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D E C I S I O N
of 19 June 2002

Case Number: T 0915/00 - 3.4.2

Application Number: 94900026.9

Publication Number: 0670916

IPC: C25D 5/18, C25D 3/12

Language of the proceedings: EN

Title of invention:
Nanocrystalline Metals

Patentee:
Integran Technologies Inc.

Opponent:
Atotech Deutschland GmbH

Headword:
-

Relevant legal provisions:
EPC Art. 83, 56

Keyword:
"Sufficiency of the disclosure (yes)"
"Inventive step (yes); circumstantial indications of inventive step: commercial implementation, licensing, recognition of the inventor's merits by the scientific community"

Decisions cited:
-

Catchword:
-

Case Number: T 0915/00 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 19 June 2002

Appellant: (Proprietor of the patent)
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Decision under appeal: **Decision of the Opposition Division of the European Patent Office posted 21 June 2000 revoking European patent No. 0 670 916 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: A. G. Klein
B. J. Schachenmann

Summary of Facts and Submissions

I. European patent No. 0 670 916 (application No. 94 900 026.9) was revoked by decision of the opposition division.

The reasons for the revocation were that the European patent did not disclose the invention defined in independent claims 1, 19 and 20 in a manner sufficently clear and complete for it to be carried out by a person skilled in the art and that the subject-matter of independent claim 18 was not novel within the meaning of Article 54 EPC in view of the disclosure in document

D13: A. M. El-Sherik et al., Deviations from Hall-Petch Behaviour in As-prepared Nanocrystalline Nickel, Scripta Metallurgica et Materialia, Vol. 27, pages 1185 to 1188, 1992.

II. The appellant (patentee) lodged an appeal against the decision revoking the patent.

III. Oral proceedings were held before the board on 19 June 2002.

The following documents and pieces of evidence were referred to by the parties at the oral proceedings, in addition to document D13:

D1: W. Kleinekathöfer et al., Die Eigenschaften von mit pulsierendem Gleichstrom (Pulse Plating) abgeschiedenem Nickel, Metall Oberfläche, 9-1982, pages 411 to 420;

D3: T.-P. Sun et al., Plating with Pulsed and

Periodic-Reverse Current, Metal Finishing, May 1979, pages 33 to 38;

D4: W. Kim et al., Pulse Plating Effects in Nickel Electrodeposition, Surface and Coatings Technology, 38 (1989), pages 289 to 298;

D5: N. R. K. Vilambi et al., Selective Pulse Plating from an Acid Copper Sulfate Bath, Plating and Surface Finishing, January 1988, pages 67 to 72;

D6: D. S. Lashmore et al., Electrodeposition of Nickel-Chromium Alloys, Plating and Surface Finishing, March 1986, pages 48 to 55;

D7: W. Kleinekathöfer, Der Einfluß von Strommodulationen auf die Eigenschaften von galvanisch abgeschiedenem Nickel, Dissertation, 1980, Fakultät für Bergbau und Hüttenwesen der Rheinisch-Westfälischen Technischen Hochschule Aachen;

D10: K. Boyan et al., An In-situ TEM Study of the Thermal Stability of Nanocrystalline Ni-P, Scripta Metallurgica et Materialia, Vol. 25, No. 12, 1991, pages 2711 to 2716;

D11: A. M. El-Sherik et al., Grain Growth Behaviour of Nanocrystalline Nickel, Materials Research Society Symposium Proceedings, Vol. 238, 1992, pages 727 to 732;

D12: D. Osmola et al., Microstructural Evolution at Large Driving Forces during Grain Growth of Ultrafine-Grained Ni-1.2wt%P, Phys. Stat. Sol.,

(a) 131, 1992, pages 569 to 575;

D17: G. Palumbo et al, Intercrystalline Hydrogen Transport in Nanocrystalline Nickel, Scripta Metallurgica et Materialia, Vol. 25, No. 3, 1991, pages 679 to 684;

D18: Experimental Report filed by the appellant with its letter dated 31 October 2000;

D19: K. J. Bryden et al., Pulsed Electrodeposition Synthesis and Hydrogen Absorption Properties of Nanostructured Palladium-Iron Alloy Films, J. Electrochem. Soc., Vol. 145, No. 10, October 1998, pages 3339 to 3346;

