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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Cement production from residues from conversion of coal or the like

Klassifikation / Classification / Classement : C04B

ENTSCHEIDUNG / DECISION
vom / of / du 11 July 1985

Anmelder / Applicant / Demandeur : EXXON

~~Patentinhaber / Proprietor of the patent / Titulaire du brevet :~~

~~Einsprechender / Opponent / Opposant :~~

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Art. 52(1), 56

"Inventive step" - "No prejudice"

Leitsatz / Headnote / Sommaire

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Case Number: T 23 / 83

DECISION

of the Technical Board of Appeal 3.3.1

of 11 July 1985

Appellant: EXXON Research and Engineering Company
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Decision under appeal: Decision of Examining Division 028 of the European Patent Office dated 14 September 1982 refusing European patent application No 80 301 609.6 pursuant to Article 97(1) EPC

Composition of the Board:

Chairman: K. Jahn
Member: P. Lançon
Member: P. Ford

Summary of Facts and Submissions

I European patent application No. 80 301 609.6 filed on 16 May 1980 and published on 10 December 1980 under publication No. 20060 claiming the priority of a prior US application of 18 May 1979 (US 41 271) was refused by the decision of the Examining Division 028 of the European Patent Office dated 14 September 1982. The decision was based on Claims 1 to 8 received on 13 February 1981 - claims 1, 3 and 5 reading as follows:

1. A process for the manufacture of cement, which comprises

(a) passing residue solids containing from 20 to 60 weight per cent carbonaceous material with the remainder being inorganic ash constituents including calcium silicates and calcium aluminosilicates, into a cement-making zone; the residue solids being a product of (i) converting a solid carbonaceous feed material in the presence of an alkali metal-containing catalyst into liquids and/or gases thereby producing particles containing carbonaceous material, inorganic ash constituents and alkali metal residues and (ii) treating the particles with a calcium-containing compound, preferably calcium oxide, in the presence of water at a temperature of from 120°C to 370°C to convert water-insoluble alkali metal constituents in the alkali metal residues into water-soluble alkali metal constituents; and

(b) combusting at least some of said carbonaceous material in said residue solids to supply at least a portion of the energy required to convert said residue solids into cement in said cement-making zone, whereby the presence of said calcium silicates and said calcium aluminosilicates decreases the amount of energy required to convert said residue solids into cement.

3. A process according to Claim 1 wherein the carbonaceous content of the residue solids is sufficient to provide substantially all of the energy required to convert the solids into cement.

5. A process according to any one of the preceding claims wherein the residue solids are contacted with water to remove water-soluble alkali metal constituents prior to passing the solids into the cement-making zone.

II The reason given for the refusal was lack of inventive step, as far as Claim 1 was concerned, knowing the content of US-A-4 022 629 (1). The first instance further stated that, because of their dependency upon an unpatentable Claim 1, Claims 2 to 8 were also not patentable.

According to the Examining Division, the concept of burning a coal gasification residue in a cement-making zone is known from document (1). This document would not suggest a prejudice against an ash content of more than 30% in the residue. No inventive step could be seen either in the use of residue with more than 30% of ash content, or in establishing that the use of already combined silicates results in a decrease in the energy required by the process.

III On 5 November 1982, the Appellant lodged an Appeal against the decision of 14 September 1982 and paid the fee within the time limit. The Statement of Grounds was received on 8 January 1983.

With the Statement of Grounds, the Appellant cited 4 new documents which included :
- The Appellant's own German specification DE-A-2 802 998 (2) referred to as the prior art mentioned on page 1 line 9 to page 2 line 11 of the application,

- the book entitled "The Chemistry of Cement and Concrete", F.M. LEA, 1971 (3)
- and "Cement Data Book" - W.H. DUDA - 1977 (4).

In the Statement of Grounds the Appellant contends that:

The teaching of document (1) would establish a prejudice against taking an ash content of more than 30%. He further adds that the residue solids treated with a calcium containing compound of Claim 1 are neither quoted not suggested in (1).
Although document (2) describes the production of such residue solids, this document would not suggest their use for the manufacturing of cement. On the contrary, in view of the newly cited document (for instance (3), page 155), the man skilled in the art would not have considered using residue solids as produced in the process of (2) because they are not devoid of alkali metal components.

IV In his answer dated 17 September 1984 to the Board's communication of 26 March 1984, the Appellant maintained his opinion that the skilled man would not have arrived at the claimed invention even if he had combined the teachings of (1) and (2).

He alleges that the cement obtained would be a good cement (i.e. better than anticipated particularly in view of the relatively high alkaline content), and that the amount of energy required during the cement making process would be reduced.

