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D E C I S I O N
of 16 April 2004

Case Number: T 0065/02 - 3.2.2

Application Number: 95111069.1

Publication Number: 0753588

IPC: C21D 8/12

Language of the proceedings: EN

Title of invention:
Method for producing a grain-oriented electrical steel sheet having a mirror surface and improved core loss

Patentee:
NIPPON STEEL CORPORATION

Opponent:
EBG Gesellschaft für elektromagnetische Werkstoffe mbH

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56

Keyword:
"Novelty - yes, (after amendment)"
Inventive step - yes, (after amendment)"

Decisions cited:
-

Catchword:
-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0065/02 - 3.2.2

D E C I S I O N
of the Technical Board of Appeal 3.2.2
of 16 April 2004

Appellant: (Opponent)
EBG Gesellschaft für elekromagnetische Werkstoffe mbH
Castroper Str. 228
D-44791 Bochum (DE)

Representative:
Simons, Johannes, Dipl.-Ing.
COHAUSZ & FLORACK
Patent- und Rechtsanwälte
Postfach 10 18 30
D-40009 Düsseldorf (DE)

Respondent: (Proprietor of the patent)
NIPPON STEEL CORPORATION
6-3 Otemachi 2-chome
Chiyoda-ku
Tokyo 100 (JP)

Representative:
VOSSIUS & PARTNER
Postfach 86 07 67
D-81634 München (DE)

Decision under appeal:
Decision of the Opposition Division of the European Patent Office posted 2 November 2001 rejecting the opposition filed against European patent No. 0753588 pursuant to Article 102(2) EPC.

Composition of the Board:

Chairman: W. D. Weiß
Members: R. Ries
U. Tronser

Summary of Facts and Submissions

I. European patent No. 0 753 588 was granted on 8 December 1999 on the basis of European patent application No. 95111069.1.

II. The granted patent was opposed by the present appellant on the ground that its subject matter lacked novelty and did not involve an inventive step with respect to the prior art.

Of the pre-published documents relied upon in the opposition proceedings, only the following have been discussed on appeal:

D1: K. Foster, J. M. Jackson: "Effect of anneal coatings and surface conditions on magnetic properties of grain oriented 3% Si-Fe" IEEE Transactions on Magnetics, volume MAG -16, No. 5, September 1980, pages 743 to 745

D2: EP-A-0 488 726

D3: Data sheet of Alcoa Industrial Chemicals Division, Product "Gilox" Normal Soda, 3rd generation calcined aluminas, October 1989

D4: Data sheet of Alcoa Industrial Chemicals Division, Product "CTF Intermediate Soda, 3rd generation calcined aluminas, February 1993

D7: DE-C-2 947 945

D8: JP-A-4120215 and D8a: abstract in English language

III. With its decision posted on 2 November 2001 the Opposition Division held that the patent satisfied the requirements of the EPC and rejected the opposition (Article 102(2) EPC).

IV. On 5 January 2002, the appellant (opponent) lodged an appeal against the decision and paid the prescribed fee on the same day. A statement setting out the Grounds for appeal was submitted on 5 March 2002.

V. In its response to the communication annexed to the summons to attend oral proceedings, the appellant submitted an Affidavit of Mr Alexander J. C. M. de Bonth confirming the public availability of documents D3 and D4.

The respondent (patentee) referred to document

D9: S. Taguchi: "Review of the Recent Development of Electrical Steel Sheet in Japan", Transactions of the ISIJ, volume 17, 1977, pages 605 to 615

The respondent doubted the public availability of documents D3 and D4 and, therefore, requested these documents to be disregarded.

VI. During the oral proceedings which took place on 16 April 2004, the technical teaching of document

D10: US-A-3785882

referred to as technical background in the introductory part of the patent at issue was also considered. At the end of the oral proceedings, the following requests were made:

– the appellant requested that the decision under appeal be set aside and the patent be revoked;

– the respondent requested that the appeal be dismissed and that the patent be maintained in amended form on the basis of claims 1 to 6 submitted at the oral proceedings, the description pages 1 to 9, submitted at the oral proceedings, and the figures as granted.