D20: H. Natter et al., Nanocrystalline Palladium by Pulsed Electrodeposition, Phys. Chem. 100, 1996, pages 55 to 64;

D21: H. Natter et al., Nanocrystalline Copper by Pulsed Electrodeposition: The Effects of Organic Additives, Bath Temperature, and pH, J. Phys. Chem. 1996, pages 19525 to 19532;

D22: H. Natter et al., Nanocrystalline Nickel and Nickel-Copper Alloys: Synthesis, Characterization, and Thermal Stability, J. Mater. Res., Vol. 13, No. 5, May 1998;

D25a: Theory and Practice of Pulse Plating, edited by Jean-Claude Puippe and Frank Leaman, published by the American Electroplaters and Surface Finishers Society, Orlando, USA, 1986;

D27-32 Experimental reports filed by the appellant with its letter dated 31 October 2000;

D34: Experimental reports filed by the appellant with its letter dated 31 October 2000;

D37: Experimental report filed by the respondent with its letter dated 27 April 2001;

D40: Affidavit by Dr W. Kleinekathöfer filed by the appellant with its letter dated 16 May 2002;

D41: G. McMahon et al., Structural Transitions in Electroplated Ni-P Alloys, Journal of Materials Science Letters 8, 1989, pages 865 to 868 as filed by the appellant with its letter of 28 April 2000 and numbered D41 by the board.

The appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of an amended set of claims filed at the oral proceedings of which independent claims 1 and 18, the only independent claims, read as follows:

"1. A process for electrodepositing a selected metallic material in nanocrystalline form on a substrate in which an aqueous, electrolyte containing ions of said selected metallic material is introduced into an electrolytic cell having an anode and a cathode, while maintaining said electrolyte at a temperature in the range between about 15° and about 75°C, characterised by passing a D.C. current, having a peak current density in the range between about 0.1 and about 3.0 A/cm^2, at pulsed intervals during which said current passes for a time period in the range of about 0.1 to about 50 milliseconds and does not pass for a time period in the range of about 1 to about 500 milliseconds, between said anode and said cathode so as to deposit said selected metallic material in nanocrystalline form and having a grain size of less than 100 nm on said cathode."

"18. A nanocrystalline nickel material produced by a process according to claim 1 characterised by an average grain size of less than 11 nanometres and by a hardness which is at a maximum in a size range of 8 - 10 nm, and by saturation magnetization properties substantially equal to those of said nickel material in normal crystalline form."

The respondent requested that the appeal be dismissed.

The board announced its decision at the end of the oral proceedings.

IV. In support of its request the appellant first submitted that the claimed process was sufficiently disclosed in the patent specification. Due account should in this respect be taken of the fact that the skilled person concerned here was a highly qualified scientist with a superior university degree in Chemical Physics, basic knowledge in electroplating as summarized for instance in document D25a, and a specialization in nanostructured materials. Although the specification of the patent only describes few specific embodiments of the process in relation to the electrodeposition of nanocrystalline nickel material, it also provides general instructions and recommendations in respect of the proper selection of parameters set out in claim 1 on the basis of which such highly qualified scientist could without undue difficulty and by way of simple empirical experiments as usual in this field, easily determine adequate electrodisposition conditions also for materials other than nickel.

Concerning novelty and inventive step the appellant submitted that the process of claim 1 was novel. In particular, the prior art documents D1 to D7 failed to disclose the essential feature that the pulse plating parameters set out in the claim are those which actually prevail between the anode and the cathode, rather than at the output of the current supply. The processes disclosed in these documents did not achieve nanocrystalline materials.

The subject-matter of independent product claim 18 was also novel in view of the cited prior art. In particular, document D13 was silent as to the saturation magnetisation properties of the nanocrystalline material disclosed there.

Concerning inventive step, although the ranges of the pulse plating conditions set out in claim 1 were known in substance from document D1 to D7, there was no obvious reason for the skilled person to expect that adequately selected conditions in the known ranges could achieve the deposition of nanocrystalline material having a grain size of less than 100 nm.

The inventive character of the invention was confirmed by a number of circumstantial indications summarized in the table "Indicia of Non-Obviousness" handed over at the oral proceedings of 19 June 2002, amongst which the quick acceptance of the claimed technology and it's commercial success as evidenced by its practical use and its licensing in the repair of nuclear reactors, or the unanimous recognition of the inventors' merits by peer experts, as evidenced by the about 50 invited presentations given by Prof. Erb, one of the inventors, around the world over the past 8 years and the three scientific awards he obtained for his contributions to the field of nanostructured materials.

