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File Number: T 212/90 - 3.2.2
Application No.: 82 301 404.8
Publication No.: 0 067 501
Title of invention: Alloy suitable for use in a radioactive radiation
environment and a reactor core component formed therefrom
Classification: C22C 38/40, C22C 38/44

D E C I S I O N
of 10 December 1992

Applicant: Hitachi Ltd.
Opponent: Creusot-Loire Industrie
United Kingdom Atomic Energy Authority
Siemens AG
Headword: Radiation resistant austenitic steel
EPC Article 54(2), 56
Keyword: "Novelty and inventive step (yes) - after amendment"

Catchwords

The disclosure of a standard steel specification is not only defined by its chemical composition, but other requirements like the mechanical properties also belong to the definition (see Point 4 of the reasons).



Case Number : T 212/90 - 3.2.2

D E C I S I O N
of the Technical Board of Appeal 3.2.2
of 10 December 1992

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Representative :

Decision under appeal : Decision of the Opposition Division 2.1.06.017 of
the European Patent Office of 7 November 1989,
dispatched on 17 January 1990, revoking European
patent No. 0 067 501 pursuant to Article 102(1)
EPC.

Composition of the Board :

Chairman : G.S.A. Szabo
Members : W.D. Weiß
M.K.S. Aúz Castro

Summary of Facts and Submissions

- I. European patent No. 0 067 501 was granted with effect from 6 August 1986 on the basis of European patent application 82 301 404.8, filed on 18 March 1982. Claim 1 of the granted patent reads as follows:

"The use, in an environment exposed to neutron-containing radiation, of a Ni-Cr austenite stainless steel wherein said neutron-containing radiation has an intensity of at least 10^{20} nvt and the steel contains not more than 0.03 wt.% C and at least 0.06 wt.% N, the total amount of carbon and nitrogen being at least 0.09 wt.%."

- II. Three oppositions were filed against the patent mainly on the grounds of lack of novelty and inventive step (Article 100(a) EPC). Numerous documents were cited in this respect.

Moreover, there has been an objection that the patent does not disclose the invention in a manner sufficiently clear and complete for it to be carried out (Article 100(b) EPC), because the energy of the neutrons to which the steel alloy is exposed during its intended use, is not specified.

- III. On 7 November 1989, the Opposition Division held oral proceedings, at the end of which the patent was revoked. The written grounds for this decision were posted on 17 January 1990.

The claim under consideration before the Opposition Division was amended insofar as the words "not more than 0.03 wt% C and at least 0.06 wt% N" were substituted by the words "0.003 to 0.01 wt% C and up to 0.15 wt% N".

The Opposition Division held that the subject-matter of the amended Claim 1 lacked an inventive step having regard to the documents:

OI(3) Revue Matériaux et Techniques - No. 6, June 1977, pages 3 to 10; Rabbe, Ferriol and Catelin: "Tôles épaisses en acier inoxydable pour réacteur nucléaire à eau légère";

OIII(1) P. Rabbe and J. Heriter: "Development of Austenitic Stainless Steels with controlled Residual Nitrogen Content; Application to Nuclear Industry ASTM STP 679 American Society for testing and Materials (1979), pages 124 to 143;

OII(3) American National Standard - ANSI/ASTM A 240-80; and

OII(6) Matériaux et Techniques Sept/Oct. 1977, pages 477 to 491.

One major argument in the reasoning of the Opposition Division was that the alloy composition used according to Claim 1 represented an obvious selection from the alloy specifications AISI 304LN and 316LN disclosed in OIII(1) in combination with OII(3), since no unexpected effect was achieved.

IV. On 16 March 1990, the Appellant (Proprietor) filed an appeal against this decision and paid the appeal fee on the same date. The Statement of Grounds was filed by telecopy on 21 May 1990.

