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D E C I S I O N
of 28 September 2004

Case Number: T 0851/99 - 3.4.1

Application Number: 94307893.1

Publication Number: 0651397

IPC: G21C 17/022

Language of the proceedings: EN

Title of invention:

In-situ palladium doping or coating of stainless steel surfaces

Patentee:

GENERAL ELECTRIC COMPANY

Opponent:

Framatome ANP GmbH

Headword:

-

Relevant legal provisions:

EPC Art. 100(a), 54(1), (2)

Keyword:

"Novelty (yes)"

"Remittal for further prosecution (yes)"

Decisions cited:

-

Catchword:

-



Case Number: T 0851/99 - 3.4.1

D E C I S I O N
of the Technical Board of Appeal 3.4.1
of 28 September 2004

Appellant: Framatome ANP GmbH
(Opponent) Freyeslebenstrasse 1
D-91058 Erlangen (DE)

Representative: Mörtel & Höfner
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Respondent: GENERAL ELECTRIC COMPANY
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Representative: Frohwitter, Bernhard, Dipl.-Ing.
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Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 27 July 1999
rejecting the opposition filed against European
patent No. 0651397 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: G. Davies
Members: R. Q. Bekkering
G. Assi

Summary of Facts and Submissions

- I. The appellant (opponent) lodged an appeal against the decision of the opposition division, dispatched on 27 July 1999, rejecting the opposition against European patent No. 0 651 397. The notice of appeal was received on 26 August 1999, the appeal fee being paid on the same day, and the statement setting out the grounds of appeal was received on 25 November 1999.
- II. Opposition had been filed against the patent as a whole, based on Article 100(a) EPC. The opposition division held that the subject-matter of claim 1 of the patent as granted was novel and involved an inventive step (Articles 100(a), 52(1), 54 and 56 EPC) and rejected the opposition accordingly.
- III. In the grounds of appeal the appellant referred to the following documents cited in the examination and opposition proceedings:
- D1: EP-A-0 514 089
- D2: R. Winkler ea, "Senkung der Korrosionsrate im Primärkreislauf von Druckwasserreaktoren zur Begrenzung radioaktiver Ablagerungen", VGB Kraftwerkstechnik 69, volume 5, May 1989, pages 527-531
- D3: P. Cohen, "Water coolant technology of power reactors", Gordon and Breach Science Publishers, New York, 1969, pages 342-359
- D4: EP-A-0 450 440

D5: EP-A-0 450 444

D6: S.P. Kowalczyk ea, "Characterization of palladium acetylacetonate as a CVD precursor for Pd Metallization", 3rd Symposium on Chemical Perspectives of Microelectronic Materials, 30 November - 3 December 1992, Boston, USA, pages 353-358

Furthermore, the following new documents were cited in the grounds of appeal:

D7: US-A-3 294 644

D8: US-A-4 759 900

D9: US-A-4 950 449

D10: P.L. Andresen, "Effects of zinc additions on the crack growth rate of sensitized stainless steel and alloys 600 and 182 in 288 °C water", Water Chemistry of Nuclear Reactor Systems 6, BNES, London, 1992, pages 169-175

D11: US-A-5 108 697

D12: Meyers Lexikon der Technik und der exakten Naturwissenschaften, volume 1, Bibliographisches Institut, Mannheim, 1969, page 484

D13: Kerntechnik, editor W. Riezler ea, B.G. Teubner Verlagsgesellschaft, Stuttgart, 1958, pages 179-193

- IV. Oral proceedings were held on 24 September 2003 and 28 September 2004.
- V. The appellant requested that the decision under appeal be set aside and the patent revoked.
- VI. The respondent requested that the appeal be dismissed and the patent be maintained as granted.

Alternatively, it was requested that the decision under appeal be set aside and the patent maintained in amended form based on one of seven auxiliary requests. It was further requested that the case be remitted to the first instance for further prosecution should the newly cited documents be admitted into the proceedings.

