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Beschwerdekammern

European Patent Office
Boards of Appeal

Office européen des brevets
Chambres de recours

Case Number: T 154 / 82

DECISION
of the Technical Board of Appeal 3.3.1
of 4 November 1983

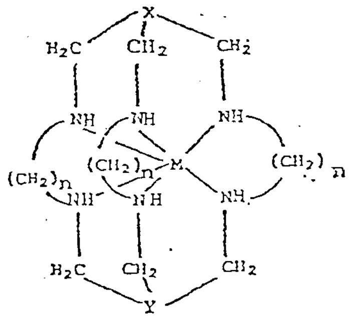
Appellant:	THE AUSTRALIAN NATIONAL UNIVERSITY Acton, Australia
Representative:	Day, Jeremy John REDDIE & GROSE 16 Theobalds Road London, WC1X 8PL England
Decision under appeal:	Decision of Examining Division 025 of the European Patent Office dated 13 May 1982 refusing European patent application No 78 300 538.2 pursuant to Article 97(1) EPC

Composition of the Board:
Chairman: D. Cadman
Member: K. Jahn
Member: L. Gotti Porcinari

SUMMARY OF FACTS AND SUBMISSIONS

I. European Patent Application No. 78 300 538.2 filed on 24 October 1978 and published on 2 May 1979 under publication No. 0 001 719, claiming the priority of two Australian prior applications of 26 October 1977 was refused by decision of the European Patent Office dated 13 May 1982 on the basis of the claims received on 26 November 1980 which have the following wording:

"1. A coordination complex of the formula:



in which n represents an integer; M represents a cobalt ion or other metal ion having at least two oxidation states; and X represents ≡C-R' and Y represents ≡N, ≡P or ≡C-R', in which R' represents a hydrogen or halogen atom, or a hydroxyl, nitro, nitroso, amino, protonated

.../...

amino, alkyl, or cyano group, or a group of the formula -COOR", -COCOOR" or -NH-COCH=CHCOOR" in which R" is a hydrogen atom or akyl group.

2. A complex according to claim 1, wherein n is an integer of from 2 to 4,.

3. A complex according to claim 1 or claim 2, wherein M represents a Rh, Pt, Cr, Cu, Re, Mo, W, Ni, V or Fe ion.

4. A complex according to claim 1, wherein n is 2, M is Co, and x and y each represent $\equiv C-NO_2$.

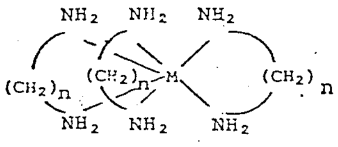
5. A complex according to claim 1, wherein n is 2, M is Co, and x and y each represent $\equiv C-NH_3^+$.

6. A complex according to claim 1, wherein n is 2, M is Pt, and x and y each represent $\equiv C-NO_2$.

7. A complex according to claim 1, wherein n is 2, M is Co, and x and y each represent ≡C-Cl.

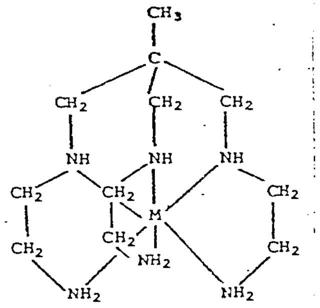
8. A complex according to claim 1, wherein n is 2, M is Co, and x and y each represent ≡C-H.

9. A process for the preparation of a complex according to claim 1, which comprises the condensation of a tris-(diamine) metal ion complex of the formula:



in which M and n are as defined in claim 1, with formaldehyde and a nucleophile appropriate for producing capping groups X and Y.

10. A process for the preparation of a complex according to claim 1, which comprises the condensation of a metal chelate of 1,9-diamino-5-(methyl)-5-(4-amino-2-azabutyl)-3,7-diazanonane of the formula:



in which M is as defined in claim 1, with formaldehyde and a nucleophile appropriate for producing capping groups X and Y.

11. A process for the production of hydrogen peroxide comprising the steps of:

(i) oxidising a coordination complex according to any one of claims 1 to 8, said metal ion being in its lower oxidation state; and

(ii) recovering hydrogen peroxide produced thereby."

