. homogeneous rhodium catalysts are given at pages 11 to 13. In example 12 elementary iodine is used and the catalyst is incorporated into the reaction medium as rhodium trichloride hydrate.

As contended by the Appellant and not contested by the Respondent, although (2) is essentially directed to the carbonylation of the ester or ether in order to produce the anhydride, it also suggests that an anhydrous alkyl iodide may be formed by heating the reaction medium comprising iodine, a carboxylic acid ester or an ether, a homogeneous rhodium catalyst and a promoter under anhydrous conditions in the presence of a carbon monoxide atmosphere. Nevertheless, in view of the fact that carbon monoxide is used, it appears that the reaction of

formation of the alkyl iodide is quite different from the process involving reaction of iodine with hydrogen. Although it is envisaged in (2) to add an inert diluting gas such as carbon dioxide, nitrogen, methane or noble gases to the carbon monoxide (cf. page 12, second paragraph), it is not suggested to replace a part of the carbon monoxide with hydrogen.

It can further be inferred from (2) that relatively low amounts of the alkyl iodide are produced, since it is regenerated and remains in the system in the case of a continuous process. Even if some loss occasionally happens, then the amount of alkyl iodide necessary to compensate this loss is very small (cf. page 15, first paragraph). As the purpose of this document is to improve the process for manufacturing a carboxylic acid anhydride such as acetic anhydride (cf. page 2, second paragraph) the yield of acetic anhydride is mentioned but there is no indication concerning the yield of alkyl iodide.

It results from the preceding that (2) does not contain any information which could provide the skilled person with an incentive to replace the known platinum catalyst used in the catalytic reaction of hydrogen with iodine to form anhydrous hydrogen iodide by the homogeneous rhodium catalyst used in (2) for a different reaction, in order to improve the yield of the desired anhydrous iodides.

- 6.2 Document (4) does not relate to the preparation of iodine compounds but, like (2), to that of acetic anhydride. The process comprises reacting methyl acetate and carbon monoxide in the presence of a catalyst containing a noble metal, in particular rhodium or a rhodium compound, and iodine or iodine compounds such as alkyl or acyl iodides. The iodine-containing component of the catalyst may be added as elementary iodine, hydrogen iodide, inorganic

salts such as sodium, potassium or cobalt iodides or organo-iodine compounds such as alkyl iodide, especially methyl iodide, or acyl iodides (cf. column 1, lines 24-30; column 2, lines 18-26; column 3, lines 9-20; Claims 1, 5-7). The catalyst is for example  $\text{RHCl}_3 \cdot 3\text{H}_2\text{O}$  with methyl iodide as iodine component (cf. examples 1, 2-4, 13).

According to (4) the presence of 5 to 50% by volume of hydrogen in the carbon monoxide needed for the carbonylation reaction has the favourable effect of suppressing the slight formation of soot and carbon dioxide which can be observed at temperatures of above  $150^\circ\text{C}$  (cf. column 1, line 64 to column 2, line 2). In example 13, where the reaction is performed in the presence of carbon monoxide and hydrogen, the yield of acetic anhydride is, however, substantially lower than in the examples using the same catalyst in the absence of hydrogen. In example 14, methyl iodide and iodine are indeed used as iodine components of the catalyst but the carbonylation reaction is carried out in the absence of hydrogen. Even if under the operating conditions prevailing in the carbonylation process of (4) anhydrous hydrogen iodide and/or an alkyl iodide might be formed as by-product by reaction of hydrogen with iodine in the case where both are present, this is neither disclosed in (4) nor derivable therefrom without knowledge of the claimed solution, since according to (4) hydrogen is added to the carbon monoxide for the totally different purpose stated above. Therefore, in the Board's opinion, the skilled person could not expect in view of the teaching of (4) that the substitution of a homogeneous rhodium catalyst for the known platinum catalyst would improve the yield of anhydrous hydrogen iodide, lithium or methyl iodide obtained by reaction of hydrogen with iodine. Without the expectation of an improvement in the yield of these products the skilled person faced with the problem defined

above would not have been prompted to replace the platinum catalyst used for the reaction of hydrogen with iodine by the catalyst disclosed in (4) for the carbonylation of methyl acetate nor to substitute hydrogen for carbon monoxide in the process of (2) (cf. decision T 2/83, OJ EPO 1984, 265).

7. For the preceding reasons, the subject-matter of Claim 1 is considered to involve an inventive step. Claim 1 being allowable, the same applies to the dependent Claims 2 and 3 whose patentability is supported by that of Claim 1.

**Order**

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

The appeal is dismissed.

The Registrar:

The Chairman:

P. Martorana

P.A.M. Lançon