



1732 82,277

Decision dated 10 February 1982  
T 20/81

Appellant: Shell Internationale Research Mij.  
(Grotenhuis, Menninga)

Key-word: Shell/aryloxybenzaldehydes

EPC Article 56

Rule 27(1)(d)

"Inventive step : significance of the technical problem"

"truly comparative examples"

#### Headnote

In order to render them relevant to the definition of the problem underlying the invention, and hence to the assessment of inventive step, alledged advantages should be supported by sufficient evidence where comparison is made with highly pertinent prior art.

Europäisches  
Patentamt

Technische  
Beschwerdekammern

European Patent  
Office

Technical Boards  
of Appeal

Office européen  
des brevets

Chambres de  
recours techniques



File Number T 20/81

DECISION  
of the Technical Board of Appeal 3.3.1  
of 10 February 1982

Appellant:

Shell Internationale Research  
Maatschappij B.V.  
The Hague  
The Netherlands

Representative:

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Decision under appeal:

Decision of the Examining  
Division 002 of 16 February  
1981 to reject European Patent  
Application No. 78 200 383.4 in  
accordance with Article 97(1)  
EPC

Composition of the Board:

D. Cadman	Chairman
K. Jahn	Member
L. Gotti Porcinari	Member

## SUMMARY OF FACTS AND SUBMISSIONS

I. European patent application No. 78 200 383.4 filed on 20 December 1978 and published on 25 July 1979 (publication number 0 003 066) was refused by the decision of Examining Division 002 of the European Patent Office dated 16 February 1981. This decision is concerned with the 10 claims, filed on 5 September 1980, having the following wording:

1. Process for the preparation of meta-aryloxybenzaldehydes by reaction of a mixture of the corresponding meta-aryloxybenzyl halides and meta-aryloxybenzal halides with hexamethylene tetramine followed by hydrolysis of the product resulting therefrom characterised in that the hydrolysis is carried out in an alkanol-water mixture containing 40%v to 80%v of an alkanol with up to four carbon atoms per molecule.

2. Process according to claim 2 characterised in that the whole process is carried out in an alkanol-water mixture.

3. Process according to claim 1 or 2 characterised in that the alkanol is ethanol.

4. Process according to claim 1, 2 or 3 characterised in that the mixture of the meta-aryloxybenzyl halide and meta-aryloxybenzal halide used as starting material is prepared by a process which comprises halogenating the

corresponding meta-aryloxytoluene with gaseous halogen at an elevated temperature in the presence of a free-radical initiator.

5. Process according to claim 4, characterised in that a mixture of meta-aryloxybenzyl chloride and meta-aryloxybenzal chloride is prepared by a process which comprises contacting a meta-aryloxytoluene in a non-polar solvent at a temperature in the range of from 40 to 100°C with gaseous chlorine.

6. Process according to claim 5, characterised in that the non-polar solvent is a halogenated hydrocarbon.

7. Process according to claim 6, characterised in that the halogenated hydrocarbon is carbon tetrachloride.

8. Process according to any one of claims 4, 5 or 6, characterised in that the free-radical initiator is azoisobutyronitrile.

9. Process according to any one of claims 4 to 8, characterised in that the reaction between the meta-aryloxytoluene and chlorine is stopped at a conversion in the range of from 95% to 99%, based on meta-aryloxytoluene.

10. Process according to any one of the preceding claims, characterised in that the meta-aryloxybenzaldehyde is meta-phenoxybenzaldehyde.

II. The stated ground for the refusal was that the subject matter of claim 1 did not involve an inventive step. As the applicant himself states in the description, the process according to claim 1 is known from NL-A-7 701 128, except that the latter employs an acid medium for the hydrolysis. Faced with the problem of avoiding the disadvantages that emerge from the use of an acid, the skilled man would have considered such nearest state of the art, and he would have also learned from Organic Reactions Vol. III, 1954, John Wiley, New York, Chapter 4, S.H. Angyal, The Sommelet Reaction, page 198, the possibility of using as a reaction medium water or aqueous ethanol instead (page 206). It was merely a matter of routine for the skilled practitioner to find the special alcohol-water mixtures and suitable alcohols other than ethanol. That there are now two different hydrolysis reactions, namely those involving benzyl and benzal halides, is of no significance, since the hydrolysis of the products resulting from both compounds were routinely treated in the cited NL-A.

