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Case Number : T 33/86



D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 20 November 1987

Appellant : STAMICARBON B.V.
Mijnweg 1
NL-6167 AC Geleen

Representative : Hoogstraten, Willem Cornelis Roeland
OCTROOIBUREAU DSM
Postbus 9
NL-6160 MA Geleen

Decision under appeal : Decision of Examining Division 025 of the European
Patent Office dated 12 August 1985 refusing
European patent application No. 81 201 341.5
pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : K. Jahn
Members : C. Gérardin
G.D. Paterson

Summary of Facts and Submissions

- I. The European patent application No. 81 201 341.5 filed on 10 December 1981, published on 30 June 1982 under publication number 54 993 and claiming priority of 20 December 1980 from a previous application filed in The Netherlands, was refused by a decision of the Examining Division of the European Patent Office dated 12 August 1985.

- II. This decision was based on Claim 1 filed on 30 March 1984 and Claims 2 to 9 filed originally. Claim 1 reads as follows:

"Process for the recovery of practically radium-free calcium sulphate, yttrium and lanthanides in a phosphoric acid preparation process by digesting phosphate rock, in the presence of more than 5% by weight of sulphate ions calculated in respect of the quantity of phosphate rock, with an excess of phosphoric acid required for the formation of monocalciumphosphate, separating out the insoluble residue, precipitating calcium sulphate from the remaining solution with sulphuric acid and separating the calcium sulphate from the phosphoric acid solution formed, characterized in that the phosphate rock is digested with phosphoric acid without barium compounds being added and yttrium and lanthanides are separated out from the insoluble residue separated off, containing radium, yttrium and lanthanides."

- III. The reason given for the refusal was that the subject-matter of Claim 1 and the dependent claims did not involve an inventive step with regard to the prior art publications

(1) DE-B-2 218 382

(2) Nouveau Traité de Chimie Minérale by P. Pascal, Volume IV, 1958, pages 934 to 950

hereinafter referred to as documents (1) and (2).

More specifically, the Examining Division put forward that the subject-matter of Claim 1 differs from the teaching of document (1) in that yttrium and lanthanides are separated out from the insoluble residue and in that the digestion step is carried out without addition of barium compounds. Since phosphate rocks were known to contain yttrium and rare earths which precipitate together with other sulphates by addition of sulphate ions to the digestion medium, the separation of these elements from the residue would be self-evident for the skilled man. As far as the addition of barium was concerned document (1) describes the use of calcium sulphate precipitate as a filter aid for the removal of the precipitated sulphate impurities by control of the amount of added sulphate ions as an alternative method for the precipitation of radium. This alternative method was even actually described in document (2).

In this regard the subject-matter of Claim 1 would appear as an obvious modification of the basic process disclosed in document (1), and as such could not be regarded as inventive.

IV. The Appellant (Applicant) lodged an appeal against the decision of the Examining Division on 22 October 1985, paying the prescribed fee and filing the Statement of Grounds on 17 December 1985.

These grounds were generally as follows:

- i) it could not be predicted from document (1) that in a digestion process of phosphate rock wherein no barium compounds are added, a first residue could be obtained that contains economically recoverable amounts of yttrium and lanthanides, and that simultaneously practically radium-free calcium sulphate could be produced as a second residue;
- ii) it is explicitly specified in document (1) that the precipitation of radium is effected in the presence of sulphate ions by addition of barium compounds and that the presence of barium sulphate is necessary to precipitate radium sulphate. Although document (1) states that calcium sulphate precipitate may be used as a filter aid in the removal of the precipitated impurities, this does not suggest at all the mere deletion of barium compounds;
- iii) the method disclosed in document (2) for removing radium from a calcium chloride solution requires a large amount of sulphuric acid as well as the presence of alcohol. This addition of alcohol would certainly be prohibitive, and the use of sulphuric acid in large excess would result in the precipitation of calcium sulphate in the digestion step; a substantial portion of radium would be present in this precipitate which would require a further expensive purification.

V. In a communication the Board indicated that although the deletion of barium compounds, in order to eliminate radium impurities from calcium compounds contained in phosphate

rocks, could be regarded as an inventive feature as such, Claim 1 would appear objectionable since it did not contain all essential features.