The independent claims 1 to 8 read as follows:

"1. Process for producing a grain oriented electrical steel sheet having a mirror surface containing 0.8 to 4.8% of Si in the form of a strip which has been subjected to a conventional series of operations including hot rolling with or without annealing, wherein sol-Al of 0.012 to 0.05 wt% is contained in the hot rolled steel sheet, cold rolling once or at least twice with intermediate annealing to obtain a final thickness, decarburizing annealing with or without nitriding treatment, immediately thereafter coating the decarburized steel sheet with an annealing separator mainly containing alumina, and final annealing, the process comprising: satisfying the relationship

$$[A] > 0.2 \times [O]$$

where
[A] is the total concentration of alkali metal impurity in the annealing separator in weight % and

[O] is the amount of oxygen contained in the surface of the steel sheet just prior to the final annealing in g/m^2."

Independent claims 2 to 4 differ from claim 1 by the following wording (in bold letters):

"2. Process for producing a grain oriented electrical steel sheet having a mirror surface containing 0.8 to 4.8% of Si, **0.012 to 0.05% of soluble Al, less than 0.01% N,** in the form of a strip which has been subjected to a conventional series of operations including hot rolling with or without annealing, wherein sol-Al of 0.012 to 0.05 wt% is contained in the hot rolled steel sheet, cold rolling once or at least twice with intermediate annealing to obtain a final thickness, decarburizing annealing with nitriding treatment, immediately thereafter coating the decarburized steel sheet with an annealing separator mainly containing alumina, and final annealing, the process comprising: satisfying the relationship

$$[A] > 0.2 \times [O]$$

where
[A] is the total concentration of alkali metal impurity in the annealing separator in weight % and
[O] is the amount of oxygen contained in the surface of the steel sheet just prior to the final annealing in g/m^2."

"3. Process for producing a grain oriented steel electrical sheet having a mirror surface containing 0.8 to 4.8% of Si, **0.012 to 0.05% of soluble Al, less than 0.01% N, 0.02 to 0.3% of Mn, and 0.005 to 0.040% S**, in the form of a strip which has been subjected to a

conventional series of operations including hot rolling with or without annealing, wherein sol-Al of 0.012 to 0.05 wt% is contained in the hot rolled steel sheet, cold rolling once or at least twice with intermediate annealing to obtain a final thickness, decarburizing annealing, immediately thereafter coating the decarburized steel sheet with an annealing separator mainly containing alumina, and final annealing, the process comprising: satisfying the relationship

$$[A] > 0.2 \times [O]$$

where

[A] is the total concentration of alkali metal impurity in the annealing separator in weight % and

[O] is the amount of oxygen contained in the surface of the steel sheet just prior to the final annealing in g/m^2."

"4. Process for producing a grain oriented electrical steel sheet having a mirror surface containing 0.8 to 4.8% of Si, **0.02 to 0.3% of Mn, less than 0.01% N,** in the form of a strip which has been subjected to a conventional series of operations including hot rolling with or without annealing, wherein sol-Al of 0.012 to 0.05 wt% is contained in the hot rolled steel sheet, cold rolling once or at least twice with intermediate annealing to obtain a final thickness, decarburizing annealing, immediately thereafter coating the decarburized steel sheet with an annealing separator mainly containing alumina, and final annealing, the process comprising: satisfying the relationship

$$[A] > 0.2 \times [O]$$

where

[A] is the total concentration of alkali metal impurity in the annealing separator in weight % and

[O] is the amount of oxygen contained in the surface of the steel sheet just prior to the final annealing in g/m^2."

VII. The appellant argued as follows:

Document D1 which represents the closest prior art discloses a process for producing regular grain oriented (RGO) and high permeability grain oriented (HGO) Si-steel sheet having reduced induction losses after decarburizing and final annealing the cold rolled sheet. It is found that using a non-reactive alumina (Al_2O_3) annealing separator coating results in a smooth (= mirror-like) oxide free surface and prevents the formation of subsurface oxides observed after conventionally annealing the sheet with MgO. These subsurface oxides are believed to adversely affect the electrical and magnetic properties, a finding that fully complies with the explanations given in the patent at issue.