V. In a communication dated 7 August 1992 the Board drew the attention to the fact that maximum values for the carbon content of a steel which were indicated in a standard

specification could not necessarily be interpreted in their formal mathematical sense but were meant as a directive for the steel manufacturer to control the process as to aim at a carbon content somewhat below this maximum value, which, in view of the possible fluctuations typical for the process used, warranted that the analysis of the final product lay somewhere in the range below the maximum. Moreover, it had to be considered that a steel was not only defined by its alloy composition but also by the mechanical test requirements which belonged to the respective standard specification as well.

The Board, in this communication, cited as a new document

(B1) Dr. Karl Kummerer, "Werkstoffe der Kerntechnik",
G. Braun Verlag Karlsruhe, 1980, pages 78 to 95,

and stated that the content of this document were in congruence with the detailed analysis, given by Appellant in its letter of 6 June 1991, of the (crack) corrosion and other damage problems arising when austenitic steel is used as a construction material in the core environment of a nuclear reactor.

VI. The Appellant's arguments which were completed during the oral proceedings held on 10 December 1992, can be summarised as follows:

The documents on which the decision of the Opposition Division was based only dealt with the "normal" intergranular stress cracking corrosion (IGSCC) which resulted from the depletion of chromium in the intergranular region in the steel being characteristic of a water environment containing certain impurities at high temperature. In contrast thereto, the patent in suit was

concerned with the additional problems of the irradiation assisted stress cracking corrosion (IASCC) and of the radiation-induced embrittlement (EP-B-0 067 501, column 1, lines 22 to 26, and column 2, line 44, to column 3, line 11). Documents OIII(1), OI(1) and OI(3), on which the decision under appeal was mainly based, did not suggest solutions to these neutron radiation induced problems.

~~The fact that in document OII(3), defining the standards for the AISI 304L and 316L, the carbon content is given as "0,030 wt.% maximum" could not be interpreted as meaning that "any value from zero up to 0,030 wt%" was made available to the public. On the contrary, the combination of the nitrogen and carbon contents indicated in Claim 1 had constituted, before the priority date of the patent in suit, an unmapped area.~~

Comparative tests, the results of which were reported in annexes to the letters of 21 May 1990 and of 6 June 1991, proved that an effect existed in this area which could not possibly have been extrapolated from the prior knowledge about the standard steels AISI 304, 316, 304L, 316L, 304LN, and 316LN.

VII. At the oral proceedings of 10 December 1992 before the Board, beside the Appellant, only Respondent I (Opponent I) was represented. In its written and oral submissions, the Respondent relied, in addition to the documents on which the decision under appeal was based, also on the following documents already submitted in the opposition proceedings:

OI(1) Matériaux et Techniques, Sept.-Oct. 1977,
pages 561 to 578;

OI(2) Matériaux et Techniques, Numéro spécial, Oct. 1977, pages 14 to 22;

OII(14) American Society for Testing and Materials STP 683, published 1979.

Its arguments can be summarised as follows:

The compositions of the steels, the use of which being claimed in the patent in suit, had already been made available to the public by the cited documents. It could be seen from two documents, which were submitted during the oral proceedings, that methods had existed which allowed to produce such extremely low carbon steels on an industrial scale and that such alloys had been produced. Moreover, the effects and the properties of these steels on which the claimed use was based were predictable from results of the research disclosed in the documents OI(1), OI(3), OI(7) and OIII(1). Consequently, the subject-matter of Claim 1 did not meet the requirements of a selection invention according to the decision T 279/89 (unpublished).

The validity of the test reported by the Appellant during the appeal proceedings was contested, because essential conditions of these experiments (e.g. the neutron energy) had not been given and the effects could well be due to a measurement error.

The Respondent III (Opponent III), in its letter dated 19 September 1990, argued that, due to the broad and general formulation of the conditions for the claimed use, Claim 1 did not state anything which would not have been expected by the normal practitioner in this field.

No substantial comment was received from Respondent II (Opponent II).

VIII. The Appellant requests that the decision under appeal be set aside and that the patent be maintained on the basis of Claims 1 to 6 filed together with the description during the oral proceedings of 10 December 1992, as well as of all the figures as granted.