II. The stated ground for the refusal was that the application lacked inventive activity in the light of the article "Sepulchrate : A macrobicyclic Nitrogen Cage for Metal Ions" in the Journal of the American Chemical Society vol. 99, no. 9, 27 April 1977, pages 3181 to 3182.

This citation discloses a coordination complex where a cobalt (III) ion is held within a cage formed by a hexadentate nitrogen-containing ligand. Such a structure is termed cage-like or "sepulchrate". Page 3182 indicates that the citation authors were investigating the preparation of Mo, W, Rh(II) and Pt(III) complexes and the capping of tris(ethylenediamine) and 1,1,1-tris(4-amine-2-azabutyl)ethane complexes with tris(methyleneamino) and substituted tris(methylenemethane) moieties. This is a clear indication that the skilled man would arrive at the claimed coordination complexes without inventive effort, and the predicted advantageous properties set forth in the cited paragraph would give the skilled man the impetus to attempt such a preparation.

The processes of claims 9 and 10 lack inventivity as well having regard to the disclosure in the above citation, where it is indicated that the complex known as / Co(sepulchrate) /$^{3+}$ (i.e. (5)-/ 1,3,6,8,10,13,16,19-octaazabicyclo/ 6.6.6 / eicosane)cobalt(III) /$^{3+}$ has been prepared by the reaction of tris(ethylenediamine)cobalt(III) with formaldehyde and ammonia (as nucleophile).

Moreover, the matter of claim 11 is considered to lack inventive step, since the use of the complex / Co(sepulchrate) /$^{3+}$ for the production of hydrogen peroxide is mentioned in the citation.

III. On 19 July 1982 the appellant lodged an appeal against the decision dated 13 May 1982. On 23 September 1982 a Statement of Grounds was filed, the substance of which is as follows: The decision reaches conclusions on the obviousness of the invention which are unjustified in view of the limited teachings of the citation and the limited knowledge of the skilled person.

The citation, originating inter alia from the three inventors of the application in suit, is a disclosure of a new structural type of cage complex and accordingly no person skilled in the art can be presumed to have any knowledge other than that taught in the citation. The only persons with the relevant background knowledge beyond that taught in the citation are the authors of the citation who are not persons skilled in the art in the sense of Article 56 in view of their privileged position.

The citation makes only a minimal disclosure regarding the production of cobalt sepulchrate complex and the skilled man would find difficulty in producing even this complex from the disclosure in the citation. The citation gives no indication whatsoever of the mechanism of reaction involved, knowledge of which would be essential before any method for the preparation of the other complexes that are now claimed could be regarded as obvious to a skilled person.

Given the lack of prior knowledge of complexes of the structural type with which the invention is concerned, and in the absence of disclosre of a mechanism for production of the very complicated molecules, the skilled person would not arrive at the carbon-capped complexes as claimed without inventive effort.

The inventiveness of the present claims both as regards complexes having two carbon caps and complexes having mixed carbon/nitrogen or carbon/phosphorous caps has been completely underestimated because of the advantage of hindsight.

IV. The Board of Appeal then, in a communication to the appellant, pointed out that the new coordination complexes as claimed must, in order to be patentable, have properties which are either unpredictable or could not be expected to that extent over the structurally closest compounds of the art.

As to the process claims, the Board ventured the opinion that the cited article gives a hint at the possibility of capping complexes of tris-(ethylenediamine) and 1.1.1-tris(4-amino-2-azabutyl)-ethane (sen) with a) tris-(methyleneamino) and b) substituted tris-(methylenemethane)-moieties. Since sen is known (Inorg. Chem. 1963, 2, 597 cf. description page 19) it was evident that in order to achieve goal a), this compound could be capped with the tris-(methyleneamino) moiety by condensation with formaldehyde and ammonia resulting in a compound with the mixed nitrogen/carbon caps.

As to goal b) the skilled man could expect that capping with tris-(methylenemethane) moieties would be achieved by condensing tris-(ethylenediamine)- complexes with

formaldehyde and compounds of the formula H_3C-substituent instead of H_3N. The only question emerging in this connection was that of the nature of the substituent. From the mode of the reaction envisaged, it was clear that only substituents which strongly activate the hydrogen atoms of the methyl group were promising. The use of such compounds in this reaction would appear obvious.