- VI. On 18 May 1987 the Appellant filed a new set of 8 claims of which the only independent Claim 1 amended on 26 June 1987 reads as follows:

"Process for the recovery of practically radium-free calcium sulphate, yttrium and lanthanides in a phosphoric acid preparation process by digesting phosphate rock, in the presence of more than 5% by weight of sulphate ions calculated in respect of the quantity of phosphate rock, with an excess of phosphoric acid required for the formation of monocalciumphosphate, separating out the insoluble residue, precipitating calcium sulphate from the remaining solution with sulphuric acid and separating the calcium sulphate from the phosphoric acid solution formed, characterised in that the phosphate is digested with phosphoric acid in the presence of such a quantity of sulphate ions that at least 80% by weight of the radium present in the phosphate rock is precipitated as radium sulphate and at most 20% by weight of the calcium present in the phosphate rock is precipitated as calcium sulphate without barium compounds being added, the residue separated out is treated with an acid in a quantity of 100 to 1000% by weight calculated in respect of the quantity by weight of residue, and yttrium and lanthanides are separated off from the acid liquor formed."

- VII. The Appellant requested the impugned decision to be set aside and the European patent be granted on the basis of Claim 1 filed on 26 June 1987, Claims 2 to 4 and 6 to 8 filed on 18 May 1987 and original Claim 5.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is therefore admissible.
2. There are no formal objections on the basis of Article 123(2) EPC to the current version of the claims since it does not extend beyond the content of the application as filed.

According to Claim 1, the quantity of sulphate ions present in the digestion of phosphate rock with phosphoric acid is such that at least 80% by weight of the radium present in the phosphate rock is precipitated as radium sulphate and at most 20% by weight of the calcium present in the phosphate rock is precipitated as calcium sulphate. This corresponds to the preferred embodiment mentioned on page 3, lines 4 to 10 of the description.

As far as the acid treatment and the separation of yttrium and lanthanides are concerned, they were the subject-matter of original Claim 8. The amount of acid used to treat the residue was originally disclosed on page 3, lines 26 to 28.

Claims 2 to 8 are identical with original Claims 2 to 7 and 9.

3. As acknowledged in the description of the application at page 1, line 25, document (1) discloses a process for the recovery of practically radium-free calcium sulphate in a phosphoric acid preparation process by digesting phosphate rock, in the presence of more than 5% by weight of sulphate ions calculated in respect of the quantity of phosphate rock, with an excess of phosphoric acid required for the formation of monocalcium phosphate, separating out

the insoluble residue, precipitating calcium sulphate from the remaining solution with sulphuric acid, and separating the calcium sulphate from the phosphoric acid solution formed (Claim 1; column 5, lines 5 to 11).

Depending on its origin the raw phosphate does contain (examples 1 and 2) or does not contain (example 3; column 3, lines 55 to 57) a certain amount of radium compounds. These radioactive impurities are eliminated before calcium sulphate is precipitated by addition of barium compounds which act as entrainers for the precipitation of radium (column 4, lines 35 to 45). As far as the level of radioactive impurities contained in the resulting calcium sulphate is concerned, this method is satisfactory since the amount of radium is below the limit of detection of 2pCi/g mentioned in examples 1 and 2; however, it requires the use of 0.1 to 5% by weight of barium per 100 parts of raw phosphate (column 4, lines 51 to 54) which has an objectionable effect on the cost of production of calcium sulphate. The problem underlying the present application can thus be seen in suggesting a more economical process for the preparation of radium-free calcium sulphate.

Among other impurities which in the process described in document (1) precipitate together with calcium are rare earth compounds (column 2, lines 38 to 45). As a second aspect of the application the claimed process aims at the subsequent recuperation of yttrium and rare earths from this precipitate.

The solution to these problems consists in the combination of process features mentioned in Claim 1 which can be summarised as follows:

- i) no barium compound is added to act as entrainer for the radium impurities;
- ii) the amount of sulphate ions present is such that at least 80% by weight of radium and at most 20% by weight of calcium precipitate;
- iii) the residue is treated with an acid to recuperate yttrium and lanthanides.

The Applicant has demonstrated with appropriate examples that this combination of features effectively solves the said problems.

