



(12) **EUROPEAN PATENT APPLICATION**
published in accordance with Art. 153(4) EPC

(43) Date of publication:
09.01.2019 Bulletin 2019/02

(51) Int Cl.:
B22F 9/26 (2006.01) B22F 1/00 (2006.01)

(21) Application number: **17759576.6**

(86) International application number:
PCT/JP2017/004499

(22) Date of filing: **08.02.2017**

(87) International publication number:
WO 2017/150105 (08.09.2017 Gazette 2017/36)

(84) Designated Contracting States:
AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO RS SE SI SK SM TR
Designated Extension States:
BA ME
Designated Validation States:
MA MD

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(30) Priority: **29.02.2016 JP 2016038185**

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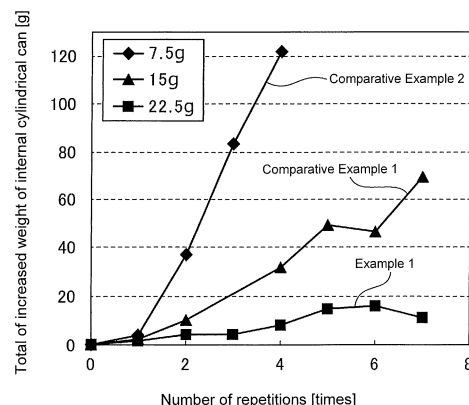
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(54) **NICKEL POWDER PRODUCTION METHOD**

(57) Provided is a method for controlling generation of scaling in a reaction vessel to reduce time and cost required for removal of the scaling in the process of producing nickel powder from a solution containing a nickel ammine sulfate complex. This is a method for producing nickel powder, including: adding, to the solution containing the nickel ammine sulfate complex, seed crystals in an amount of 0.3 times or more and 3 times or less the weight of nickel in the solution to form a slurry; and blowing hydrogen gas into the slurry to reduce a nickel complex ion and to thereby form a nickel precipitate.

Fig. 1



Description

Technical Field

[0001] The present invention relates to a method for controlling scaling in a reaction vessel in a method for producing nickel powder from a solution containing a nickel ammine sulfate complex. Particularly, the present invention can be applied to a treatment of an in-process intermediate solution generated from a nickel hydrometallurgical process.

Background Art

[0002] Methods for producing fine nickel powder have been known including atomization methods of dispersing molten nickel in gas or water to obtain fine powder, and dry processes, such as CVD processes, of volatilizing nickel, and reducing the nickel in a gaseous phase to obtain nickel powder as described in Patent Literature 1.

[0003] Methods for producing nickel powder by a wet process include a method of producing nickel powder using a reducing agent as described in Patent Literature 2, and an atomization thermal decomposition method of atomizing a nickel solution in a reduction atmosphere at a high temperature to obtain a nickel powder through a thermal decomposition reaction as described in Patent Literature 3.

[0004] However, these methods are not economical because these methods require expensive reagents and a large amount of energy.

[0005] In contrast, a method as described in Non Patent Literature 1 is industrially inexpensive and useful, in which hydrogen gas is fed to a nickel ammine sulfate complex solution to reduce nickel ions in the complex solution, obtaining a nickel powder.

[0006] This method involves the following process: a small amount of fine particles called seed crystals is added, and a reducing agent is fed thereto to grow the seed crystals into a powder having a predetermined particle size. However, in this method, the so-called scaling, which is part of nickel precipitated on the walls of the reaction container in the apparatus rather than on the seed crystals, may be formed in the reaction vessel, causing failure such as clogging of the pipes.

[0007] For this reason, an operation of removing scale is required by, e.g., stopping of the apparatus regularly, resulting in a reduction in productivity and an increase in maintenance cost required for the removal. Accordingly, the problem has been control of the generation of scaling as much as possible.