Claim 1 corresponds to the version under consideration before the Opposition Division.

The Respondents I and III request that the appeal be dismissed.

No request has been received from Respondent II.

Reasons for the Decision

1. The appeal is admissible.
2. Amendments

Claim 1 is based on the original Claim 1 and the original description, page 3, paragraph 4, and page 6, paragraphs 1 and 2. The lower limit for the nitrogen content results automatically from the condition that the total amount of carbon and nitrogen is at least 0.09 wt.%. Moreover, the scope of protection of the amended Claim 1 is restricted when compared to its granted predecessor.

Claims 2 to 6 are based on the original Claims 6, 7, 8, 11, and 9, respectively.

There is, therefore, no objection to the current claims under Article 123 EPC.

3. Sufficiency of the disclosure

The disclosure of the patent specification (i.e. column 1, lines 19 to 45) states that the material, the use of which is claimed, should have a high resistance against typically occurring neutron radiations in the core environments of light-water reactors, fast breeder reactors, as well as fusion reactors. Applications which are typical for this use are enumerated as control rods, neutron counter tubes, core supporters, core shrouds, neutron source pipes, wrapper tubes etc. (EP-B-0 067 501, column 4, second paragraph, and column 6, lines 5 to 9). Therefore it is clear from the description that the dose level of 10^{20} nvt is valid for the whole energy spectrum of neutrons, from thermal neutrons up to the level of 0.1 MeV and more.

This dose level has not been chosen arbitrarily, because the expert in this field has known that it is beyond this threshold where the various radiation induced effects start to negatively influence the mechanical properties of austenitic steels (see for instance document (B1), page 93, Figure 35, and page 94, Figure 36). On the other hand doses above this level are typical for the core environment of the various types of reactors mentioned above (cf. document (B1), page 92, Table 35).

Since the European patent discloses the invention in a manner clear and complete for it to be carried out by a person skilled in the art, the provisions of Article 100(b) or of Article 83 EPC, respectively, are therefore met.

4. State of the art and novelty

Document OI(3) reports about researches which aimed at the development of stainless steels which could stand the stress which is typical for support structures in the core of a PWR reactor (cf. OI(3), page 3). This intended use falls under the scope of the use of Claim 1 (cf. EP-B-0 067 501, right column, line 12). Austenitic steels according to the United States standard specifications AISI 304LN and 316LN are denominated as prospective materials for this use.

The content of document OII(3), which is the respective US standard specification, has to be considered as being incorporated into the content of document OI(3) due to this cross-reference. Table I on page F-13 of document OII(3) which defines the chemical compositions of austenitic steels also contains such data for the types 304LN and 316LN. According to the explanation on page G-1 of the footnote "A" which is affixed to the title of Table I, a numerical value indicated in this table means "Maximum, unless range or minimum is indicated. Having this in mind, the austenitic steel types 304LN are both specified as having a "0.10% to 0.16% nitrogen" and a "maximum of 0.030% C".

There is no doubt about how the range for the nitrogen content has to be interpreted. The nitrogen content of the two standard steels broadly overlaps with the claimed range.

It would, however, be against the generally known rules of application of standard specifications, if the indication "maximum of 0.030% C" were merely mathematically interpreted as meaning "0 to 0.030% C". If it were so, it would not have been possible to define the steels 304L and

316L, the chemical compositions of which are also specified in Table I of document OII(3), as standards independent of the respective standards 304 and 316 from which they only differ by a lower maximum for the carbon content. The characteristics of a standard steel are, however, not only defined by the "Chemical Requirements Composition" (cf. Table I of document OII(3)), but also by the "Mechanical Test Requirements" (cf. Table II on page G-2 of document OII(3)). There is, for instance, stated that the steel type 304 must have a minimum yield strength of 205 MPa. Since carbon acts as a strengthening element in this type of steel, this minimum value can no longer be warranted, whenever the carbon content is lowered to a value of 0.030% or less which is valid for the steel type 304L with a minimum yield strength of only 170 MPa. The minimum yield strength of the steel type 304LN is readjusted to 205 MPa because the addition of nitrogen has a strengthening effect.

