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D E C I S I O N
of 14 March 2003

Case Number: T 0565/00 - 3.3.6

Application Number: 93401320.2

Publication Number: 0572304

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Language of the proceedings: EN

Title of invention:

Carbon dioxide in neutral and alkaline sizing processes

Patentee:

CANADIAN LIQUID AIR LTD L'AIR LIQUIDE CANADA LTEE

Opponent:

AGA AB

Headword:

Carbon dioxide in sizing processes/CANADIAN LIQUID AIR

Relevant legal provisions:

EPC Art. 54, 56

Keyword:

"Main request: novelty of a process - no (implicit disclosure of all process features in a citation)"
"Auxiliary request: inventive step - no (selection of measures which are technically feasible in order to put into practise an incentive in the prior art)"

Decisions cited:

G 0002/88

Catchword:

-



Case Number: T 0565/00 - 3.3.6

D E C I S I O N
of the Technical Board of Appeal 3.3.6
of 14 March 2003

Appellant: CANADIAN LIQUID AIR LTD
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 25 April 2000
revoking European patent No. 0 572 304 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman: P. Krasa
Members: G. Dischinger-Höppler
C. Rennie-Smith

Summary of Facts and Submissions

I. This appeal is from the decision of the Opposition Division to revoke European patent No. 0 572 304 relating to the use of carbon dioxide in neutral and alkaline sizing processes. The decision was based on a set of five claims as granted as a main request, on a first auxiliary request containing only amendments to the description, and on two further sets of amended claims according to second and third auxiliary requests.

Claim 1 of the granted set of claims reads (after amendment of an obvious clerical error):

"1. A process for sizing paper comprising:

forming (12, 14) an aqueous pulp of cellulosic paper-forming fibers and an aqueous vehicle,

contacting the fibers in said aqueous pulp with an alkylketene dimer sizing agent at a non-acidic pH,

dissolving carbon dioxide gas (64, 68, 66, 54) in the aqueous vehicle to provide a catalytic amount of bicarbonate ions for the reaction between the alkylketene dimer sizing agent and the cellulose of the fibers, and

forming a paper from said pulp."

II. A notice of opposition had been filed against the granted patent wherein the Respondent (Opponent) sought revocation of the patent inter alia on the grounds of Article 100(a) EPC, i.e. for lack of novelty

(Article 54 EPC) and lack of inventive step (Article 56 EPC).

The opposition was based on eleven documents including the following:

E2: T. Lindström et al., "On the mechanism of sizing with alkylketene dimers, part 4", Nordic Pulp and Paper Research Journal No. 2/1986, pages 39 to 45; and

E8: EP-A-0 281 273.

The Respondent further relied on document

E12: T. Lindström et al., "On the mechanism of sizing with alkylketene dimers, part 2", Nordic Pulp and Paper Research Journal No. 1/1986, pages 34 to 42

filed late during the opposition proceedings.

- III. Oral proceedings before the Opposition Division were postponed, to give the Appellant (Proprietor) upon its request sufficient time to consider the late-filed document.
- IV. In its decision, the Opposition Division held that the subject-matter of the claims as granted according to the Appellant's main request and as amended according to the second and third auxiliary requests was novel due to the feature, which was not disclosed in documents E12 and E2, of "dissolving carbon dioxide" to provide a catalytic amount of HCO_3^- (bicarbonate) ions in the aqueous vehicle. Opinion G 2/88 of the Enlarged Board of Appeal (OJ EPO 1990, 93, corr. 469) was

referred to in this respect. The claimed subject-matter was, however, not inventive in view of document E12 as the closest prior art since it was common knowledge that HCO_3^- ions may be generated by dissolving CO_2 (carbon dioxide) gas in water, this being an obvious option in particular where the addition of NaHCO_3 (sodium bicarbonate) was not desired. Further, document E8 confirmed that the introduction of CO_2 gas into the pulp was technically feasible.

The first auxiliary request which contained merely an amendment in the description was held inadmissible under Rule 57a EPC.

