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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Photographic element containing substituted 1-phenyl-3-pyrazolidinone electron transfer agent

Klassifikation / Classification / Classement : G03C 5/54

ENTSCHEIDUNG / DECISION
vom / of / du 15 December 1987

Anmelder / Applicant / Demandeur : Eastman Kodak Company

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant : Fuji Photo Film Co., Ltd.

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step (affirmed)"

Leitsatz / Headnote / Sommaire

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Case Number : T 337/86

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 15 December 1987

Appellant : (Proprietor of the patent)
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Decision under appeal : **Decision of the Opposition Division of the European Patent Office dated 29 July 1986 revoking European patent No. 0 009 837 pursuant to Article 102(1) EPC**

Composition of the Board :

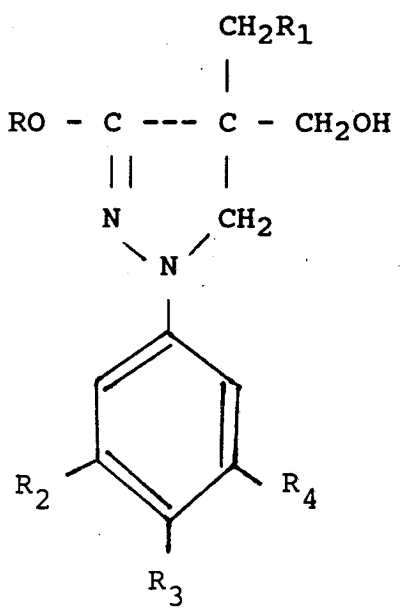
Chairman : P. Lançon
Members : G. Szabo
E. Persson

Summary of Facts and Submissions

I. European patent No. 9837 was granted with six claims on 22 February 1984, in response to European patent application 79 200 530.8.

Claim 1 of the patent-in-suit reads:

A color diffusion transfer photographic element comprising a support having thereon at least one photosensitive silver halide emulsion layer having associated therewith a redox dye releasing compound capable of being oxidized by an oxidized silver halide electron transfer agent characterised in that said element contains a substituted 1-phenyl-3-pyrazolidinone silver halide electron transfer agent or a precursor having the formula:



wherein:

R is hydrogen or a hydrolyzable moiety;
R_1 is hydrogen or a hydroxy group;
R_2, R_3 and R_4 each represent hydrogen, hydroxy, an alkyl group of 1 to 3 carbon atoms, an alkoxy group of 1 to 3 carbon atoms, a phenyl group, $--NHSO_2R_5$, $--O(CH_2)_nOR_6$, $--(CH_2)_nOR_6$, $--O(CH_2)_nC_6H_5$, or $--(CH_2)_nNHSO_2R_5$;
R_5 is hydrogen or an alkyl group of 1 to 3 carbon atoms;
R_6 is hydrogen or a methyl group;
n is a positive integer of 1 to 2;
with the proviso that at least one of said R_2, R_3 and R_4 is a substituent other than hydrogen.

II. The patent was opposed by the Respondent on 22 November 1984 on the grounds of Article 100(a) in conjunction with Article 56. The opposition was supported inter alia by the following documents:

(a) Photographic Science and Engineering, vol. 20, No. 4, July/August 1976, pages 155 to 160,

(b) W.E. Lee in C.E.K. Mees, "The Theory of the Photographic Process", rev. (3rd) ed., T.H. James, ed., The Macmillan Co., New York 1966, page 300,

(c) W.E. Lee in C.E.K. Mees, "The Theory of the Photographic Process", rev. 4th ed., T.H. James, ed., The Macmillan Co., New York 1977, page 322.

III. In a decision dated 29 July 1986, the Opposition Division revoked the patent. The reasoning was that document (c) taught:

(i) that 1-phenyl-3-pyrazolidinones were known as electron transfer agents (ETAs) in black and white or colour photography;

(ii) that the p-tolyl homologue was somewhat more active than Phenidone (1-phenyl-3-pyrazolidinone) itself, and

(iii) that Phenidone derivatives with substitutions on the 4-position of the pyrazole ring especially 1-phenyl-4-methyl-4-hydroxymethyl-3-pyrazolidinone, compound (57), had great stability, were much more soluble in aqueous solution and were highly resistant to hydrolysis in an alkaline medium.