Furthermore, no indication of an inventive step could be seen in the absence of any surprising effects. The yield of aldehydes and the total reaction time were nearly the same for the known and for the claimed processes, when the pressure used was taken into account. In the absence of a valid claim 1 the other claims were not allowable since they had no inherent significance (claims 2, 3 and 10) which would overcome the objections, or were directed to a combination with known processes of preparing the starting material (claims 4-9).

- III. The appellant filed an appeal against the decision of 16 February 1981 on 7 April 1981, and submitted a Statement of Grounds on 9 June 1981.
- IV. The Board of Appeal then, in a communication to the appellant, introduced a new document, Zh. Prikl. Khim 1966 39(7), pages 1669-1670 (hereinafter "Libman"), reported in Ullmanns Encyklopädie der technischen Chemie, (1974) Vol. 8 pages 346-347, which discloses the reaction of combined benzyl and benzal halides with hexamethylene tetramine in water as solvent. The appellant responded to the communication in due time and when doing so eventually developed the following argument:

The problem, according to the appellant, was not simply to eliminate the disadvantages emerging from the use of an acid in the process of the NL-A cited above, but to improve the known process for preparing meta-phenoxy benzaldehyde (hereinafter POAL).

Facing this problem the person skilled in the art had to select which of the many conceivable process variables offered a promising improvement. Only when elimination of acid had been identified as a possibly advantageous improvement did the next problem emerge, namely in what way that could be done.

The closest prior art, viz NL-A-7 701 128 (hereinafter "old Shell process"), teaches unambiguously that acid must be used. Even common general knowledge in the art, namely the article in Organic Reactions cited above (hereinafter "Angyal"), teaches that acid is preferred in the Sommelet reaction. If the disadvantages of using

acid were well known then the skilled man would have expected the "old Shell process" to describe the process without the use of an acid.

Libman described the reaction of a mixture of benzyl and benzal chlorides to make benzaldehydes. However, even with knowledge of this prior art, the man skilled in the art would not have arrived at the solution claimed in the application, since firstly there was no teaching of the special alkanol-water solvent system and secondly there would be a doubt whether the conclusion could be drawn that the Libman process would work to produce POAL in place of benzaldehyde, in view of the teaching of Angyal about the hindrance of the Sommelet reaction by electron withdrawing substituents (page 201).

Furthermore, the Libman Process was not believed to represent the correct mechanism, for in the "old Shell process" unchanged benzal halide was found at the end of the first step in the process, i.e. the reaction of hexamethylene tetramine with the mixed aromatic halides (page 2 lines 26 etc).

Finally a further document, GB-A-1 557 421 (hereinafter American Cyanamid) was submitted by the appellant which referred in the introductory part to the Libman article, and described the hydrolysis step, leading to POAL, with a dilute mineral acid. If the process according to the application, which avoids any use of an acid, was really so obvious, then the "old Shell process" and the "American Cyanamid" process would have referred to that possibility.

The appellant has requested that the decision under appeal be set aside and the patent be granted with the above cited 10 claims. The appellant has also indicated his willingness to delete claims 4 to 9.



Reasons for the decision

1. The appeal is in accordance with Articles 106-108 and Rule 64 EPC; it is therefore admissible.
2. There is no formal objection to the current version of the claims, since it is adequately supported by the original documents. Claim 1 is based on the original claims 1, 2 and 4; claim 2 is supported by the description page 3, lines 12-17 and 27-31 and examples 1-4 and 6; claim 3, claims 4-10 correspond to claim 3 and to claims 5-11, respectively.
3. As indicated in the preamble of the patent application the applicant starts from the process of the NL-A-7 701 128 (old Shell process), which involves two steps, the first step comprising the reaction of a mixture of meta-aryloxybenzyl halides and meta-aryloxybenzal halides with hexamethylene tetramine and the second step consisting of the hydrolysis of the resulting product in an acidic medium to form meta-aryloxybenzaldehyde (cf. page 1, lines 9-15). The acid necessary in the second step makes it obligatory to use an acid-resistant vessel (page 1 lines 16-19). It is evident from this that the applicant felt the use of such a vessel disadvantageous and addressed himself to the problem of improving the known process in such a way that he could dispense with the acid-resistant vessel i.e. to avoid the use of the corrosive acid medium in this step, without substantial loss in the yield of POAL.