Although document D1 distinguishes between normally processed RGO steels sheet and "fully processed" RGO and HGO Si steel sheet, whereby after a stress relief annealing step the coating is removed by etching and thereafter the sheet is re-annealed with an Al_2O_3 coating, a skilled person would learn from document D1 as a whole that an alumina annealing separator exhibits a beneficial influence on the surface condition and the electric and magnetic properties of both RGO and HGO Si steel sheet. It is true that document D1 does not specify the purity of the alumina used in the final annealing. However, the composition of the alumina separator typically applied in the art and used also by

the opponent for recrystallisation annealing Si-steel sheet is specified in documents D3 and D4. These data sheets disclose a 99% high purity alumina comprising as a main impurity about 0.5% Na (D3) or max. 0.5% Na (D4), which represents the typical impurity level mentioned also in paragraphs [0020] and [0048] of the patent.

The typical amount of oxygen [O] after decarburisation annealing, also undisclosed in D1, can be estimated from documents D2 and D8a. It normally ranges from 0.4 to 1.6 g/m^2, a typical value being about 1.0 g/m^2 for both surfaces = 0.5 g/m^2 for one surface (cf. D2, page 6, lines 47 to 49 and page 4, lines 18 to 20; D8a: < 0.5 g/m^2). This level corresponds to the values for [O] of the examples given in the patent. The amount of Na as an impurity, therefore, suffices to satisfy the formula [%Na] > 0.2 x [O] specified in the patent. Having regard to the fact that the grain oriented Si steel sheet produced according to D1 has the same final properties, i.e. (a) exhibits a mirror-like surface and (b) is free of sub-surface oxides, it has to be duly assumed that the same process conditions as in the patent have been applied: the use of alumina having a purity of about 99% and comprising about 0.5% Na, and a steel sheet having conventional amounts of [O] in g/m^2 after decarburization. Thus even if the subject matter of independent claims 1 to 4 of the disputed patent was rated as being novel with respect to the teaching of document D1, which is denied, the claimed process does not involve an inventive step since it amounts to nothing more that what is conventionally carried out by a person skilled in the art. This statement is confirmed by document D7 showing that the positive

effect of minor amounts of Na in a non-hydratable annealing separator mainly composed of MgO rather than Al_2O_3 upon the watt loss is also known. Hence, the independent claims 1 to 4 do not comprise patentable matter.

VIII. The respondent (patentee) argued as follows:

Document D1 discloses neither the addition of alkali metals Na, Li, K to the alumina separator nor the interrelationship of [%(Na, K, Li)] > 0.2 x [O] claimed in the patent. As set out in claims 1 to 4, the Si steel sheet comprises 0.012 to 0.05 soluble Al (normally needed in HGO Si steel sheet when (Al,Si)N is the main grain inhibitor). If such a sheet is decarburized, oxides mainly composed of mullite ($3Al_2O_3 \cdot 2SiO_2$) are precipitated directly under the surface and a mirror-like smooth surface of the sheet cannot be obtained in the final anneal. Conventionally, these oxides and precipitates need to be removed by etching or acid pickling. It is the merit of the applicant who has found for the first time that small amounts of alkali metals, either comprised as residual impurities in or intentionally added to a high temperature coil Al_2O_3 annealing separator, effectively counteract the formation of subsurface mullite. Neither the authors of document D1 nor the inventors of document D7 have recognized the strong inhibiting effect of alkali metals and the dependency of Na, K, Li on [O] expressed by the claimed formula. Moreover, the appellant failed to prove beyond any doubt that the types of alumina disclosed in the data sheets D3 and D4 actually have been applied as an annealing separator coating. It is, therefore, unknown which type of

alumina was used as an annealing separator in document D1.