Besides, the possibility to tie the claimed complexes to a polymer chain was considered obvious in view of DE-C-1 029 807.

Moreover, the version of claim 1 was objected to with regard to the term "protonated amino group".

V. The appellant contested these arguments. The new complexes do have at least one property which is clearly unpredictable from the prior art, namely that they are so reactive in excited states that they are capable of serving as electron transfer agents in the photo chemical reduction of water to produce hydrogen. Furthermore, the fact that the complexes can be incorporated into a polymer chain involves an inventive step. The citation DE-C-1 029 807 does not relate to the incorporation of metal complexes in polymer chains, it merely relates to the incorporation of certain compounds that can exist in two oxidation states in a polymer chain to produce a resinous material that can be used as a redox material. None of the prior art that has been cited in connection with this application shows the incorporation of a metal complex in a polymer chain, for any purpose, even less to produce a polymer material with redox properties.

As to the claimed reaction, this is not a conventional one since it could not clearly be predicted that formaldehyde would react with the amino groups of the tris-(ethylenediamine) cobalt ion, and that having so reacted ammonia would then act as a capping agent. It can be seen from the specification that the mechanism proposed for the reaction is only postulated and, accordingly, it is again submitted that the skilled man is quite likely to have problems in producing the complex. The knowledge of the mechanism of a reaction is of primary importance where the document is being relied on to establish obviousness of an analogous process. Unless the mechanism of reaction is known nobody, however skilled in the art, can tell what other reactants can be used to produce analogous compounds.

It is submitted that the reasoning of the board is all based on hindsight. There is no mode of the reaction referred to in the citation and there is certainly no indication that if one is to cap the complexes with substituted tris-(methylenemethane) moieties, one should react the complex with formaldehyde and a substituted methane with a substituent which strongly activates the hydrogen atoms of the methyl group.

The appellant maintained the claims while expressing his willingness to delete claim 11. Moreover, he did not insist on the originally requested refund of the appeal fee.

VI. At the appellant's request, oral proceedings were appointed for 15 September 1983 at which the appellant failed to appear. In a letter received on 14 September 1983 it was stated that the appellant, who would not be

represented at the oral proceedings, wished the matter to be decided on the basis of the papers currently on file.

From the foregoing it must be concluded that the appellant requests that the decision under appeal be set aside and that the patent sought should be granted with claims in these terms.

REASONS FOR THE DECISION

1. The appeal is in accordance with Articles 106-108 and Rule 64 EPC; it is therefore admissible.

2. The question as to whether or not claim 1 is adequately supported by the original documents can be left in abeyance, since the application fails for other reasons.

3. As indicated in the description of the present application (cf. page 5, paragraph 2) the applicant starts from the known hexadentate complex (S) (1,3,6,8,10,13,16,19-octaazabicyclo (6.6.6.)eicosane) cobalt (111)$^{3+}$ which has been given the trivial name Co(sepulchrate)$^{3+}$ ('sepulchrate' may hereafter be abbreviated to 'sep'), and which may be prepared by condensing tris(ethylenediamine)cobalt (III) with 2 molecules of ammonia and 6 molecules of formaldehyde to form a cage-like structure, as described in I.I. Creaser, J. MacB. Harrowfield, A.J. Herlt, A.M. Sargeson, J. Springborg, R.U. Geue and M.R. Snow, J. Amer. Chem. Soc. April 27 99:9, 3181 (1977). This Co(sep)$^{3+}$ can be reduced by zinc dust to Co-(sep)$^{2+}$ which reoxidises with O_2 quantitatively to Co(sep)$^{3+}$ and hydrogen peroxide (cf. page 3181 right hand column last line to page 3182 left hand column line 1).

In the absence of other evidence, the technical problem underlying the application in suit must be seen in providing further hexadentate coordination complexes which are able to produce hydrogen peroxide (complexes with metals in their lower oxidation state) and their precursors (complexes with the metal in its higher oxidation state). This capability was clearly emphasised as the main objective in the present application (cf. original page 1 paragraph 1, pages 2, 3, 4 lines 13/14, page 8 paragraph 2, pages 12 and 22, and claim 11 on file.