In addition, the Appellant submitted three auxiliary main Claims, the first main claim being restricted to the subject-matter of Claim 3, the second main claim being

restricted to the subject-matter of Claim 5, and the third main claim being restricted to the subject-matter of Claims 3 and 5.

V The Appellant requests that the decision of the Examining Division be set aside and a European Patent be granted on the basis of the main set of claims or, failing those, on the basis of one of the sets of auxiliary claims.

The Appellant also requests a refund of the Appeal fee.

Reasons for the Decision

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

2. There is no formal objection to any of the current versions of the claims since they are adequately supported by the original disclosure. The broadest form of Claim 1 has been limited with the terms of original Claims 3 and 5 and with the insertion of features of the original description (page 4, lines 12 to 30).

New Claims 3, 4 and 5 have been added. Their content is supported by the description (page 3, lines 28 to 31; page 6, lines 5 to 21; page 20, lines 17 to page 21, line 1).

Claims 2, 6, 7 and 8 have been maintained. Thus, this broadest set of claims can form the basis for a decision.

3. The relevant closest state of the art is described in document (1).

Document (1) describes a continuous process for the manufacture of cement clinker. This process comprises

- introducing unsintered cement raw material comprising calcium carbonate containing material, silica containing material, alumina containing material and optionally Fe_2O_3 providing material into an inclined rotary vessel;
- heating said raw materials at calcination temperatures ranging from 1000°F to 2000°F to effect thermal decomposition of calcium carbonate, and then heating said calcined material at sintering temperatures ranging from 2300°F to 2900°F in a clinkering zone.

It is in this zone that calcined materials are converted into the typical cement compounds: tricalcium silicate, dicalcium silicate, tricalcium aluminate and tetracalcium-alumino-ferrite.

Residual fuels having a volatiles content of not more than 40% and an ash content of less than 30% to provide from 5 to 75% of the heat required for said process are incorporated into the cement raw materials. The expression "residual fuels" means carbonaceous fuels having a desired low volatiles content and high ignition temperature of the non-volatile portion.

Char and coal gasification residues are explicitly quoted as representative residual fuels.

Later on, it is suggested that adjustments in the chemical compositions of the cement making feed material may be desirable to compensate for the incorporation into the clinker of substances such as iron, from the ash of the residual fuel. Immediately thereafter it is admitted that, in order to improve the setting properties of the ultimate cement product, it is known in the art to add a small amount of calcium sulfate to cement clinker, usually during the final grinding (See column 1, lines 51 to 54; line 65 to column 2, line 3; lines 20 to 55, column 4, lines 1 to 35 and Claims 1 and 9).

In addition to the foregoing it must be emphasized that one object of this process was to permit the utilization of residual fuels, including char and coal gasification residues.

4. In view of this particular state of the art, the problem of the present invention can only be seen as providing an alternative to the process known from (1) with the use of other starting materials than those defined in (1).

As a solution to this problem, the Appellant proposes a process for the manufacture of cement generally comprising the features of the process according to (1) whereby the starting materials contain 20 to 60% carbonaceous material with the remainder being inorganic ash constituents including calcium silicates and calcium aluminasilicates. These starting raw materials are, essentially, a product of (i) converting a solid carbonaceous feed material in the presence of an alkali metal-containing catalyst thereby producing particles and (ii) treating the particles with a calcium-containing compound to convert water-insoluble alkali metal constituents in the alkali metal residues into water-soluble alkali metal constituents, (see Claim 1 of the application).

The production of these starting raw materials is, however, described in document (2) which corresponds to the background art indicated on page 1, line 19 to page 2 line 11 of the application, as later recognized by the Appellant (see Statement of Grounds, page 4, 1st paragraph).

5. Document (2), the Appellant's own specification, deals with the problem of recovery of alkali metal constituents from the char particles produced during coal gasification carried out in the presence of an alkali metal-containing catalyst. The chars normally include unconverted carbonaceous constituents and various inorganic constituents generally referred to as

ash. Although the composition of ash varies, the principal constituent, expressed as oxides, are generally silica, alumina and ferric oxide. During the catalytic conversion, at least a portion of the alkali metal compounds that are used as gasification catalyst react with the aluminosilicates and the other ash constituents to form water soluble and water insoluble alkali metal residues. According to document (2), there had been proposals for the recovery of the water soluble alkali metal constituents by water washing, but unless the alkali metal constituents in the insoluble alkali metal residues could be recovered, they are lost for the process. (Page 7, last 4 lines, page 8 and page 9, 7 first lines page 10, 8 last lines and page 11).