- VII. Claim 1 of the patent as granted reads as follows:

"A method for reducing corrosion of stainless steel components in a water-cooled nuclear reactor or associated components, characterized by the step of injecting a solution of a compound containing a metal into the water of said reactor while said reactor is operating, said metal having the property of increasing the corrosion resistance of stainless steel when incorporated in or deposited on an oxide film formed on a surface of the stainless steel, and said compound having the property that it decomposes under reactor thermal conditions to release atoms of said metal into the reactor water".

VIII. The appellant argued that the subject-matter of claim 1 as granted lacked novelty with respect to document D2. In particular, document D2 disclosed a method for reducing corrosion of stainless steel components in a water-cooled nuclear reactor. A continuous dosing of a solution of a compound containing a metal into the coolant water at temperatures above 200°C was envisaged. Consequently, the method of D2 was carried out while the reactor was at elevated temperatures, pressurised and with the circulation pumps running and thus while the reactor was operating in its broadest sense. Furthermore, the method disclosed in document D2 involved the use of compounds containing titanium or zirconium, like the patent in suit. In particular, according to the description of the patent in suit, suitable metal containing compounds included compounds of metals such as titanium or zirconium. Accordingly, the compounds used in D2 had the properties defined in claim 1 as granted.

IX. The respondent submitted that claim 1 as granted required the injection of the solution to be performed while the reactor was operating, which could only be understood in the sense that the control rods were removed and nuclear reaction took place. The method of document D2 was not disclosed to be carried out while the reactor was operating, neither was it compatible with reactor operating conditions. Furthermore, the titanium and zirconium containing compounds used in document D2 did not have the properties defined in claim 1 of the patent in suit. In particular, in D2 the metal was not incorporated in or deposited on an oxide film formed on the surface of the stainless steel, but rather metal oxide particles were deposited.

Furthermore, in D2 the compound hydrolysed instead of decomposing to release atoms into the reactor water, as defined in claim 1.

Reasons for the Decision

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

2. *Novelty of the main request*

2.1 Document D2

Document D2 is concerned with reducing the corrosion rate in the primary cooling water circuit of a pressurised water nuclear reactor in order to limit the radioactive deposits. According to D2, before the first operation of the reactor or after a complete decontamination the surfaces of the cooling water circuit show a protective passive layer. Within hours after exposure to hot water over 185-195°C, however, this passive layer is destroyed or altered. Only after the subsequent build-up of a new protective layer is the corrosion rate reduced (cf page 527, "Aufhebung des Passivzustandes"). It is noted in D2 that at water temperatures of 300°C, continuing corrosion of the steel surfaces is primarily caused by the porosity of the protective layer (cf page 530, "Konsequenzen aus der Deckschichtporosität"). Document D2 addresses ways of reducing this porosity (cf page 530, "Wege zur Verminderung der Porosität"). According to D2, if during the formation of the protective layer compounds

of titanium or zirconium are dissolved in the hot water, amorphous layers providing high corrosion protection may form. In particular, in a running large-scale experiment, the complex Ti/H₂O₂/EDTA is continuously dispensed into the hot cooling water circulating in the reactor of a power plant. Furthermore, the use of a Zr/EDTA complex is suggested for reactor circuits with stainless steel (X8 CrNiTi 18 10) piping.

Claim 1 as granted requires that the metal has the property of increasing the corrosion resistance of stainless steel when incorporated in or deposited on an oxide film formed on a surface of the stainless steel.

The board concurs with the appellant that, since according to the description of the patent in suit metals such as titanium and zirconium can be used for the invention (cf page 4, lines 28 to 29; page 6, lines 49 to 50), these metals, which correspond to the metals used in D2, necessarily have this required property. Moreover, it is noted that the method of D2 is based on a continuous supply of titanium or zirconium at the surface of the steel surface during the formation of the corrosion protective layer. This process presumably leads to an incorporation of titanium or zirconium, ie of the metal, into the spinel lattice of the growing corrosion protective oxide layer. However, even if it were to result in a filling of pores in this oxide layer through a superficial deposition of metal oxide particles, as argued by the respondent (cf D2, page 531, left-hand column, second paragraph), the metal still can be said to be included in or deposited on an oxide film formed on the surface, as required by claim 1 as granted.

Claim 1 as granted furthermore defines the compound as having the property that it decomposes under reactor thermal conditions to release atoms of the metal into the reactor water.