The applicant has failed to persuade the Board with his argument that the problem was to find a worthwhile improvement of the old process requiring a selection within the range of conceivable variables. Apart from the fact that the problem as defined by the applicant is too vague to be resolved by specific technical means, the nature of the problem should be determined on the basis of objective criteria, as stipulated in the Board's earlier decision "Carbonless copying paper" (cf. Official Journal EPO 1981, 206). This requires the assessment of the technical success vis-à-vis the state of the closest art. There can be no doubt that the particular success achieved by the application in suit lies in making the reaction independent of the use of an acid-resistant vessel (cf. page 2, lines 8-10).

Moreover, the applicant refers to the advantage that hydrolysis can now be carried out in a shorter time to obtain a slightly higher yield of POAL (page 2, lines 1012). This advantage is said to be proven by the results of the comparative example within example 5 on file. Contrary to the submission of the applicant, the comparison is not pertinent, since different temperatures were used, namely 110°C for the hydrolysis step pursuant to the present application, but only 106°C for that of the old Shell process. It is well known that the employment of different reaction temperatures entails a different speed of reaction. In general, a rise of temperature by 10°C will cause the speed to double or triple.

The objection made by the Board that the old Shell process was apparently giving better yields and in shorter time (example<sup>3</sup> 3 of the mentioned NL-A) has not been met

by the submission of a strictly comparable experiment, as might have been expected, but by an argument about the non-relevance of such a comparison in view of the different proportions of meta-phenoxybenzyl chloride and meta-phenoxybenzal chloride used as starting materials in admixture (NL-A example 3: 60:40; present application example 5: 50:50). Neither was an explanation given why such a small shift (10%) in the proportion of the two components of the mixture should have resulted in a significantly different yield and reaction time, in spite of the same reaction temperatures (reflux temperature), nor was claim 1 restricted accordingly.

Consequently, the conclusion must be drawn that the additional advantages referred to by the appellant have not been properly demonstrated. Such alleged but unsupported advantages cannot be taken into consideration in respect of the determination of the problem underlying the application. For this reason, the problem the applicant must have faced remains the one already defined.

4. In order to solve this problem the applicant proposes to carry out the hydrolysis step in an alkanol-water mixture containing 40%v to 80%v of an alkanol with up to four carbon atoms per molecule.
5. Notwithstanding the statement in the old Shell document that the presence of acid in the hydrolysis step was an essential feature, it was common textbook knowledge that Libman had already described the analogous reaction of a mixture of benzyl chloride and benzal

chloride with hexamethylene tetramine, and the subsequent hydrolysis of the product so obtained with water alone to produce benzaldehyde in high yields (85-90%).

6. Moreover, the notional skilled man would also have had the general knowledge that the similar Sommelet reaction, i.e. between benzyl halides and hexamethylene tetramine resulting in benzaldehydes, which is considered by the applicant as a reaction going side by side with the hydrolysis of benzal halides, could be properly carried out in water as a solvent if the starting material and the intermediates have sufficient solubility therein, although the addition of an organic solvent is preferred (Angyal document, page 206, paragraph 4). He would also know that aqueous ethanol (50-80%) was the solvent favoured by the early workers (Angyal page 206, penultimate paragraph), although in many cases in which comparative runs were made with several solvents, 50% acetic acid proved to be the best medium (Angyal page 207, paragraph 2). Notwithstanding that, aqueous ethanol could not be disqualified as obsolete because it was stated to be one of the two recommended solvent systems (cf. Angyal, the last two lines of page 208, to line 10, especially lines 7-10 of page 209).
  
7. From the point of view of the problem of avoiding the acid medium in the hydrolysis step of the old Shell process, the prior art cited was well adapted to offer a solution, because it clearly opened up the prospect of using aqueous ethanol, within the percentage range as claimed, in place of the commonly used acetic acid.