The process proposed in document (2) allows recovery of these water insoluble alkali metal compounds - (Page 2, lines 3 to 19). This process comprises treating char particles with a calcium compound in the presence of water at elevated temperature. The calcium ions from the calcium compound evidently react with the water insoluble alkali metal compounds in the char particles thereby liberating alkali metal constituents which dissolve in the water to form a solution which is recycled, and producing water insoluble calcium silicates. These water insoluble containing precipitates may be processed to recover valuable components such as the calcium silicates which may subsequently be used in the manufacture of cement. (Page 9, last paragraph, page 23 and 24, 14 first lines, page 27 last 5 lines, pages 28 and 29, page 30, 10 first lines, page 32, lines 12 to 16).

The tests carried out with potassium compounds indicate a recovery of insoluble potassium ranging from 63 per cent to 95.2 per cent (see page 40). With reference to the same

passages as quoted by the Appellant, it is apparent from the foregoing that the process of document (2) provides a system which makes it possible to significantly increase the amount of alkali metal constituents that are recovered in soluble form (top of page 23, Table I page 40, pages 41 and 42).

6. In its communication dated 26 May 1984, the Board already accepted the novelty of Claim 1, for the following reasons:

Document (1), which discloses the use of coal gasification residues in making cement, specifies a maximum ash content for such a material which lies outside the claimed 40-80% by weight range in the application.

Document (2), which describes the production of the starting material of the claimed process, does not suggest the direct use of that material in the manufacture of cement. Thus, the proposed Claim 1 is novel.

7. As regards the inventive step, the question to be clarified is whether the use of residues such as obtained in the process of document (2) in the process according to document (1) is obvious to the skilled man.

The Appellant alleges that there are two reasons which would dissuade the skilled man from this use:

(a) the ash content (40-80 weight %) in the residues obtained in the process of document (2) is clearly higher than the ash content clearly stated in Claim 1 of document (1) (less than 30%),

(b) the residues obtained in the process of document (2) are not deprived of alkali metal constituents. These metal constituents would be deleterious in a cement-making process.

In addition, the Appellant submits three arguments in order to support an inventive step:

(c) even if the skilled man had combined the teachings of documents (1) and (2), he would have used the char as supplementary fuel and not used it alone as fuel let alone as both fuel and feed component,

(d) it would be surprising to find that at least a portion of the calcium used in the hydrothermal treatment in document (2) was converted to a form common in cement, and that this partial conversion during the hydrothermal treatment reduces the amount of energy required during the cement making process,

(e) with all the expected disadvantages, it would be surprising that the used char would produce a good cement.

8. Considering the first reason (a) relating to the ash content, the Board has already raised, in its communication, the question of the reduction of the need for additional cement forming material when manufacturing cement from char. The Board submitted that the use of a high ash char might be an obvious way.

In this connection a fair comparison of the ash content in document (1), and in Claim 1 of the application is necessary.

Document 1 precribes the use of ashes in an amount of less than 30% in the cement raw materials. However, adjustments in the compositions of the cement may be desirable to compensate for the incorporation of certain substances from the ash of the residual fuels. However, also, a small amount of calcium

sulfate may be added in order to improve the setting properties of the ultimate cement product (See point 3). This suggests clearly that ashes according to document (1) should not include calcium, contrary to what is required according to Claim 1 of the application.

Indeed, Claim 1 of the application specifies that the ash constituents include calcium silicates and calcium aluminates. Balancing the carbonaceous materials, these ash constituents form 40 to 80 weight % of the cement forming materials. Although not necessary, a further support may be found in document (2) which describes the production of the residues used in the claimed process. According to document (2), although the composition of ash varies, the principal constituents are generally silica, alumina and ferric oxide. Thus calcium is not mentioned. However, after treatment according to the process described in document (2), the spent solids may be further processed to recover calcium silicates which may subsequently be used in the manufacture of cement (See point 5).

In conclusion, the composition of the ashes in the present case and in document (1) may differ significantly as far as calcium is concerned. Should account be taken of calcium in considering document (1), then the ash content might come very close to that in Claim 1.

Consequently, in the absence of any convincing counter arguments, the Board maintains the view that the upper limit of the ash content does not constitute a prejudice against the use of the char obtained in the process of document (2) in the process of document (1).

9. In relation to the point concerning the alkali metal content (b), the Appellant has made three citations in support of his argument. All of them show the need for a low alkali metal content where the cement is to be used in concrete with aggregates that may be deleteriously reactive.