- V. An appeal was filed against this decision. Subsequent to the Respondent's written counter-statements and to the Board's communication of 18 October 2002, the Appellant in its letter of 17 January 2003 withdrew its objection to late filing of document E12 and filed one single auxiliary request. The only claim of this request differs from Claim 1 as granted by replacing the term "dissolving" by "injecting" and by the addition at the very end of the claim of the feature "wherein said carbon dioxide is introduced into a flowing stream of the aqueous pulp (52), said stream (52) flowing at a liquid velocity effective to producing turbulent mixing and a hydraulic residence time of carbon dioxide in the flowing stream of at least 30 seconds".
- VI. Oral proceedings were held before the Board of Appeal on 14 March 2003 in the absence of the Appellant as announced by its letter of 17 January 2003.
- VII. The Appellant submitted the following arguments in

writing:

- The claimed subject-matter was novel since none of the cited prior art documents disclosed the sizing of paper with AKD (alkylketene dimer) at non acidic pH, with generation of HCO_3^- ions by injecting CO_2 gas into the aqueous support in an amount sufficient to catalyse the reaction between the AKD and the fiber while staying at non-acidic pH conditions.
- The technical problem to be solved in view of document E2 as the closest prior art was to avoid using NaHCO_3 for generation of HCO_3^- ions including the disadvantages of an imprecise dosage of NaHCO_3 and the introduction of Na^+ ions which was detrimental in the process circuit.
- None of the cited prior art documents gave a skilled person any incentive to solve this problem by injecting and dissolving CO_2 gas into the aqueous vehicle to generate a catalytic amount of bicarbonate ions while maintaining a non acidic pH.
- The feature "... to provide a catalytic amount of bicarbonate ions ..." was per se clear and distinguishing over the prior art since no catalytic amount of HCO_3^- ions was generated if CO_2 was injected below pH 7 and above pH 9.
- In document E12 a different reaction rate was observed in tap water as compared to deionized water and it was suggested that these differences could come from CO_2 absorbed from the air at pH 8

and 10, but it was not indicated that the absorbed CO_2 would increase the rate of the sizing reaction.

- According to document E8, the function of CO_2 was to control the pH of the solution, preferably to decrease the pH below 7. HCO_3^- ions were not mentioned in this document.

VIII. The Respondent filed a figure on carbonic acid dissociation (as presented at the 9th PTS-CTP Deinking Symposium, Munich, May 9-12, 2000) and submitted in writing and at the oral proceedings the following arguments:

- Opinion G 2/88 was not applicable in the present case since the claimed process did not include a new technical effect. Instead, the process of Claim 1 was anticipated by the disclosure of document E12 that the HCO_3^- ion was a strong AKD reaction catalyst which increased the reaction rate and which could be provided by absorbing at pH 8 and thereby passively dissolving CO_2 from the air into the aqueous vehicle.
- The claimed process was, in any case, not inventive over document E12 since injecting CO_2 gas was the only technically feasible way of dissolving CO_2 in an aqueous vehicle on an industrial scale such as in paper making and since it was part of the skilled person's general knowledge that the pH should not go below 7 when sizing with AKD. The catalytic amount was no distinguishing feature since it was not defined in the patent in suit, so that any amount of HCO_3^- ions was sufficient for that purpose.

- As regard the Appellant's auxiliary request, a skilled person would get from document E8 the information required to know that the CO₂ should be added at the discharge side of the pump so that it was intimately mixed into the turbulently flowing pulp and adequate contact time was provided for the CO₂ to dissolve.

IX. The Appellant, in writing, requested that the decision under appeal be set aside and that the patent be maintained as granted (main request) or on the basis of its single auxiliary request filed on 17 January 2003.

The Respondent requested that the appeal be dismissed.

Reasons for the Decision

1. Main request
 - 1.1 Novelty of the subject-matter of Claim 1 has been contested in view of document E12 which is a scientific article entitled "On the mechanism of sizing with alkylketene dimers; Part 2. The kinetics of reaction between alkylketene dimers and cellulose".
 - 1.2.1 The article is based on lab scale experimental results with bleached kraft pulp using either tap water or deionised water as the aqueous vehicle, wherein AKD was added to the aqueous stock and paper sheets are formed from this stock after sufficient contact time (30 s) between the AKD and the pulp (page 34, summary, second paragraph and right-hand column, last paragraph to page 35, left-hand column, third paragraph). It was found that the reaction between the AKD and the

cellulose of the fibers essentially takes place if the paper sheets are almost dry after a dry or wet curing treatment (page 34, Summary, second paragraph and right-hand column, lines 14 to 22). The experiments were carried out at pH 4, 6, 8 and 10 (in particular Figures 1 to 8) and showed that the rate of reaction increased with the alkalinity, i.e with increasing pH, both after dry and wet curing, as well as when using tap and deionised water (page 36, left-hand column, lines 1 to 9, page 37, left-hand column, last sentence above Figure 5 and right-hand column, last sentence above Figure 7 to page 38, left-hand column, first line below Figure 8).