Citation List

Patent Literature

[0008]

Patent Literature 1: Japanese Patent Application Laid-Open No. 2005-505695

Patent Literature 2: Japanese Patent Application Laid-Open No. 2010-242143

Patent Literature 3: Japanese Patent No. 4286220

Non Patent Literature

[0009] Non Patent Literature 1: "The Manufacture and properties of Metal powder produced by the gaseous reduction of aqueous solutions", Powder metallurgy, No. 1/2 (1958), pp. 40-52.

Summary of Invention

Technical Problem

[0010] In such circumstances, the present invention provides a method for controlling the generation of scaling in a reaction vessel to reduce time and cost required for removal of the scaling in the process of producing nickel powder from a solution containing a nickel ammine sulfate complex.

Solution to Problem

[0011] A first aspect of the present invention to solve the above problem is a method for producing nickel powder, including: adding, to a solution containing a nickel ammine sulfate complex, seed crystals in an amount of 0.3 times or more and 3 times or less the weight of nickel in the solution to form a slurry; charging the slurry into a reaction vessel, maintaining a liquid phase portion occupied by the slurry and a gaseous phase portion other than the liquid phase portion within the reaction vessel; and blowing hydrogen gas into the slurry to reduce a nickel complex ion and to thereby form a nickel precipitate.

[0012] A second aspect of the present invention is a method for producing nickel powder, wherein of a polyacrylate salt dispersant is added to the slurry according to the first aspect in an amount of 0.5 to 5% based on the weight of the seed crystals added to the slurry.

[0013] A third aspect of the present invention is a method for producing nickel powder, wherein a solution containing a nickel ammine sulfate complex is added to the nickel precipitate according to the first and second aspects, and hydrogen gas is blown thereinto to precipitate and grow nickel on the nickel precipitate.

[0014] A fourth aspect of the present invention is a method for producing nickel powder, wherein the temperature of the slurry is 150 to 200°C when hydrogen gas is blown into the slurry according to the first to third aspects.

[0015] A fifth aspect of the present invention is a method for producing nickel powder, wherein the pressure of the gaseous phase portion in the reaction vessel is in the range of 1.0 to 4.0 MPa when hydrogen gas is blown into

the slurry according to the first to fourth aspects.

Advantageous Effects of Invention

[0016] The present invention can control the generation of scaling. For this reason, the frequency of removal of scale can be reduced, resulting in a reduction in time and cost.

Brief Description of Drawing

[0017] [Figure 1] This is a diagram illustrating a relation between the amount of nickel powder to be added, the number of repetitions, and the amount of scaling generated.

Description of Embodiments

[0018] The present invention is a method for producing nickel powder by adding seed crystals to a nickel ammine sulfate complex solution, and blowing hydrogen gas thereinto to produce a nickel powder.

[0019] Hereinafter, the method for producing nickel powder according to the present invention will be described.

[Nickel ammine sulfate complex solution]

[0020] A nickel ammine sulfate complex solution that can be used in the present invention is not particularly limited, but it is suitable to use a nickel ammine sulfate complex solution obtained by dissolving a nickel-containing material, such as an industrial intermediate including one or a mixture of two or more selected from nickel and cobalt mixed sulfide, crude nickel sulfate, nickel oxide, nickel hydroxide, nickel carbonate, and nickel powder, with sulfuric acid or ammonia to prepare a nickel-containing leachate (solution containing nickel), subjecting the nickel-containing leachate to a liquid-purification step such as solvent extraction, ion exchange, or neutralization to remove impurity elements in the solution, and adding ammonia to the resulting solution.

[Preparation of slurry]

[0021] In this step, seed crystals are added to the nickel ammine sulfate complex solution to prepare a slurry.

[0022] The seed crystals added here are preferably a powder having a particle size of 20 μm or less. A nickel powder is suitable as a substance which does not contaminate the final nickel precipitate. The nickel powder used as the seed crystals can be prepared, for example, by adding a reducing agent such as hydrazine to the nickel ammine sulfate complex solution.