Contrary to the claimed Si steel sheet, the RGO Si steel sheet tested in document D1 did not comprise soluble Al and, therefore, the problems associated with the formation of subsurface mullite did not arise. Even if soluble Al was initially present, as is the case in HGO Si steel sheet also tested in D1, this aluminium was oxidized during the decarburisation and stress relief annealing steps to form an oxide film which then was removed by acid pickling or etching before coating the sheet with the alumina separator and final annealing. Contrary to this "fully processed sheet" mentioned in document D1, acid pickling is dispensed with in the claimed process due to the absence of subsurface mullite. Thus, the claimed process not only enables the production of grain oriented Si steel sheet having the core loss effectively reduced by imparting it with a mirror-like surface free of subsurface precipitates, but it also provides a simplified and less expensive treatment by eliminating the acid pickling step. The subject matter of independent claims 1 to 4 is therefore novel and involves an inventive step.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

Claim 1 results from a combination of claims 1 and 6 as granted with the description paragraph [0037] specifying the presence of 0.012 to 0.05% acid soluble aluminium in the steel. The further restricting feature of "immediately thereafter coating the decarburized steel sheet" in amended claim 1 finds support in the description paragraphs [0042, 0043].

The same statement is true for independent claims 2 to 4 which are amended correspondingly. Dependent claims 5 and 6 remain unchanged and correspond to claims 5 and 7 as granted.

The description has been suitably adapted to the revised wording of the claims, and parts no longer falling within the scope of the claims have been deleted.

The requirements of Article 123(2) and (3) EPC are, therefore, satisfied.

3. *Admissibility of documents D3 and D4*

The public availability of documents D3 and D4 has been challenged by the patentee.

Enclosed with its letter received 11 March 2004, the appellant submitted an Affidavit of Alexander J. C. M. de Bonth, a employee of Alcoa World Chemical, who confirmed the publication date for data sheets D3 (February 1993) and D4 (October 1989). The appellant did not produce any evidence putting Mr de Bonth’s

statement in doubt. Hence, documents D3 and D4 belong to the state of the art.

4. *Novelty*

Lack of novelty has been objected to only in view of document D1 which discloses a HGO Si steel sheet having a smooth mirror-like surface after final annealing with a non-reactive alumina separator coating.

However, document D1 remains silent about the type of alumina used in the process, the precise percentage of soluble aluminium [Al_{sol}] comprised in the HGO steel sheet and the oxygen content per surface after decarburization annealing. Consequently, there is no proof that the formula [A] > 0.2 x [O] set out in independent claims 1 to 4 is definitively met. Moreover, the fully processed HGO steel sheet is, after decarburisation, subject to etching or acid pickling in order to remove an oxide film formed on its surface, a treatment which is excluded from the claimed process.

Although document D10 discloses the use of a high purity 99% alumina annealing separator comprising only (unspecified) trace amounts of soda, silica and iron oxide, the soluble aluminium in the silicon iron composition is restricted to 0.009% at most (cf. D10, column 3, lines 11 to 13; lines 51 to 55).

In the processes disclosed in documents D2, D7, D8 and D9, a conventional annealing separator mainly containing MgO is used (cf. D2, page 4, lines 33, page 8. line 40, page 9. line 4, page 11, line 14; D7, claim 1; D8, page 77, right hand column, line 4,

page 80, right hand column, line 19, page 82, right hand column, line 6, page 85, right hand column, line 4; D9: page 604, right hand column, lines 17 to 22; page 606, right hand column, lines 10 to 18).

Therefore the subject matter of independent claims 1 to 4 is novel.

5. *Inventive step*

5.1 The patent in suit, as set out in the independent claims 1 to 4, relates to a process for producing grain oriented electrical steel sheet which comprises 0.012 to 0.05% soluble aluminium. This amount of aluminium (Al_{sol}) is necessary to combine with nitrogen so as to provide (Al,Si)N as the main grain inhibitor for obtaining a high magnetic flux density (see the patent, paragraph [0037]). If, however, the Al_{sol}-containing steel sheet is decarburized, subsurface oxides are precipitated and a mirror finish is not obtained. Therefore, the oxide film needs to be removed by etching or pickling prior to final recrystallisation annealing the steel sheet with an alumina separator. This process and product are described in document D1 as "fully processed HGO high permeability grain oriented steel sheet". Based on these considerations, it has been common ground for all parties and for the Board that document D1 represents the closest prior art.