In view of the preconditions demonstrated above, the standard for the steel type 304LN is, in addition, not to be considered to cover any carbon content below the above maximum value of 0.030%, because an excessive decrease of the carbon content would result in a failure to comply with limits for "Mechanical Test Requirements". The steelworker who receives an order under the condition of a standard specification which contains a maximum value for an unavoidable impurity, like carbon in steel, knows that, as a general rule, he has to aim at 70% of the respective maximum value. This precaution warrants that, taking into account the possible fluctuations of the manufacturing as well as the analysis processes, the actual composition of the final product only lies safely within this maximum but not in the range below the value where important mechanical safety properties may be lost. This means that the steelworker will aim only at a value

about 0.02%, when a maximum of 0.030% is indicated in the standard, but not at 0.01% or even less.

The validity of this general rule is confirmed by the state of the art cited by the Respondents. Document OI(4) (page 132, second paragraph) states that a good-quality AISI 304L steel has a carbon content of 0.020%. Document OI(7) (in the discussion on page 490) discloses that, for an AISI 316L steel with a maximum of 0.030%, a carbon content of 0.024% is aimed at. Figure 12 of document OI(1), where tests on various steels corresponding to AISI 304/316 (C_{\max} 0.08%) down to the low carbon versions 304/316L (C_{\max} 0.03%) are reported, does not display values below about 0.017% carbon.

In summary, standard specifications for the steel types AISI 304LN and 316LN the use of which is suggested in document OI(3)/OII(3) do not disclose a carbon content in the range of 0.003 to 0.01%. The same applies to the documents OI(1), OI(2), OI(4) and OII(6).

Document OII(14), on page 369, mentions the composition of an austenitic steel which, starting from a AISI 316 basic composition, was modified down to very low contents of certain residual elements including carbon and nitrogen. Considering the fact that the composition there is given in atomic percent, the alloy contains about 0.0012 wt% carbon and about 0.006 wt% nitrogen. Thus, both contents are far below the ranges according to Claim 1.

This document cannot serve to support the allegation that the standard specification AISI 316 is meant to disclose carbon contents down to zero percent either, because this known alloy was, starting from a standard alloy, deliberately constructed for experimental purposes to study the cross section for thermal neutrons of nickel and

to exclude the influence of disturbing effects. Because this modified alloy, due to its low carbon and nitrogen contents, does not meet the mechanical minimum requirements, it is not an AISI 316 kind of standard alloy.

The Respondent I has during the oral proceedings of 10 December 1992 submitted copies of two new documents as a further proof for the lack of novelty or inventive step, respectively. One of these documents, according to its title page, originates from a presentation at the "25^e Colloque de Métallurgie de Saclay" from 23 to 25 June 1982 and is obviously no state of the art according to Article 54(2) EPC. The second document consists of a title page bearing the date of "October 1980" and two annexed sheets. One of these sheets was said by the Respondent to originate from page 4 of the final research report mentioned on the title page. The second sheet is a copy of page F-13 of document OII(3). The compositions of four test samples of an austenitic steel are specified on the first sheet, none of which complying with the combination of the nitrogen and carbon contents according to Claim 1. No intended use is disclosed. Consequently, this document, even if it turned out to be prepublished, would be more remote to the subject-matter of Claim 1 than the documents discussed above.

The Board has also examined the other documents which were cited during the opposition procedure but were not mentioned during the appeal proceedings and are, therefore, not specified above. None of these documents discloses the composition of an austenitic steel the use of which is claimed by the patent in suit.

Since, the composition the use of which is claimed by the patent in suit, is outside the alloy definition which has

been a state of the art, the question of the novelty of a selection according to the decision T 279/89 cited by the Respondent does not arise.

Consequently, the subject-matter of Claim 1 is novel.

5. Closest art and difference

The Board concurs with the Opposition Division and the parties that document OIII(1) is the closest prior art and that document OI(3), originating from the same research group, is equivalent in this respect.