For instance, according to document (3), the alkali content of clinkers has been reduced commercially to a value of 0.6 per cent (page 155). According to document (4), the alkali content of Portland cement clinker is between 0 and 1% (Table 1.5.1, page 8). The third citation, ASTM Standard C-150, specifies a maximum of 0,6% alkali.

Turning now to document (2), it has to be examined whether the residues produced in the process described there would not be considered as suitable for cement making.

As already shown in point 5 above, the results of the tests given in document (2) convincingly illustrate the effectiveness of the process for removing alkali described in the document. In view of the same passages in document (2) as quoted by the Appellant in his Statement of Grounds (page 4, 1st paragraph), the Board is convinced that the data and information in document (2) give a clear suggestion to the skilled man that the hitherto unsuitable alkali containing residues have become, after removal of the alkali by the described process, a suitable new starting material for cement making.

In short, the Board considers that the use of the residue produced in the process of document (2) in the process of document (1) was obvious to the skilled man.

10. Further, referring to point (c), the skilled man, aware of the teachings of document (2), knows that the char produced in the process of this document contains all the essential elements present in the starting material of the process of document (1), namely, in addition to unconverted carbonaceous materials, alumina, silica, ferric oxide, calcium compounds and a significantly reduced alkali metal content. (See points 3 and 5). Therefore, he would not have exhibited any inventive ingenuity in using this char as main component (that is fuel as well as cement raw material) in the process of document (1). Thus, this basic allegation of the Appellant fails to convince the Board.

Nothing surprising can be seen in the finding that at least a portion of the calcium used in the hydrothermal treatment (document (2)) was converted into a form common in cement making - calcium silicates - and that consequently the amount of energy required during the cement making process was reduced. As a matter of fact, this conversion had already been explained in document (2) (see point 5, and also page 32, lines 12 to 16 of document (2)). Consequently, reason (d) of the Appellant also fails to convince the Board.

Lastly, coming to point e), no support has been given to the allegation that the Appellant's process produces a cement with unexpected properties. The Appellant asserts that it is surprising that the cement produced is good and much better than anticipated in view of the expected disadvantages. Such an allegation is too indefinite to be convincing and could not be anything else in view of the absence of any prejudice against the use of the char produced according to document (2), as demonstrated above.

It follows that the arguments (c), (d) and (e) given by the Appellant do not support the presence of an inventive step. In conclusion, Claim 1 lacks inventive step.

11. The result arrived at if (2) is taken as the most relevant state of the art instead of (1), and if the technical problem addressed by the present application is taken as that of making sufficient use of the char arising from a process for the catalytic gasification of coal, as suggested by the Appellant in his answer to the Board's communications, would not lead to a more favourable outcome for the Appellant. In view of the composition of the produced char and in view of the common use of such chars (see point 5), the use in the process for the manufacture of cement described in document (2),(see point 5), would have been obvious to the skilled man.

12. The suggested restriction of the main claim in the auxiliary requests on the basis of Claim 3, Claim 5 or even on the basis of Claims 3 and 5 cannot impart inventive merit to an otherwise obvious process.

On the basis of Claim 3, which includes the added feature of the carbonaceous content of the residue solids being sufficient to provide substantially all of the energy required to convert the solids into cement, the first instance already pointed out that the concept of burning a coal gasification residue in a cement making process to supply up to 75% of the energy required by the process was clearly known from (1) (see also 3). The Board also considers that in view of the thermal efficiency of the process in relation to the carbonaceous material content of the residue, such a feature is obvious.

The same result applies to a restriction on the basis of Claim 5, because the feature of water washing alkali metal constituents prior to passing the solids into the cement making zone is already part of the description of document (2) which is recognized by the Appellant as prior art.

Furthermore, in view of the problem defined under point 4, and more particularly in view of the need to use a starting material substantially free from alkali metal constituents, the feature of Claim 5 is an essential feature, and as such, it should have been incorporated in Claim 1. Accordingly, it has already been discussed under point 9. Thus, Claim 5 does not add any true limitation to the process of Claim 1.

In view of the above, the main claims in the three auxiliary requests for relief likewise lack inventive step.

The same applies to the common subsidiary claims 2, 4 and 6 to 8, since they are dependent on the main claims and fall with them.

13. The requested reimbursement of the appeal fee, for which the Appellant has shown no cause, may only be ordered in a case in which the appeal is deemed to be allowable. This requirement is not met in the present case.

Order

For these reasons, it is decided that :

1. The appeal against the decision of the Examining Division of 14 September 1982 is dismissed.

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