Furthermore, it could be expected that the yield achieved in the old Shell process would be maintained in aqueous alcohol, because the analogous unsubstituted benzaldehyde had been obtained according to Libman in an excellent yield (85-90%), using as reaction medium water alone, which also forms a substantial part of the reaction medium in the old Shell process and in the process of the present application. That the teaching of the present application is that the old Shell process does not require an acidic medium must be regarded, in the light of the problem, as predictable and hence involving no inventive step.

8. The applicant has also advanced the argument that if the disadvantages of using acid had been well known, then the skilled man would have expected the old Shell process to be described without the use of acid. However, consideration of the old Shell document reveals that it is concerned with the quite different problem of avoiding complicated control in the preparation of halogenated toluene intermediates (cf. page 1, paragraph 2) and there is, in the view of the Board, no reason to suppose that the inventor of the old Shell process should at the same time have solved any other problem associated with the process, even where such solution would have been obvious. The development of any technical process commonly proceeds in a series of short steps during the course of which the skilled man focusses his attention ever closer on questions which initially have been regarded as of lesser importance, and the fact that workers in a field of activity had not earlier addressed themselves to solving a particular problem should not of itself be taken as a reliable indication that the solution eventually proposed was not obvious.

Thus the fact that the inventors of the old Shell process only adopted one of the alternatives recommended by Angyal, notwithstanding his teachings and those of Libman, is no indication of prejudice against the existence of the other alternative, as later confirmed by the applicant's own use of aqueous alcohol. It is true that the old Shell process produced a very good result, if one considers the high yield (97%) and the short reaction time (4 1/4 hours). But, as it has been represented by the applicant, no process is perfect in every respect. However, with the recognition of serious problems as a consequence of the use of acid, it was obvious to profit from the state of the art, as submitted in detail in Sections 5-7 above.

9. Furthermore, the appellant contends the non-obviousness of the application in suit by the argument referring to an inhibition of the Sommelet reaction by electron withdrawing substituents. However, in our view this argument lacks persuasive power. It is stated in the Angyal article (page 201, paragraph 3) that an accumulation of strongly electron-attracting substituents may prevent the reaction. Consequently 2,4-di-nitrobenzaldehyde cannot be prepared, but one strongly electron-attracting substituent fails to impair significantly the reaction because nitrobenzaldehyde can be properly obtained (63% yield, cf. page 210). With the relatively weak electron-withdrawing phenoxy substituent a hindrance of the reaction was not to be feared.

10. Contrary to the view of the applicant, it is not crucial for the assessment of the question of inventiveness whether or not a reaction scheme (e.g. (2) in Libman's paper, page 1669) given in a document represents the correct mechanism of a reaction. The only matter that eventually counts is the result, not its explanation. The same result, an aromatic aldehyde, is obtained according to both documents, Libman and Shell. Nothing can be derived in favour of the inventivity of the process claimed from the statement in the old Shell document that the first step results in a mixture of a benzylhalide complex with unchanged benzalhalide, whereas pursuant to Libman benzalhalide is said to form a similar complex.
  
11. Finally the applicant refers to GB-A-1 557 421 (American Cyanamid process) of later publication date which, notwithstanding that the Libman article is mentioned in its preamble, describes hydrolysis with a dilute mineral acid, but makes no mention of neutral hydrolysis conditions and particularly no mention of hydrolysis in aqueous ethanol. It is concluded by the applicant from this that the latter system could not have been obvious since neither Shell nor American Cyanamid had taken any advantage of the allegedly obvious solvent system. This argument is unconvincing partly for reasons already explained in Section 8. In addition, the American Cyanamid document is concerned with the problem of improving the preliminary halogenation step (page 2 lines 3-15), which is thus quite different from the sole interest here; i.e. the avoidance of acid conditions. Of course this document cannot itself be taken into account for the assessment of the inventive step, because it was not published before the priority date of the application in suit.

12. Our conclusions above extend not only to a process according to claim 1, but also for those of claims 2-10. These claims are dependent from claim 1 and fall with the same in the absence of any feature introducing non-obvious subject matter.

For these reasons it is decided that:

The appeal against the decision of the Examining Division 002 of the European Patent Office dated 16 February 1981 is rejected.

The Registrar:

J. Rückerl

The Chairman:

D. Cadman