Document OIII(1) reports about the development of optimised grades of austenitic steels for parts which are in direct contact with the primary and secondary water in light water reactors (pages 124 to 127, first paragraph). In particular in pressurised water reactors (PWR), these parts undergo severe loadings under high pressure and at elevated temperatures. The three major types of damages which are considered in this publication are enumerated

1. Intergranular stress cracking corrosion (IGSCC),
2. damage from fabrication, and
3. damage from fatigue corrosion.

In particular from the chapter "Method for Determination of the Intergranular Corrosion Resistance" beginning on page 127 it becomes evident that document OIII(1) considers the normal IGSCC only which is due to a chromium depletion by carbide formation at the grain boundaries. IGSCC is not specific for the environment in a light water reactor but occurs, whenever an austenitic steel is subjected to static and/or cyclic stress in pressurised water which is loaded with certain impurities such as oxygen or halides (for general background in this respect

see document OI(1), pages 563 and 564). The content of some of these impurities is, however, enhanced in the primary water of a reactor, because they typically result from decay reactions. The other two types of damages are not exclusive to pressurised water circuits of nuclear reactors either. Their consequences there are more serious, due to the reduced accessibility of the parts in the computer core.

The essence of document OIII(1) (pages 127 to 133) is to suggest two optimised steels called ICL 473 BC and ICL 167 CN the compositions of which lie within the ranges of the AISI standards 304L and 316L, respectively. The fact that the carbon content (average about 0.02%) of these optimised steels is precisely balanced with the chromium and molybdenum contents warrants that these steels combine the good intergranular corrosion resistance of AISI 304L or 316L grade steels with the mechanical property specifications of the higher carbon types AISI 304 and 316 grades, respectively. These results are achieved, although the nitrogen content of these optimised steel types is set in the range of as low as 0.05 to 0.08%.

Although these optimised steels require extreme care during their production which involves higher costs, their use has been preferred to the selection of steels belonging to the families of types AISI 304LN and 316LN with increased nitrogen contents of 0.1 to 0.15% (see Standard Specification OII(3), page F-13). This is because such selection would have "entailed the use of a new material which is not yet retained by current codes, and of which industrial experience is limited" (page 125, paragraph 4 from the bottom).

Document OI(3) is essentially congruent with the teaching of document OIII(1). This document is particularly

concerned with the development of an austenitic steel for core support structures (page 3, paragraphs 3 and 4). Here again, it is preferred to optimise a type 304L/316L steel than to select steels of the types 304LN or 316LN, respectively. The same applies to document OI(2) (page 20, Table 14) and to document OI(1) (pages 570 to 577).

Consequently, the subject-matter of Claim 1 differs from the closest prior art as represented by the documents OIII(1) or OI(3) in that the nitrogen content is higher and the carbon content is lower than in the known optimised steels, and the carbon content is even lower than in the type 304LN or 316LN steels, i.e. 0.003 to 0.01 wt%.

6. Problem and solution

Austenitic steel which is subjected to a high irradiation dose of neutrons may, depending on the energy of the neutrons, suffer from a variety of damages in addition to IGSCC mentioned above (EP-B-0 067 501, column 1, lines 19 to 45). These effects are also discussed in document (B1) (pages 91 to 95, chapter 34.) and they are specified as

- "low temperature embrittlement which occurs at temperatures below 500 °C and is due to dislocations provoked by fast neutrons;
- "high temperature embrittlement" which is the consequence of (n,α)-reactions which are provoked by thermal and by fast electrons; and
- "swelling" which is the consequence of voids provoked by fast neutrons (E > 0.1 MeV).

Depending on which energy level is dominant in a specific reactor type, one of these damage types is prevailing when an austenitic steel is used as a structural material in a

respective reactor core. A dose on the level of about 10^{20} nvt is the threshold for the occurrence of most of these effects (e.g. document (B1), page 93, second paragraph, page 94, Figure 36, page 95, Figure 37).