V. The respondent's arguments in support of its request can be summarized as follows.

The skilled person concerned here was not a highly qualified scientist, but rather a graduated chemical engineer with basic knowledge and practical experience in the art of electrodeposition. The claimed combination of electrodeposition parameters was well-known from the prior art as illustrated for instance by documents D1 to D7, and there was no doubt that the skilled person would have understood from these documents that the current conditions defined in the claim are those which actually prevail between the electrodes, as is evidenced for instance by the affidavit D40 by the author of documents D1 and D7 filed by the appellant himself. Therefore, claim 1 did not define any contribution to the state of the art and, if it was admitted that the prior art did not achieve nanocrystalline structures, then obtaining such structures could only result from technical circumstances which were not set out in the claims and were not available to the skilled person at the filing date of the patent, such as for instance particular compositions of the deposition baths or specific choices of grain refining agents. Experimental report D37 in this respect showed that the electrodeposition conditions stated in claim 1 did not yield a nanocrystalline structure.

The specification of the patent also showed that different techniques were available to determine grain size of the deposited product, which provide different grain size values (see column 7, lines 43 to 47). Claim 1 failing to specify which technique should be employed for determining whether the deposited material exhibits the claimed grain size of less than 100 nm, the skilled person could not use this criterion to experimentally determine adequate deposition parameters.

Thus, from the specification of the patent which only discloses the deposition of nanocrystalline nickel from a single bath composition, the skilled person could not practise the claimed process which is not restricted to any particular material.

Concerning the patentability of the claimed subject-matter, documents D1 to D7 not only disclosed the claimed ranges of electrodeposition parameters, but they also expressly pointed at the grain refining effect of short current pulses. Accordingly, if it was admitted that the processes disclosed in these documents did not yet achieve nanocrystalline structures, these would result from an obvious extrapolation of the process conditions disclosed there in conjunction with the manufacturing of microstructures.

In addition, nanocrystalline materials, and in particular the nanocrystalline material defined in independent claim 18, were known from documents D10 to D13 and said there to be obtainable by electrodeposition. Accordingly, the material of independent claim 18 was not novel over document D13 and the process of independent claim 1 also resulted from the obvious implementation of the processes of documents D1 to D7 for manufacturing the products praised in documents D10 to D13.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

The alternative (b) set out in dependent claim 2 as granted was deleted, and so were independent claims 19 and 20 as granted and dependent claims 21 to 24 as appended thereto.

Independent claim 18 was supplemented by an indication that the nanocrystalline nickel material it defines is "produced by a process according to claim 1".

Dependent claims 25 to 26 were re-numbered claims 19 to 20.

The specification was merely adapted to the amended version of the claims and Figure 5 was deleted.

These amendments undisputedly meet the requirements of Article 123(2) and (3) EPC.

3. *Sufficiency of the disclosure*

Claim 1 defines a process for electrodepositing a non-specified metallic material in nanocrystalline form on a substrate in an electrolytic cell having an anode and a cathode between which a direct current is passed at pulsed intervals. The claim defines ranges for the temperature of the electrolyte, the peak current density and the time periods for which current passes or not, and it specifies that the process shall be so conducted as to deposit the metallic material in nanocrystalline form and having a grain size of less than 100 nm on the cathode. Since such nanocrystalline material is not obtained for each arbitrary combination of parameter values in the ranges set out in the claim, as is admitted by both parties and is evident from the prior art documents D1 to D7 which disclose several embodiments in which process conditions within the ranges of claim 1 do not achieve nanocrystalline material, claim 1 shall be construed as meaning that, within the ranges it defines, combinations of parameter values have still to be selected so as to achieve the desired nanocrystalline material.

To assist the skilled person in selecting an appropriate combination of parameter values for a given material the specification of the patent on the one hand comprises four examples, which all describe the deposition of nanocrystalline nickel from a same electrolyte bath and under the same electroplating conditions and which differ only by the amount of stress reliever and grain refining agent (see column 5, lines 2 to 57). The specification on the other hand provides a series of recommendations as to the proper selection of the electrodeposition conditions: the quality of the deposit and the nanocrystalline structure thereof are functions of the peak current density in the cell and the rate of pulsing the current, the time off is generally longer than the time on, if the peak current density is too high, there is a risk that the deposited material will burn and, if too low, the grain size will increase (see the paragraph bridging column 4 and 5).