Consequently, it was obvious to combine the substitutions at the pyrazole ring 4-position, as taught in compound (57) in document (c), with the p-tolyl substitution which was more active to obtain an ETA which gave faster access times and lessened bubble formation, since faster access times were caused by high activity, and lessened bubble formation and crystal growth was caused by high solubility and high resistance to hydrolysis of an ETA.

IV. A Notice of Appeal was filed by the Appellant, who is the Patentee, on 22 September 1986, the fee being paid on the same day. The statement of grounds for appeal was filed on 1 December 1986.

Oral proceedings were held before the Board on 15 December 1987.

V. In their written submissions and at the oral proceedings the Appellant argued substantially as follows:

(i) The most relevant prior art was document (c), in particular its disclosure of compound (57), in the context of the use of such compounds as activators in colour diffusion transfer elements, as disclosed in the standard text (a), the compound (57) being

particularly soluble and resistant to alkaline hydrolysis;

Compared with this, the invention achieved the surprising results of (1) shorter access times, (2) lessened crystal growth/bubble formation on the processed print, and (3) the possibility of using lower quantities of ETA - the results (1), (2) and (3) being achieved simply by substituting a methyl group on the benzene ring of compound (57);

(ii) The Opposition Division had misdirected itself in concluding that the skilled person would inevitably have chosen the p-tolyl substitution for shorter access times and 1-phenyl-4-methyl-4-hydroxymethyl-3- pyrazolidone for lessened bubble formation, since there was no evidence that these parameters were related to the actually disclosed qualities of higher activity and higher solubility/resistance to alkaline hydrolysis respectively;

(iii) The idea that the skilled person would turn automatically to modifying the ETA was in any case erroneous. There were many more obvious strategies available to solve the technical problem of providing the effects (1), (2) and (3);

VI. The Respondent presented substantially the following arguments:

(i) The nearest prior art was represented by document (a), disclosing unspecified derivatives of Phenidone, and not document (c); because (a) referred specifically to (b) as a source of suitable Phenidone derivatives and disclosed the p-tolyl homologue; updating this knowledge by

looking at the later edition (c) revealed additionally the usefulness of the 4-hydroxy-methyl substitution on the pyrazolidone ring as a way of increasing solubility and resistance to alkaline hydrolysis; thus inviting the skilled man to combine both types of substituents to obtain the higher activity while compensating for any loss of solubility occasioned by introducing the methyl group on the benzene ring;

(ii) The skilled person would realise that higher activity would lead to shorter access times and that higher solubility/resistance to alkaline hydrolysis would lead to reduced crystal growth/bubble formation respectively on the processed prints;

(iii) From the description of the patent-in-suit it was clear that the only strategy for improving access times which would confront the skilled man consisted in arriving at a suitable derivative of Phenidone and not in any of the alternative strategies suggested by the Appellant.

VII. The Appellant requests that the decision of the Opposition Division be set aside and the patent maintained as granted. The Respondent requests that the appeal be dismissed.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. The patent-in-suit relates to a colour diffusion transfer (CDT) photographic element comprising a support having thereon at least one photo-sensitive silver halide emulsion layer having associated therewith a redox dye releasing compound capable of being oxidised by an oxidised silver halide electron transfer agent (ETA), the ETA being a 1-phenyl-3-pyrazolidinone compound or precursor.

3. Such CDT elements are known from the prior art of which document (c) represents, in the Board's view, the closest state of the art.

4. Document (c), which is regarded by those skilled in the art as the Photographic technician's "Bible", discloses various derivatives of 1-phenyl-3-pyrazolidone (Phenidone) for use as ETA's in CDT transfer elements. It suggests that the p-tolyl homologue is somewhat "more active" than the unsubstituted Phenidone, but states that most of the pyrazolidones substituted on the phenyl group are much less soluble than Phenidone itself (left-hand column, last paragraph, lines 6-9). Other derivatives of Phenidone substituted at the 4 position of the pyrazolidone ring by a hydroxyalkyl group are, however, said to be much more soluble in aqueous solution and in an alkaline medium to be highly resistant to hydrolysis. In this connection compound (57), which is 1-phenyl-4-methyl-4-hydroxymethyl-3-pyrazolidone, is mentioned as an example which is more soluble and resistant to alkaline hydrolysis. Since the other compounds disclosed in this citation, including the p-tolyl derivative of Phenidone itself, are structurally more remote from what is claimed in the patent-in-suit, compound (57) is regarded as the closest approach to the subject-matter claimed.