[0023] The weight of the seed crystals added here is preferably 0.3 times or more and 3 times or less the weight of nickel in the solution. An amount of less than 0.3 times the weight of nickel in the solution cannot

achieve a sufficient effect of reducing scaling. An amount of more than 3 times has no influences over the effect; rather, this results in addition of excess crystal seeds.

[0024] Then, a dispersant may be added to disperse the seed crystals in the slurry. A dispersant used here is not particularly limited as long as it has polyacrylate salt, but sodium polyacrylate, which is industrially available at low cost, is preferable. The amount of the dispersant to be added is preferably 0.5 to 5% based on the weight of the seed crystals. An amount of less than 0.5% cannot achieve the dispersion effect. An amount of more than 5% has no influences over the dispersion effect; rather, this results in addition of an excess dispersant.

[0025] Next, the slurry prepared by the addition of the seed crystals or the seed crystals and the dispersant is charged into a reaction vessel in a container resistant to high pressure and high temperature, forming a liquid phase portion occupied by the slurry and a gaseous phase portion in the reaction vessel. Subsequently, hydrogen gas is blown into the slurry in the reaction vessel to reduce a nickel complex ion in the solution and precipitate nickel on the seed crystals added.

[0026] At this time, the reaction temperature is preferably in the range of 150 to 200°C. A reaction temperature of less than 150°C reduces the efficiency of reduction. A reaction temperature of 200°C or more has no influences over the reaction; rather, such a reaction temperature increases loss of thermal energy, and is not suitable.

[0027] Furthermore, the pressure of the gaseous phase portion in the reaction vessel during the reaction is preferably 1.0 to 4.0 MPa. If the pressure is less than 1.0 MPa, reaction efficiency will be reduced, and even if the pressure exceeds 4.0 MPa, there will be no influence on the reaction, and the loss of hydrogen gas will increase.

[0028] A precipitate of nickel is formed on the seed crystals through the reduction and the precipitation treatment under such conditions so that nickel can be extracted and recovered from the solution in the form of a fine powdery precipitate by the effect of the dispersant.

[0029] The nickel powder thus produced can be used in, for example, an application to nickel pastes as an inner constitutional substance for stacked ceramic capacitors. Besides, particles of the nickel powder can be grown through repetition of the above hydrogen reduction to produce high purity nickel metal.

Examples

[0030] Hereinafter, the present invention will be described with reference to Examples.

[Example 1]

[0031] 191 ml of 25% aqueous ammonia, 22.5 g of 1 μm nickel powder (0.3 times the weight of nickel in a mixed solution used here) as seed crystals, and 0.4 g of sodium polyacrylate (42% solution) were added to the

mixed solution of a nickel sulfate solution containing nickel equivalent to 75 g and 330 g of ammonium sulfate, and the total volume of the mixture was adjusted to 1000 ml to prepare a slurry.

[0032] The resulting slurry was then charged into an internal cylindrical can of an autoclave used as a reaction vessel to define a liquid phase portion and a gaseous phase portion. The slurry was heated to and held at 185°C with stirring. In this state, hydrogen gas was blown and fed into the slurry so that the pressure of the gaseous phase portion in the internal cylindrical can of the autoclave became 3.5 MPa, to perform a reduction treatment. Thereby, a reduced slurry was generated. After the lapse of 60 minutes from the start of feeding hydrogen gas, the feed of hydrogen gas was stopped, and the internal cylindrical can was cooled.

[0033] After the cooling, the reduced slurry in the internal cylindrical can was filtered to recover nickel powder. To the recovered nickel precipitate, a nickel sulfate solution containing nickel equivalent to 75 g, a solution containing 330 g of ammonium sulfate, and 191 ml of 25% aqueous ammonia were added and the total amount of the solution was adjusted to 1000 ml to prepare an adjusted slurry.