Thus, the issue to be decided in respect of the sufficiency of the disclosure is whether the skilled person could on the basis of his general knowledge and of the above indications and without undue burden determine adequate combinations of parameter values allowing the obtaining of nanocrystalline structures also of materials other than nickel.

The board in this respect first notes that according to the jurisprudence of the boards of appeal, an objection for lack of sufficient disclosure shall only be raised if there are serious doubts, substantiated by verifiable facts. The mere fact that the claim is broad is not in itself a ground for considering the application as not complying with the requirement of sufficient disclosure under Article 83 EPC (see T 19/90 OJ 1990, 476). In the present case, however, the respondent in substance only relied upon experimental report D37 to show that applying parameter values in the claimed ranges in three experiments failed to achieve nanocrystalline copper deposits. There is however no doubt that any arbitrary combination of parameter values will not necessarily in the obtaining of nanocrystalline material. The mere failing of three such arbitrary combinations cannot establish that the skilled person could not possibly have devised successful electrodeposition conditions within the claimed parameter ranges, from his normal knowledge and capacity.

The board can in this respect agree to the appellant's definition of the skilled person as a highly qualified scientist well aware of the latest developments in nanocrystalline materials and electrodeposition. This view is indeed consistent with the observation that most of the numerous relevant prior art citations in the file consist of articles from scientific publications, disclosing fundamental research work rather than for instance practical developments in industrial equipment.

The credibility of the numerous experimental reports filed by the appellant to demonstrate that various materials can be deposited in a nanocrystalline form under conditions meeting the parameter ranges of claim 1 (see D18, D27 to D32 and D34) is supported by documents D19 to D22 published after the filing date of the patent in suit. These scientific publications do not in any way suggest the existence of particular difficulties in the selection of proper deposition conditions or bath compositions.

The respondent also questioned the sufficiency of the disclosure in the patent in suit on the ground that the specification did not unambiguously specify how the grain size of less than 100 nm referred to in claim 1 was to be measured, but disclosed instead two distinct procedures which gave different results, namely scanning electromicroscopy and x-ray diffraction. In the board's view, however, the specification clearly indicates that the 100 nm value was referred to merely as a generally recognised grain size limit below which the material was defined as being nanocrystalline (see column 1, lines 29 to 33). In addition, the results of the two grain size measurement procedures referred to in the specification are fully consistent, scanning electromicroscopy indicating a grain size less than 100 nm while x-ray diffraction gave grain size values of about 10 to 15 nm with some grain sizes up to about 37 nm (see column 7, lines 43 to 47).

For the above reasons, the board is satisfied that the process of independent claim 1 is disclosed in the patent in a manner sufficiently clear and complete for it to be carried out by the person skilled in the art.

The same conclusion holds true for the nanocrystalline nickel material defined in independent claim 18, the preparation of which is described in details in lines 2 to 57 of column 2 of the specification, with reference to Examples 1 to 4.

Independent claims 19 and 20 as granted, the subject-matter of which had also been considered insufficiently disclosed by the opposition division in the decision under appeal have not been maintained by the appellant.

4. *Novelty*

4.1 Independent process claim 1

Documents D1 to D7 disclose processes for electrodepositing various metallic materials by passing a direct current between electrodes under electrolyte temperature, peak current density and pulse time conditions which fall within the ranges set out in claim 1. These documents do not however disclose that the process is so conducted as to deposit said metallic materials in nanocrystalline form.

Document D17 discloses nanocrystalline nickel material obtained by continuous, rather than pulsed, electrodeposition. Documents D10 to D13 also disclose nanocrystalline materials, but fail to describe how these can be obtained.

The remaining documents in the file do not come closer to the subject-matter of claim 1, which accordingly is novel within the meaning of Article 54 EPC.