According to an embodiment of the invention disclosed in the patent in suit, palladium acetylacetonate decomposes in the hot reactor water to release palladium, in ionic form actually (cf page 5, lines 17 to 23). In the method known from document D2, the titanium or zirconium/EDTA complex hydrolyses in the hot reactor water, whereby, at first, the complex decomposes to release a metal ion. The respondent's argument that hydrolysis is a chemical reaction and not a decomposition is not found convincing in this respect. In document D2, eventually the hydrolysis results in the formation of titanium or zirconium oxide particles. In the board's view, however, the same is bound to occur in the patent in suit when eg titanium or zirconium is used as the metal. Accordingly, also this feature of claim 1 in suit cannot provide a distinction over document D2.

There remains, however, the feature of claim 1 as granted according to which the solution of the compound is injected while the reactor is operating. The appellant argued that the expression "while the reactor is operating" in its broadest sense would include an operating condition of the reactor with the cooling water at elevated temperatures and pressurised, and with the circulation pumps running. In the board's opinion, however, to the skilled reader claim 1 as

granted requires that the nuclear reaction takes place in the reactor.

In document D2 the solution of the compound is injected in the hot circulating cooling water of a nuclear reactor of a power plant. There is, however, no clear indication that the nuclear reactor is operating at this time. The respondent argued that D2 is concerned with a treatment of the surfaces of the coolant circuit prior to the operation of the reactor. In particular, the indication in document D2 that the undesirable formation of complexes of the released EDTA and corrosion products of the steel may be eliminated by varying the temperature of the cooling water (cf D2, page 531, fourth paragraph), was seen as evidence that the reactor was not in operation. Furthermore, forming the protective layer during operation was considered to go against the aim of D2 to reduce radioactive deposits. On the other hand, the board notes that document D2 indicates that the protective layer takes about 1000 hours to form. Although arguably, the addition of the metal complex could shorten this time period, doubts may arise whether it is plausible that the cooling water is circulated and heated by means of the pumps, or possibly auxiliary heaters, without the nuclear reactor being switched on for such time spans. In order to be prejudicial to the novelty of the claimed subject-matter, however, the feature that the injection of the solution of the compound takes place while the reactor is operating should be derivable directly and unambiguously from document D2, which is not the case, as is apparent from the above.

Accordingly, the subject-matter of claim 1 of the patent as granted is novel with respect to document D2 (Articles 100(a), 52(1), 54(1) and (2) EPC).

2.2 Further documents

Documents D7 to D13 were filed by the appellant with the grounds of appeal, in particular to counter the argument underlying the decision under appeal that the skilled person usually would not intervene in an operating reactor. It is clear, from a *prima facie* analysis of the documents, that all of them, to a greater or lesser extent, are relevant to the issue of inventive step, and, as a matter of fact, at least some of them even appear relevant to the issue of novelty. In the oral proceedings before the board the appellant has in particular already indicated that document D9 would be prejudicial to the novelty of the subject-matter of claim 1 of the patent as granted.

These documents, together with documents D4 to D6 already considered relevant in the examination procedure, are therefore admitted into the proceedings.

2.3 Remittal

The respondent has requested remittal of the case to the first instance for a consideration of the documents newly filed in the appeal proceedings, should the board decide to admit these documents.

A concluding consideration by the board of the issue of novelty and inventive step with respect to all documents in the proceedings would clearly present the

advantage of a swift final decision on the case, thereby meeting the interests of the public and the office. On the other hand, in the present case a remittal of the case would meet the interests of the respondent in that it would allow it to defend its case in relation to the newly admitted documents before the first instance.

In view of the fact that the newly filed documents give rise to a new situation in particular with respect to the issue of novelty, and also in view of the fact that the respondent has explicitly requested remittal of the case for consideration of the documents at first by the opposition division and the fact that the appellant gave its consent in the oral proceedings to the remittal, the board makes use of the powers conferred on it by Article 111(1) EPC to remit the case to the first instance for further prosecution.

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 for further prosecution.

The Registrar:

The Chairman:

R. Schumacher

G. Davies