[0034] The adjusted slurry was reacted in the autoclave in the same manner as described above, and a thus recovered nickel precipitate was again reacted in the manner described above. This operation was repeated to grow nickel powder.

[0035] In each operation, the reduced slurry in the internal cylindrical can was extracted, and was dried. The weight of the internal cylindrical can was measured to measure a change in weight before and after the reaction.

[0036] The results are shown in Figure 1 (see a legend of 22.5 g).

[0037] As seen from Figure 1, if the seed crystal nickel powder is 22.5 g (proportion of addition: 30%), the change in weight is small according to "the number of repetitions" and the generation of scaling can be reduced.

[Example 2]

[0038] A nickel powder was grown under the same condition as in Example 1 except that the nickel powder as seed crystals was added in an amount of 3.0 times the weight of nickel in the mixed solution, i.e., 225 g.

[0039] The results similar to those in Example 1 were obtained. Even if the number of repetitions was increased, the weight of scaling generated was 20 g or less each time.

(Comparative Example 1)

[0040] The growth was repeatedly performed in the same manner as in Example 1 except that initially 15.0 g of 1 μm nickel powder (proportion of addition: 20%) and 0.3 g of sodium polyacrylate (42% solution) were added.

[0041] As a result, as shown in Figure 1 (see a legend of 15 g), it is found that the amount of scaling increased according to the number of repetitions of the reaction, i.e., as the repetition of the reaction was larger, although the increase was mild.

(Comparative Example 2)

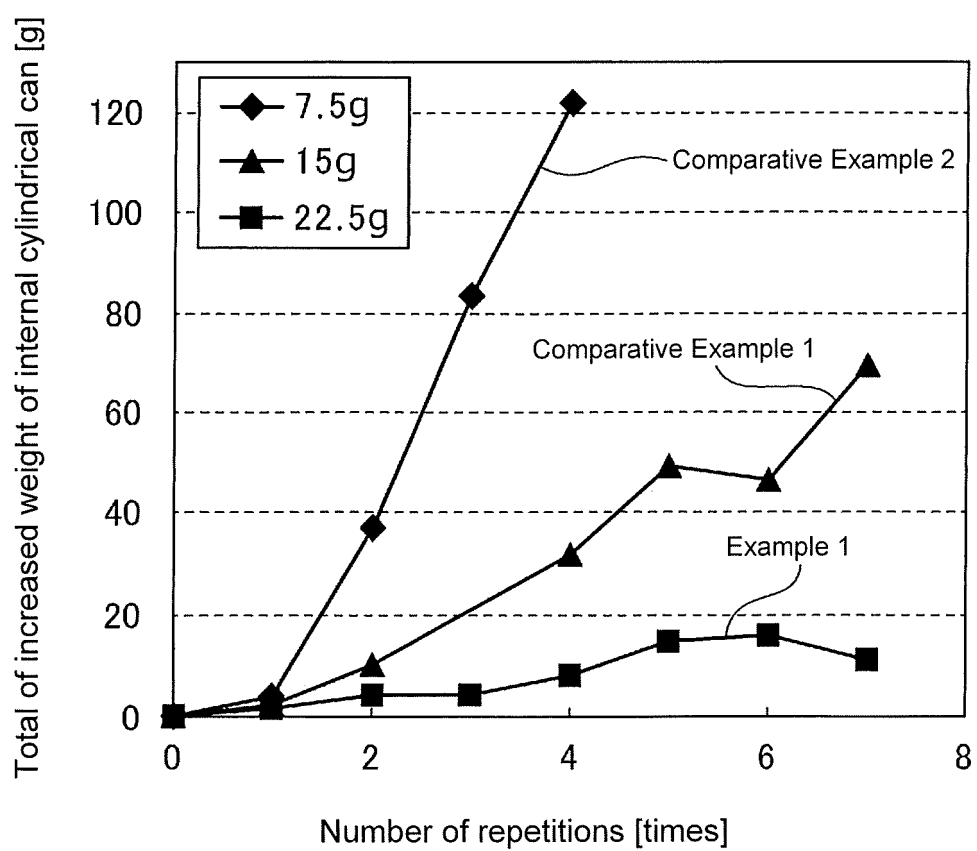
[0042] The growth was repeatedly performed in the same manner as in Example 1 except that initially 7.5 g of 1 μm nickel powder (proportion of addition: 10%) and 0.1 g of sodium polyacrylate (42% solution) were added.