4.2 Independent product claim 18

Document D13 is the only citation in the file to disclose a nanocrystalline nickel material with an average grain size of less than 11 nm and a hardness which is at a maximum in a size range of 8 to 10 nm (see the similarity between Figure 3 of document D13 and Figure 3 of the patent). The document does not specify whether the saturation magnetisation properties of this material are substantially equal to those of nickel material in normal crystalline form as is set out at the end of claim 18. In any case, document D13 completely fails to disclose how the material it describes was or could be obtained. Neither is there any indication that the material as such was available to the public. This document therefore does not provide an enabling disclosure which might anticipate the product defined in claim 18 (see decision T 206/83, OJ 1987, 5 and G 1/92, OJ 1993, 277).

The remaining documents in the file do not come closer to the above product. Documents D10 to D12 in particular describe nanocrystalline materials having an average grain size of less than 11 nm as set out in claim 18, but these materials are constituted by nickel alloys instead of nickel, and the documents do not disclose their hardness and saturation magnetization properties. Moreover, they were obtained by continuous rather than by pulsed electrodeposition, which certainly implies structural differences as compared to materials susceptible of being produced by the pulsed electrodeposition process of claim 1 as referred to in claim 18. Document D17 describes nanocrystalline nickel material which is also obtained by continuous electrodeposition and has an average grain size above 11 nm.

For these reasons, the subject-matter of independent claim 18 is novel within the meaning of Article 54 EPC.

5. *Inventive step*

5.1 The patent in suit generally relates to the manufacturing of nanocrystalline materials by electrodeposition and to materials so obtained.

Such a manufacturing process is disclosed in document D41 (see page 865, the third and the fourth paragraphs) and nanocrystalline materials obtained thereby are disclosed in documents D10, D11 and D12 (document D41 is bibliographic reference 4 in document D10, reference 1 in document D11 and reference 7 in document D12). The process of document D41, in which current is passed continuously between the electrodes, in the board's view comes closer to the process set out in claim 1 of the patent in suit than the processes described in any of documents D1 to D7, because the latter do not achieve nanocrystalline materials.

5.2 The method of present claim 1 is distinguished from the continuous electroplating method of document D41 essentially in that it comprises passing direct current at pulsed intervals and under peak current density and timing conditions selected in the ranges set out in the claim so as to deposit nanocrystalline material having a grain size of less than 100 nm on said cathode, instead of passing direct current in a continuous manner.

The nanocrystalline materials obtained by the process of D41 exhibit grain sizes comparable to those disclosed in the patent in suit (see D10, pages 2711, the penultimate paragraph: between 5 and 10 nm; D11, page 728, Table 1 and Figure 1: between 5 and 10 nm; D12, page 573, Figure 5: mostly between 4 and 10 nm as compared to the 6 nm or the 11 nm grain size of Examples 3 and 4 of the patent in suit).

Thus, the technical problems solved by the process of claim 1 can be seen in proposing an alternative to the known manufacturing process.

5.3 Although, as established under point 2 above in relation to the question of the sufficiency of the disclosure, once the skilled person has contemplated using pulse electrodeposition for the manufacturing of nanocrystalline material there would be no undue difficulty for him to select adequate deposition conditions so as to obtain the desired material, the prior art in the board's view is devoid of any encouragement for him to do so.

Documents D1 to D7 indeed expressly point at the grain refining effect of current pulsing, but only in the context of microcrystalline structures and the skilled person had no obvious reason to foresee that this could still be extrapolated to structures smaller by at least two orders of magnitudes, if not with the benefit of hindsight.

The practical application of the claimed process in the nuclear reactor maintenance technology, its licensing in the electrical power generation industry and the recognition of the inventors' contribution by the scientific community as evidenced by the list of invited presentations given by Prof. Erb and of awards he received, as filed by the appellant at the oral proceedings, in the board's view also constitute convincing further circumstantial indications of the presence of an inventive step.

For the above reasons, the board came to the conclusion that the process set out in claim 1 of the patent in suit involves an inventive step within the meaning of Article 56 EPC.

5.4 The same conclusion applies to the subject-matter of independent product claim 18, the structure and properties of which could only result from the performance of the inventive process of claim 1 and to the subject-matter of the remaining dependent claims by virtue of their appendence to independent claim 1.

6. For the above reasons, the patent as amended in accordance with the appellant's request and the invention to which it relates meet the conditions of the convention.

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 as follows:

    - claims 1 to 20 and description pages 2 to 6, presented at the oral proceedings of 19 June 2002;

    - drawings as in the patent specification without Figure 5.

The Registrar: The Chairman:

P. Martorana E. Turrini