In order to solve this problem, the applicant proposes complexes as set out in claims 1 to 8, a process for producing them as specified in claims 9 and 10 and finally a process for the production of hydrogen peroxide by using the complexes of claims 1 to 8, said metal being in its lower oxidation state.

4. A teaching as defined above cannot be gathered from the publications before the Board. The cited article, which indicates that complexes derived from capping tris(ethylenediamine) metal complexes with tris(methyleneamino) and substituted tris(methylenemethane) moieties are currently being investigated (cf. page 3182 right hand column, paragraph 2), is silent on the nature of the substituents for the last mentioned cap, in contrast to the definition in the present application (cf. claim 1, R'). Therefore, the application in suit is deemed to be novel.

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

As mentioned above, the cited article itself offers the prospect of the synthesis of tris(methylenmethane) capped hexadentate metal coordination complexes and points like a signpost to these compounds. In view of the structural vicinity of these carbon capped complexes vis à-vis the known nitrogen capped cobalt sepulchrates, the skilled reader would expect that the first-named complexes had rhe inherent property of producing hydrogen peroxide and, therefore, presented a solution of the envisaged problem, irrespective of the particular chemical nature of the substituent of this capping group.

Consequently, the particular choice of the said substituent as set out for the variable R in claim 1 is of no importance and might be governed by aspects of manufacture of the new complexes. Thus the subject matter of claims 1 to 8 does not involve an inventive step.

6. The appellant fails in his allegation that the discloure of the mode of manufacture of the cobalt sepulchrates described in the cited article is insufficient to enable the skilled person to succeed in the production of these compounds and, therefore, all the more so for the production of the claimed complexes. This article gives all the information needed for the production of the described cobalt sepulchrates, i.e. the three partners for the reaction: cobalt tris(ethylendiamine), formaldehyde and ammonia. Moreover, the proportion in which these reactants should normally be applied could be seen from the structure of the sepulchrates (see as well the present description page 5 paragraph 2). In the absence of evidence to the contrary, the fact that the article is silent on the solvent, the temperature and the catalyst

does not impair the feasibility of the method described, since knowledge of such features is within the ordinary skill of a chemist.

7. The appellant advances the argument that the complexes as claimed do at least have the unpredictable property of serving as electron transfer agents in the photochemical reduction of water to produce hydrogen. No evidence has been presented in this respect. Nevertheless, on page 8 of the present specification the possibility for the reduction of water to hydrogen was stated (lines 24/25) which is not disputed by the Board. However, the appellant failed to submit that this property was unique and characteristic only of the claimed complexes, but not of the known Co(sep)$^{+2}$ as well. The existence of such a significant difference in properties of structurally close related compounds is also very unlikely, since the known redox couple Co(sep)$^{2+}$/Co(sep)$^{3+}$ establishes a reduction potential of -0.45 V (cf. the above citation page 3181 right hand column last paragraph) which is roughly the same as for the claimed complexes (cf. page 10 of the present description). The identity between the authors of the above article and the inventors of the present application justifies the conclusion that the reduction potential of the known and the claimed complexes was measured under the same conditions. In any case, the appellant relinquished his opportunity of clarifying this point by not participating in the oral proceedings.

But even if the property of producing hydrogen from water was evidently inherent only in the complexes as claimed, such an additional effect would not have been crucial for the question of obviousness. Apart from that, such an effect, in the Board's view, cannot be incorporated in the

definition of a realistic technical problem. The cited article in effect, set a signpost pointing at the carbon capped complexes. This signpost could not be neglected by the skilled man who was engaged with the development of further similar complexes capable of producing hydrogen peroxide, having regard to one facet of the double problem, i.e. the production of hydrogen peroxide.

8. Moreover the appellant considers the fact that the complexes can be incorporated into a polymer chain as involving an inventive step. This advantage cannot be claimed for all complexes falling within the term of claim 1, e.g. for complexes where R' is hydrogen or alkyl, since it is stated in the present application that the substituent R' due to be tied to the polymer chain shall be suitable for that purpose such as an amino group (cf. page 8 last paragraph).