[0043] As a result, as shown in Figure 1 (see a legend of 7.5 g), it is found that the amount of scaling significantly increased at the second reaction in the repetitions, and the tendency continued thereafter.

Claims

1. A method of producing nickel powder, comprising: adding, to a solution containing a nickel ammine sulfate complex, seed crystals in an amount of 0.3 to 3 times a weight of nickel in the solution to form a slurry; charging the slurry into a reaction vessel, maintaining a liquid phase portion occupied by the slurry and a gaseous phase portion other than the liquid phase portion within the reaction vessel; and blowing hydrogen gas into the slurry to reduce a nickel complex ion and to thereby form a nickel precipitate.
2. The method of producing nickel powder according to claim 1, wherein a polyacrylate salt dispersant is added to the slurry in an amount of 0.5 to 5% based on the weight of the seed crystals added to the slurry.
3. The method of producing nickel powder according to claim 1 or 2, wherein a solution containing a nickel ammine sulfate complex is added to the nickel precipitate, and hydrogen gas is blown therein to precipitate and grow nickel on the nickel precipitate.
4. The method of producing nickel powder according to any one of claims 1 to 3, wherein a temperature of the slurry is 150 to 200°C when hydrogen gas is blown into the slurry.
5. The method of producing nickel powder according to any one of claims 1 to 4, wherein a pressure of the gaseous phase portion in the reaction vessel is in a range of 1.0 to 4.0 MPa when hydrogen gas is blown into the slurry.

Fig. 1



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2017/004499

A. CLASSIFICATION OF SUBJECT MATTER

B22F9/26(2006.01) i, B22F1/00(2006.01) i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

B22F9/26, B22F1/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho 1922-1996 Jitsuyo Shinan Toroku Koho 1996-2017

Kokai Jitsuyo Shinan Koho 1971-2017 Toroku Jitsuyo Shinan Koho 1994-2017

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 5796696 B1 (Sumitomo Metal Mining Co., Ltd.), 21 October 2015 (21.10.2015), paragraphs [0020] to [0033], [0048] to [0052]; example 5 & WO 2016/117138 A1	1-5

☒ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

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Date of the actual completion of the international search
02 May 2017 (02.05.17)

Date of mailing of the international search report
16 May 2017 (16.05.17)

Name and mailing address of the ISA/
Japan Patent Office
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Tokyo 100-8915, Japan

Authorized officer

Telephone No.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2017/004499

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2015-166489 A (National University Corporation Kochi University), 24 September 2015 (24.09.2015), paragraphs [0017] to [0028], [0033] to [0037], [0041] to [0044] & JP 2015-166488 A & US 2016/0354844 A1 paragraphs [0023] to [0042], [0049] to [0052], [0060] to [0067] & US 2016/0368059 A1 & WO 2015/122535 A1 & WO 2015/122534 A1 & EP 3108986 A1 & EP 3144084 A1 & AU 2015216113 A1 & AU 2015216114 A1 & CA 2939493 A1 & CA 2939513 A1 & CN 106029268 A & CN 106029269 A	1, 3-5 2
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REFERENCES CITED IN THE DESCRIPTION

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- JP 2005505695 A [0008]
- JP 2010242143 A [0008]
- JP 4286220 B [0008]

Non-patent literature cited in the description

- The Manufacture and properties of Metal powder produced by the gaseous reduction of aqueous solutions. *Powder metallurgy*, 1958, 40-52 [0009]