This compound, which has been tested under the designation "control compound B" in the patent specification, however, exhibits the disadvantages of producing a large number of bubbles which are visible and physically degrade the image on the processed print, as well as having rather slow access times (cf. Table on page 13, control compound B, which is compound 57 itself, producing 168 bubbles/cm^2 in the visually sensitive low colour density - D_{min} - area of the print, and Table on page 11 showing an initial access time of 52 seconds).

5. Compared with this state of the art, the technical problem was to be seen in providing a lessening of bubble growth and faster initial access times. The solution, according to Claim 1 of the patent-in-suit, lay in the modification of compound (57) by the provision of one or more substituents R_2, R_3, R_4 on the benzene ring, as specified in the claim, as well as the option of generalising the 4-methyl group on the heterocyclic ring to a CH_2R_1 group as specified in the claim. The compound carrying a 4-methyl group on the benzene ring is disclosed in the patent specification as compound 1, and is found to produce zero bubbles/cm^2 on the finished print compared with 168 bubbles/cm^2 for control compound B, which is compound (57) of the cited art (c) and to have an initial access time of 31 seconds compared with 52 seconds for the same control compound (cf. Table on page 13 and Table on page 11, respectively). It is evident that the technical problem is credibly solved.

6. No document of the prior art discloses a colour diffusion transfer photographic element comprising a compound of the formula in Claim 1 and thus the subject-matter claimed in the patent-in-suit is novel. Novelty was in any case not impugned during the proceedings.

7. In the assessment of inventive step, the question has to be asked, whether the skilled person would have been induced or encouraged to modify compound (57) of document (c) in the photographic element in such a manner as to arrive at something falling within the scope of Claim 1, by the hope or expectation of lessened bubble growth and/or shorter initial access times. The simplest modification of compound (57), which will lead to something falling within the claims of the patent-in-suit, is the provision of a methyl substitution at the 4-position of the benzene ring to give the p-tolyl homologue of compound (57). This already shows a significant reduction of bubble formation and a decrease of access time.

8. The question thus arises, was there any hint or indication for the skilled man from the prior art (including general knowledge) that such a substitution would result in such improvements in the properties of the compounds?

9. In arriving at its decision to revoke the patent, the Opposition Division considered that the knowledge that compound (57) was resistant to alkaline hydrolysis would have indicated to the skilled person that its tendency to bubble formation would also be reduced because the products of decomposition of the pyrazolidone ring could cause bubble formation. Furthermore, the knowledge that the p-tolyl derivative of Phenidone was "somewhat more active" would have been an indication to the skilled person that the "p-tolyl" homologue had faster access times, because higher activity meant faster reaction rates and faster reaction rates are necessary for shorter access times. Thus, it would have been obvious to obtain both advantages by combining the substituents in the form of the p-tolyl homologue of compound (57) in (c).

It will be necessary to examine both aspects of this argument in turn.

10. Firstly, although it is not inconceivable that alkali induced decomposition of the ETA might lead to bubbles on the print, there is no concrete evidence before the Board that this is actually what happens. In any case, the increased resistance to hydrolysis was, according to (c), due to the introduction of a 4-hydroxymethyl substitution to the pyrazoline ring, which applies equally to compound (57) and to those covered in the present patent. Unexpectedly, control compound (57) itself, in spite of its high resistance to alkaline hydrolysis, nevertheless produced 168 bubbles/cm^2 on the D_{min} part of the print compared with the same compound substituted with a methyl group in the benzene ring (compound 1) which under the same conditions produced zero bubbles (cf. Table on page 13). In the light of this, the Board is not prepared to accept the unsupported allegation of the Respondent that resistance to alkaline hydrolysis would have been

understood by the skilled person to correspond to a promise of lessened bubble formation on the processed print, in consequence of some other modification of the molecule indifferent to the action of alkali. It is conclusive for the Board that the presence of the 4-methyl group on the 1-phenyl substituent is correlated with the unexpected drastic reduction of bubble formation irrespective of the retention of the identical set of other substituents in the molecule.