1.2.2 However, it is stated in document E12 that the reaction rate is slower in deionized water than in tap water and that the pseudo first order reaction rate constant K_1 (min^{-1}) given in Table 4 shows for the former a decreased reaction rate over the whole pH range. Indeed, Table 4 contains K_1 values for tap water and deionised water at pH 4, 6, 8 and 10 which are throughout higher in tap water and it is noted "that CO_2 is probably absorbed at pH 10 in tap water and at pH-values exceeding pH 8 in deionized water" or "that CO_2 from air may have been absorbed during sheet handling" and that " HCO_3^- is a strong reaction catalyst" (page 38, left-hand column, below Figure 8 and Table 4). From the analysis in Table 1 (page 35) it is known that tap water already contains this strong catalyst HCO_3^- , typically in a concentration of 0.48 mmol/l.

1.3 Thus, given this information, it is evident that the higher reaction rates given for tap water in Table 4 of document E12 are due to its content of HCO_3^- ions right from the start and that the increase of the K_1 values,

at pH 10 for tap water and above pH 8 in deionised water, is due to absorption of CO₂ from the air at alkaline pH into the aqueous vehicle either of the pulp or of the wet sheets formed therefrom (see also point 1.5.5 below). Such absorption of CO₂ occurs as a technical inevitability during the subsequent dissolution in the pulp of CO₂ under the formation of H₂CO₃ (carbonic acid) which in turn will dissociate, depending on the pH, in the manner well-known to the skilled person as confirmed by the figure on carbonic acid dissociation filed by the Respondent, to produce HCO₃⁻ ions, the maximum concentration of which are obtainable at a pH value of between 7 and 9.

- 1.4 The Appellant's arguments in favour of the novelty of the claimed process over that disclosed in citation E12 were the following:

It was not known from document E12 to inject CO₂ gas in order to produce HCO₃⁻ ions in a catalytic amount, i.e. in a sufficient amount to have a catalysing effect on the reaction between AKD and cellulose, while staying at non-acidic pH conditions. In particular, it was argued that the amount of CO₂ present in air was low as was its solubility in water and that the term "probably absorbed" in document E12 did not suggest the injection of CO₂ into the pulp. The term was rather used to explain the discrepancies contained in Table 4 insofar as the ratios K₁(tap water) : K₁(deionized water) were inconsistent with each other at the various pH levels. Nor did document E12 indicate that CO₂ absorbed from the air would increase the rate of the sizing reaction. The Appellant agreed that CO₂ injection generates a maximum amount of HCO₃⁻ ions when the pH of the aqueous medium was maintained at between 7 and 9 but concluded that,

as a consequence, below pH 7 and above pH 9 a catalytic amount of HCO_3^- ions was not adequately provided.

1.5.1 These arguments are not convincing since the process of Claim 1 does not call either for a particular method of "dissolving of carbon dioxide gas in the aqueous vehicle" such as actively introducing the CO_2 gas (e.g. by injection), or for a particular "catalytic" amount, let alone for a maximum amount obtainable at pH 7 to 9. Considering that CO_2 injection decreases the pH of an aqueous solution, as submitted by the Appellant, Claim 1 is not even limited to conditions where the pH remains non-acidic after dissolving the CO_2 gas in the pulp.

1.5.2 Instead Claim 1 covers, in the Board's judgment, any process where paper is made from an aqueous pulp which is formed from cellulosic paper-forming fibers and water and contacted with AKD at non-acidic pH, i.e. above pH 7, and in which pulp HCO_3^- ions are provided by CO_2 gas entering the aqueous medium, be it by active injection or by passive absorption from the air, to catalyse the reaction between AKD and cellulose or, in other words, to increase the rate, i.e. the velocity, of that reaction, without being limited to conditions where the pH remains non-acidic after CO_2 addition.

1.5.3 Such a process was, however, already suggested in document E12 where it is stated that HCO_3^- ions, probably generated by absorption of CO_2 from the air at a pH above 8, act as catalyst for the AKD sizing reaction. This finding is confirmed in Table 4 of document E12 showing an increased reaction rate at non-acidic pH (in particular above pH 8) even in deionised water (see above point 1.2.2).