5.2 Starting from this prior art, the problem underlying the patent at issue resides in providing a simplified and less costly process which effectively prevents the formation of subsurface oxide in the Al_{sol}-containing steel sheet during the decarburising treatment to

provide a mirror-like surface so that the acid pickling treatment can be eliminated (see paragraph [0013] of the patent specification).

5.3 The solution to this problem consists in applying in the final annealing step a separator consisting of alumina and alkali metals in an amount sufficient to satisfy the formula [A]>0.2x[0]. In doing so, the formation of oxides mainly composed of mullite ($3Al_2O_3 \cdot 2SiO_2$ which cannot be reduced when finally annealing the steel sheet in the high temperature reducing atmosphere) is avoided and a mirror surface is obtained.

5.4 The appellant has referred to document D1, page 743, abstract and the introduction, page 744, right column, first paragraph, arguing that by annealing with a non-reactive alumina separator smooth surfaces for RGO and HGO Si steel sheet have been obtained. It follows from this that the claimed correlation rule [A]>0.2[0] is implicitly satisfied.

It is, however, noted that in D1 only regular grain oriented RGO steel (not comprising Al_{sol} in the claimed amounts) has been processed without intermediate removal of the oxide coating. By contrast, HGO steel sheet (comprising Al_{sol}) has been "fully processed" meaning that the decarburised steel sheet is stress relief annealed (SRA) and, after removing the oxide coating formed on the surface by etching, is finally annealed with an alumina separator (see Tables II and III), and a smooth surface is obtained. It remains, however, unknown from document D1, whether or not a high purity 99% Al_2O_3 has been chosen (as for instance

mentioned in document D10, column 3, lines 51 to 55) and if so, which amounts of alkali metals it actually comprised as an impurity.

The appellant has pointed in this context to the type of alumina specified in D3 and D4 typically comprising up to 0.5% Na and alleged that such a material is commonly used in the processes in the art, including the process disclosed in D1, as an annealing separator. However, no convincing evidence has been produced by the appellant to prove this allegation "up to the hilt". The type of alumina used as an annealing separator in D1, therefore, remains speculative.

It remains also undisclosed in D1 which amount of oxygen on each steel surface was present after decarburisation, stress relief annealing and etching. The appellant pointed in the context to document D2, disclosing a "marked oxygen content" on both surfaces of the sheet after decarburisation in the range of 0.4 to 1.6 g/m^2, and to document D8 disclosing $<0.5\ g/m^2$. There is, however, nothing in these documents for concluding or implying that these values are to be rated as being typical in the art. Given this situation, it remains speculative whether the claimed relationship $[Na,K,Li] > 0.2 \cdot [0]$ is actually satisfied by the process given in document D1.

5.5 Furthermore, it is observed that none of the prior art documents not even remotely envisaged the addition of small amounts of alkali metal Na, K, Li (albeit as an impurity or by adding it on purpose) to the Al_2O_3 annealing separator and to correlate this constituent to the oxygen content of the decarburized sheet in

order to minimize or completely suppress the formation of subsurface oxides, as does the claimed patent, so that the pickling step can be dispensed with. Only in the process disclosed in document D7, a separator mainly composed of non-hydratable MgO and including Na-borate up to 0.2% Na is proposed (cf. D7, claims 4 and 6, column 8, line 58 to column 9, line 15). The natrium compound and MgO is found to react and form a glass film which provides a high tension to the sheet surface whereby the watt losses of the Si steel sheet is reduced effectively. However, no information whatsoever is given in document D7 leading a skilled person to replace non-hydratable MgO with Al_2O_3 and to control the amount of Na, K or Li with respect to the oxygen content on each surface of the decarburized steel sheet in order to suppress the formation of subsurface oxides and to establish a mirror-like surface. Hence, also the combined teaching of documents D1 and D7 would not make the claimed process obvious.

6. In view of these considerations, the subject matter of independent claims 1 to 4 involves an inventive step. The dependent claims 5 and 6 relate to preferred embodiments of the process set out in claims 1 to 4 and are, therefore, likewise allowable.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to maintain the patent in amended form on the basis of:

   – claims 1 to 6 and the description pages 1 to 9 submitted at the oral proceedings, and

   – the figures as granted.

The Registrar: The Chairman:

V. Commare W. D. Weiß