The skilled person would not therefore have been induced, at least on the basis of a hope of reduced bubble formation, to adopt any particular substitutions at the 4-position of the pyrazolidone ring.

11. The second question, namely that of the relationship to access time is more complex. At the oral hearing, the Respondent submitted that "activity" meant faster reaction time or that one could use less ETA. The Appellant accepted only the former alternative. The only reference to "activity" of the ETA in the patent-in-suit is in Example 4; however this is in the context of "threshold speed" which is in effect a measure of film exposure sensitivity and as such has nothing to do with access times. It does, however, appear to the Board that higher "activity", while not necessarily predicating shorter access times, is not inconsistent with shorter access times, particularly insofar as it implies a faster reaction speed of the ETA. The introduction of the methyl substitution in the para-phenyl position would have had some attraction to the skilled person considering modifications of the molecule provided there are no other discouraging circumstances.

12. The argument of the Appellant, that there are other alternative strategies which would equally well suggest

themselves, such as the use of different redox dye releasing agents, boosters or different mordants, was not supported by any documentary evidence from the prior art beyond an assertion that a patent for boosters had been applied for. The Board considers that the skilled man must be expected to read what is put in front of him. Document (c) is an extract from a basic reference book for the photographic technician. In the left-hand column of page 322 of (c) it is stated that "The p-tolyl homologue is somewhat more active than Phenidone." The next sentence, however, begins "Most of the phenyl-substituted pyrazolidones are much less soluble than Phenidone." The underlining is by the Board. The contrast here is between the words "somewhat more active" and "much less soluble".

13. As was argued especially in the oral proceedings, a sufficient solubility of the ETA was an absolute pre-requisite of a CDT system, since diffusion through the multiple layers of the photographic element was an essential mechanism in the formation of the picture and if the ETA was insoluble it could not diffuse. Thus, the reference to "much less" solubility, is a very strong discouragement to the skilled person interested in obtaining high activity from using a p-tolyl derivative of Phenidone. Moreover, even the promise of the higher activity is watered down by the word "somewhat". Taking the teaching in (c) as a whole, the Board considers that the encouragement to try the p-tolyl derivative to get higher activity is outweighed by the discouragement in much stronger terms arising from the threat of loss of the high solubility already achieved by the 4-hydroxymethyl substitution of the pyrazolo-ring in compound 57 (right-hand column of page 322, lines 10-12). Therefore, such a teaching read by the skilled person cannot amount, on the whole, to an encouragement for increasing activity by the use of the p-tolyl homologue. Consequently, no direct

unproblematic one-way street situation existed even in respect of increasing activity in respect of the p-tolyl homologue, let alone any prediction of the important further improvement in access time.

14. Thus, the Board disagrees with the view of the Opposition division that the critical feature of the solution of the technical problem was deducible in an obvious manner from the teachings of document (c). None of the other documents cited in these proceedings implied the consequences of such modification either.

15. This view is not altered by choosing, as the Respondent preferred to do, document (a) referring to "derivatives of Phenidone" as the closest prior art. The mere fact that document (b), to which (a) directly refers, is an earlier edition of (c) which mentions the p-tolyl homologue of Phenidone but not compound (57), the latter compound only appearing in the later edition, does not, in the Board's view, mean that the separate disclosures can be regarded as cumulative. The modifications of the Phenidone referred to in (a) are contextually separate. However, even if they had not been, the terms used in (b) to refer to the p-tolyl homologue are even less encouraging than those in (c), since (b) goes to say that Phenidone itself is barely soluble enough to be useful (see (b), page 300, lines 9-13).

Consequently, neither the route starting from (c), nor the route leading from (a) via (b) to (c), gives the skilled person the encouragement or inducement he needs to embark on the modification necessary to arrive at something falling within the scope of the claims of the patent-in-suit. In this connection, the remaining compounds disclosed in the documents (a), (b) and (c) are still more remote from the subject-matter of Claim 1 of the patent.

16. Thus, the subject-matter of Claim 1 involves an inventive step within the meaning of Article 56. The remaining Claims 2-6 are fully dependent on Claim 1 and therefore their subject-matters by the same token also involve an inventive step.

Order

For the above reasons, it is decided that:

1. The decision of the Opposition Division is set aside.

2. The patent is to be maintained as granted.

The Registrar: The Chairman:

F. Klein P. Lançon