Furthermore, the idea of tying to a polymer support an organic compound which acts as a redox couple and enables the production of hydrogen peroxide is known from DE-C-1 029 807. The application of this idea to the hydrogen peroxide-producing complexes as claimed must be considered obvious as no more than a mere workshop variation.

9. For the assessment of the inventive step of the present process claims it is of minor importance that the citation does not tell the reader how the capping of tris-(ethylendiamine) complexes with substituted tris(methylenemethane) moieties would be achieved. In the light of the teaching of this document that the capping of tris-(ethylendiamine) complexes with tris(methyleneamino) moieties is carried out by reaction with formaldehyde and

ammonia, it would be crystal clear to the reader that the reaction partner, ammonia, with its active hydrogen atoms, had to be replaced by a methane compound with strongly activated hydrogen atoms. Such compounds are well known in the art (strongly activating groups like cyano, nitro, carboxylic and ketocarboxylic ester groups). Mention should be made in this context that the applicant himself considers it sufficient to define this reaction partner as a nucleophile appropriate for producing capping groups X and Y.

The appellant ventured the view that unless the mechanism of a reaction is known, nobody, however skilled in the art, can tell what other reactants can be used to produce analogous compounds. The Board does not share this position. The knowledge of the mechanism of a reaction which is of scientific interest, is of no importance to a teaching disclosed in the sense of the European Patent Convention. Moreover, in the present case, the carbon capped complexes and their manufacture were already foreshadowed in the cited article by naming the two possible groups of starting materials and the two relevant capping moieties, i.e. the tris(methyleneamino) and the substituted tris(methylenemethane) moiety. Since it was known that the capping of tris(ethylenediamine) complexes with the first mentioned cap could be achieved by reaction with formaldehyde and ammonia, it was clear that the capping of the starting complexes with the last mentioned cap would be performed in an analogous reaction with formaldehyde and a single substituted methane the nature of the substituent being clear, as set out above.

Therefore, the subect matter of claim 9 cannot be regarded as involving an inventive step.

10. The same conclusion must hold true for claim 10 which comprises the reaction of a metal chelate of 1,9-diamino-5-methyl-5-(4-amino-2-azabutyl)-3,7-diazanonane with such a nucleophile. These chelates already contain one of the two caps, i.e. the carbon cap. Again, it was clear to the skilled reader of the cited article that these chelates were well adapted to the manufacture of the coordination compounds mentioned at the end of this article, since these chelates had only to be made complete by the missing cap. It may be mentioned that the higher homologue of the starting chelate as used according to claim 10, which bears an ethyl group instead of a methyl group at the carbon cap, and its manufacture also, was described (cf. Journal of Inorganic Chemistry 2, 1963, pages 597 to 600). Therefore, the skilled man would have no difficulties in producing the starting chelates required in the process according to claim 10 in suit. Consequently, this claim must fail for lack of inventive step as well.

11. The appellant submits that the reasoning of the Board as sketched in its communication is all based on hindsight. As set out in this Board's decision T 24/81 dated 13 October 1982 "Metal refining" (O.J. EPO 1983, 133, 137 particularly paragraph 4) an ex-post-facto approach in the assessment of inventive step can be avoided if, by taking account of the technical relevant merits of the invention beyond the state of the art, the problem which objectively underlies the invention is defined and then the question is asked as to whether or not the solution of this problem as proposed by the applicant was obvious from the point of view of the problem. The Board has followed this principle in the present case. Therefore, the appellant's allegation that the Board's considerations about inventive step have been made with hindsight is unjustified.

12. As set out in detail in paragraph 5, it was obvious to provide the coordination complexes according to claims 1 to 8 for the production of hydrogen peroxide. The subject matter of claim 11 which likewise concerns this production therefore cannot be considered as involving an inventive step.

13. Because the Board has not deemed the appeal to be allowable, it follows that the request for the reimbursement of the appeal fee is rejected.

ORDER

It is decided that

1. The appeal against the decision of the Examining Division of the European Patent Office dated 13 May 1982 is dismissed.

2. The request to reimburse the appeal fee is rejected.

Cadman