Starting from the closest prior art described above, the basic technical problem of the patent in suit is to create an austenitic steel material which has a considerable long time resistance against all these irradiation induced types of damage and which, therefore, can be used as a universal core material in light-water reactors, fast breeder reactors, and fusion reactors.

As the Appellant has demonstrated by unchallenged test results (Figure C annexed to the letter dated 21 May 1990, and Figure D annexed to the letter dated 6 June 1991), this problem has been solved by the distinguishing features denominated in the last paragraph of Point 5 above.

7. Inventive step

Of the documents relied upon by the Respondents during the appeal procedure, only document OII(14) deals with the "Helium production in fast breeder reactor out-of-core structural components". All the other documents investigate only the IGSCC effects and neglect the additional irradiation induced effects enumerated above. Document OI(2) (page 4, penultimate paragraph), published 1978, even acknowledges the existence of such irradiation induced damages but explicitly declares that they were not considered.

Document OII(14) reports about measurements of the cross sections of (n, alpha) reactions occurring with nickel and boron in a stainless steel which, starting from a AISI 316

basic composition, was modified down to very low contents of carbon (about 0.0012 wt.%) and nitrogen (about 0.006 wt.%) (see page 369, Table I). Document OII(14) has concentrated on these reactions which are provoked by relatively slow neutrons, because these reactions are a major source of damages in out-of-core structural elements of fast breeders the investigation of which is the major concern of this document (page 378, "Conclusion"). ~~Threshold reactions which play a major role with the types~~ of damages which are typically provoked in austenitic steels by the hard in-core neutron irradiation were completely neglected in these investigations. There is no reason whatsoever based on this document which could lead a skilled person to the measure of adjusting the nitrogen and carbon contents of a 316 type austenitic steel to the well defined and narrow ranges indicated in Claim 1. On the contrary, the skilled person would have expected that the relatively high amount of nitrogen would constitute an additional source of helium production in an in-core environment (for instance wrapper tubes), where neutrons having an energy above the threshold for the respective nitrogen reaction are of considerable importance (document OII(14), page 366).

The optimised steels on the basis of the standard types AISI 304L and 316L suggested in the closest prior art documents were developed with respect to a maximum resistance to IGSCC. The skilled metallurgist knows that a further reduction of the carbon content to below 0.02% would not have brought about a further improvement in this respect, but would have resulted in a considerable loss in mechanical strength which in the optimised steels had just been maintained by a sophisticated adjustment of the basic composition of the steel. Consequently, the skilled person would have had a strong aversion against a further reduction of the carbon content from the level typical for

304L and 316L steels down to the level indicated in Claim 1. The adverse influence of the (n, alpha) reaction on a steel that was subjected to a hard neutron irradiation was known from OII(14), and there was also no incentive to increase the nitrogen content of a steel which was going to be used for manufacturing for instance a wrapper tube.

The Board is aware that methods had existed on an industrial scale to produce steels reproducibly with a defined but very low carbon and nitrogen contents before the priority date of the patent in suit. Consequently, there is no doubt that the subject-matter of the patent in suit can be realised (see also other aspects of sufficiency in Point 3 above). No proof has, however, been submitted that an austenitic steel with such a composition was made available to the public before priority date and that, at least, there had been obvious reasons for a skilled person for making the material for use in order to solve the basic technical problem of the patent in suit.

Many experts in this technical field must have considered the basic problem of the patent in suit and it appears that the Appellant was the first to find a solution in a non-obvious manner.

The Board, therefore, comes to the conclusion that the subject-matter of Claim 1 cannot be derived in an obvious manner from the documents cited by the Respondents and must accordingly be seen as involving an inventive step in the meaning of Article 52(1) in combination with Article 56 EPC.

8. The independent Claim 1, together with the dependent Claims 2 to 6 and the revised description adapted thereto, can, therefore, form the basis for maintaining the patent as amended.

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 on the basis of Claims 1 to 6 filed together with the description during the oral proceedings as well as all the figures as granted.

The Registrar



S. Fabiani

The Chairman


G. Szabo