1.5.4 The ratios $K_1(\text{tap water}) : K_1(\text{deionized water})$, calculated for whatever reason by the Appellant for the different pH levels (see above point 1.4), are not mentioned in document E12, let alone any discrepancies or inconsistencies in this respect. Whilst the differences in these values may be based on various reasons, such as e.g. the different content of ionic species in tap and deionized water, possibly corresponding to a different CO_2 absorption equilibrium, and/or a different dissociation behaviour of H_2CO_3 at the different pH levels, it is nevertheless clear that document E12 in Table 4 in combination with the corresponding explanations suggests that the reaction rate increases at a pH above 8 even for deionised water due to CO_2 which has probably been absorbed dissociating after dissolution into the strong HCO_3^- catalyst.

1.5.5 The Board agrees that the only explanation given in document E12 for the results in the said Table 4 is that they are "**probably**" caused by the absorption of CO_2 . It is only for that reason that the process of Claim 1 is not explicitly disclosed in document E12.

However, as conceded by the Appellant, CO_2 is usually always present in the atmosphere. Therefore, to work in an CO_2 -free environment would have required particular measures to remove this gas from the air. No such measures were reported in relation to the experiments of document E12 which, therefore, have not been carried out in the absence of CO_2 . Otherwise the suggestion in document E12 that CO_2 absorption might have occurred would make no sense. It is generally known in the art that CO_2 is absorbed by an aqueous medium, in particular if the pH of the medium is high. Therefore, a skilled reader would infer from document E12 the implicit

teaching that CO₂ was actually absorbed and consequently dissolved in the aqueous vehicle. This conclusion of the skilled reader is supported by the findings in Table 4 of document E12 (see points 1.2.2 and 1.5.4) for which the Appellant did not provide any other plausible explanation.

- 1.6 The decision under appeal referred to the Enlarged Board's of Appeal opinion G 2/88 in support of the novelty of the claimed subject-matter. However, Claim 1 does not relate to a new use of a known compound to achieve an effect as does opinion G 2/88 (see reasons Nos. 5.1 and 7) but to a process for the manufacture of a product. This opinion is thus not applicable in the present case.

The Board holds, therefore, that no technical process feature apt to provide novelty over the process disclosed in document E12 is added simply by indicating, in accordance with an embodiment covered by present Claim 1, that it is **actually** this absorption of CO₂ which enhances via dissociation into HCO₃⁻ ions the reaction rate in AKD sizing.

- 1.7 The Board therefore concludes that the process of Claim 1 is anticipated by the disclosure of document E12.

2. *Auxiliary request*

2.1 Amendments

By the amendments made to the claims of the auxiliary request, for which a basis can be found in the application as originally filed, the scope of

protection has been limited. The requirement of Articles 123(2) and (3) EPC are, therefore met. The amended claims also comply with the requirements of Article 84 EPC. This not having been contested by the Respondent, no further comment on that matter is necessary.

2.2 Novelty

Novelty of the subject-matter of Claim 1 over the disclosure of document E12 has been acknowledged by the Respondent due the new technical features of how to add the CO₂ to the aqueous vehicle, namely by injecting it into the pulp flowing at a velocity effective to produce turbulent mixing, and a residence time of the CO₂ in the flowing pulp of at least 30 seconds.

The Board agrees that these features are not mentioned in document E12 or, in combination with the other features of Claim 1, in any of the other cited prior art documents. The Board is, therefore, satisfied that the subject-matter of Claim 1 is novel.

2.3 Inventive step

2.3.1 The patent in suit is concerned with the general technical problem in the field of AKD sizing of paper in neutral or alkaline conditions that the reaction rate between AKD and cellulose is slow (column 1, line 56 to column 2, line 1).

2.3.2 The parties disagreed on the issue of whether document E2 or document E12 should be considered as the closest prior art. Both documents belong to the same series of articles under the common heading "On the mechanism of

sizing with alkylketene dimers". Document E2 relates as "Part 4" of this series to a problem related to that in the patent in suit, namely to "The effects of HCO_3^- ions and polymeric reaction accelerators on the rate of reaction between alkylketene dimers and cellulose". It has been found that from the various additives investigated (page 41, Table 2) in this respect, HCO_3^- and cationic basic amines were the most efficient sizing accelerators (page 42, left-hand column, fourth paragraph). Document E12, as mentioned under 1.1 above, is part 2 of the same series and concerns the kinetics of the AKD sizing reaction. It includes, however, the issues concerning the rate of reaction and the catalytic influence of HCO_3^- ions (Summary, page 39, left-hand column, third full paragraph and Table 7). The Board holds, therefore, that both documents are equally suitable as a starting point for assessing inventive step.

2.3.3 Technical problem and solution

According to documents E2 and E12 the HCO_3^- ions are preferably provided by adding NaHCO_3 (document E2, Table 2, document E12, Table 7).

Whilst the Respondent did not accept the Appellant's argument that the introduction of Na^+ ions might be a problem in the sizing process, the parties nevertheless agreed insofar as the technical problem solved by the claimed process in view of such prior art can be seen in avoiding the using of NaHCO_3 for HCO_3^- generation for one reason or another. The Board also follows this approach, the more so as it is evident that this technical problem is solved by injecting CO_2 gas into the aqueous pulp under conditions which provide

effective mixing and a residence time sufficient for the CO₂ to be dissolved in the pulp. The Board considers it credible that this can be achieved by injecting the CO₂ gas at a point where the liquid velocity is high enough to produce turbulent mixing and which allows a hydraulic residence time of the CO₂ in the flowing stream of at least 30 seconds in accordance with the claimed process.

2.3.4 It remains to be decided whether, in view of the cited prior art documents, it was obvious for someone skilled in the art to solve this technical problem by the above means.

2.3.5 It follows from the above (see in particular 1.2 and 1.5) that the experiments disclosed in document E12 which have been carried out in a laboratory suggest CO₂ gas absorbed from the air as being a possible source for HCO₃⁻ generation. Whilst it is true, as indicated by the Appellant, that injection of CO₂ gas is not suggested in document E12, it is, nevertheless, obvious for those skilled in the art that it is not technically feasible in a large scale process simply to wait and expect that the gas be passively absorbed in amounts sufficient to accelerate the AKD sizing reaction to the same extent as would be possible by adding NaHCO₃. On the other hand, the technical possibilities for introducing a gaseous compound into a liquid in a practicable manner are limited and the Appellant has not provided any evidence that a skilled person would select any other methods than absorption and injection. Thus, the Board agrees with the Respondent that for those skilled in the art injection is at least the most convenient way of introducing the CO₂ gas into the pulp on a large scale.

2.3.6 The Appellant did not comment on the effects provided by the features relating to the turbulent mixing and the residence time of at least 30 seconds of the CO₂ gas in the pulp. The patent in suit merely mentions in this respect that the point of injection is located downstream of a pump where the velocity of the pulp is sufficient to provide turbulent mixing in a feed line which is long enough to provide "in this way adequate dissolution of the carbon dioxide in the flowing stream is achieved" (column 4, lines 45 to 49). Certainly, this does not necessarily mean that this way is the best one or the only one for mixing and dissolving the gas "adequately".

In the Board's opinion, it is, however, within the general knowledge of those skilled in the art that a reaction catalyst should be evenly distributed in the reaction medium to provide homogenous reaction and that a retention time is needed for dissolution of the CO₂ in the aqueous vehicle, as a prerequisite for the generation of HCO₃⁻ ions (see 1.3). Therefore, the skilled person is in any event required to take such steps.

The Appellant did not provide evidence as to any unexpected effect such as a particular advantage of the claimed injection at a point where the velocity of the liquid produces turbulent mixing and which allows a hydraulic residence time of the CO₂ in the flowing stream of at least 30 seconds. Further, it is apparent from document E8 that there existed no prejudice in the technical field of paper making against doing so since it discloses exactly the same procedure of injecting CO₂ under turbulent conditions at the discharge side of a pump in order to effect acidification, or by

implication dissolution of the CO_2 , in the subsequent line (column 2, lines 47 to 54 and Figure).

The features related to mixing and residence time are, therefore, considered to be one option amongst others which a skilled person would select according to circumstances in order to find adequate conditions to generate HCO_3^- ions from CO_2 gas in an industrial process.

- 2.4 Therefore, the Board concludes that, for the purpose of avoiding NaHCO_3 addition as a source for HCO_3^- generation as disclosed in documents E2 and E12, the skilled person, following a suggestion in document E12 to generate it from CO_2 , would have tried to produce the HCO_3^- ions efficiently by injecting CO_2 under conditions known to produce effective mixing and dissolution in the pulp.

Consequently the subject-matter of Claim 1 lacks an inventive step and does not meet the requirements of Article 56 EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

G. Rauh

P. Krasa