4. Having examined the documents on file, the Board has come to the conclusion that a process comprising all the features of Claim 1 is not disclosed in these citations; the subject-matter of Claim 1 is thus novel. Since novelty has never been questioned by the Examining Division, further considerations in this respect are superfluous.
5. According to document (1) the addition of barium compounds is unambiguously related to the presence of radium compounds in the phosphate rock (Claim 1b and column 2, lines 26 to 28). It is explicitly specified that the precipitation of radium compounds occurs by addition of barium compounds and that barium sulphate and radium sulphate are removed together from the monocalcium phosphate solution by filters or other separating devices (column 4, lines 35 to 45).

That barium sulphate resulting from the addition of barium compounds is necessary to precipitate radium sulphate is further confirmed by the examples. The treatment of raw phosphates according to examples 1 and 2 which contain radium impurities requires the addition of barium

compounds which act as entrainers whereas the process applied to Kola raw phosphate (example 3) which does not contain such radium impurities (column 3, lines 55 to 57) does not involve the addition of barium compounds.

According to a preferred embodiment calcium sulphate precipitate may also be used as a filter aid for the removal of the impurities by suitable control of the added sulphate ions; in this case the quantity of precipitate calcium sulphate should not exceed 20% of the precipitable amount (column 5, lines 5 to 13). This feature corresponds exactly to the condition expressed in the present application (page 3, lines 4 to 10), i.e. the digestion of the phosphate rock is preferably carried out in the presence of such a quantity of sulphate ions that at most 20% by weight of the calcium present in the phosphate rock is precipitated as calcium sulphate.

This raises the question whether the skilled man at the priority date could have been inclined to use the method of calcium sulphate precipitate alone in order to eliminate radium impurities instead of using this method together with the addition of barium compounds.

6. Document (2) describes several methods of separation of radium impurities which do not require the use of barium entrainers. One of them consists of entraining radium by calcium sulphate dihydrate prepared by addition of sulphuric acid to a solution of calcium chloride in the presence of alcohol (page 946).

Although the amount is not specified, the use of alcohol could hardly be compatible with the envisaged economic digestion process of phosphate rock since this would involve prohibitive amounts of this compound. Besides, the efficiency of such a method is closely related to the

excess of sulphuric acid added, since excesses of 100 to 200% are necessary in order to precipitate 85 to 95% of calcium sulphate on which 65 to 92% of radium ions are adsorbed (page 946, paragraph on "Adsorption"). The skilled man trying to interpret the preferred embodiment suggested in general terms in document (1) in the light of the teaching of document (2) would thus be faced with subsequent purification problems as a result of the precipitation of most of the calcium sulphate containing most of the radium sulphate which would occur in the digestion step due to the large excess of sulphuric acid. This shows that the method described in document (2) which is to be used alone to separate radium from calcium has little in common with the additional treatment mentioned in document (1).

7. In reality the features disclosed in document (2) are not compatible at all with the conditions set out in Claim 1. It is explicitly required that the digestion of the phosphate rock should be carried out in the presence of such a quantity of sulphate ions that at least 80% by weight or more of the radium present in the phosphate rock is precipitated as radium sulphate and at most 20% by weight of the calcium present in the phosphate rock is precipitated as calcium sulphate. This last condition of a limited amount of calcium allowed to precipitate, would lead the skilled man away from the teaching of document (2), thus from the possibility of eliminating radium impurities without barium entrainers.
8. The present formulation of Claim 1 includes all the essential process features which are necessary for the actual separation of radium from calcium and the further extraction of yttrium and lanthanides from the residue. Their combination is regarded as inventive and therefore supports the patentability of the entire process.

Although Claim 1 is drafted as a process claim for the subsequent recovery of these elements, this final step is performed by methods which are described as usual in the art (page 3, lines 29 to 32) and whose incorporation in the claim does not appear mandatory.

9. Claims 2 to 8 are concerned with preferred embodiments of the process according to Claim 1. Their patentability is supported by that of Claim 1.

Order

For these reasons, it has been decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division with the order to grant a patent on the basis of following documents:
 - description: pages 1 to 3 filed on 18 May 1987 and
original pages 4 to 21
 - claims: Claim 1 filed on 26 June 1987
Claims 2 to 4 and 6 to 8 filed on 18 May 1987
Claim 5 as originally filed
 - drawings: original drawings 1/3 to 3/3.

The Registrar:



The Chairman:

