Patented May 24, 1949

2,471,079

UNITED STATES PATENT OFFICE

2,471,079

GLASS SEALING ALLOY

Carl B. Post, Wyomissing, and Warren S. Eberly, Temple, Pa., assignors to The Carpenter Steel Company, Reading, Pa., a corporation of New Jersey

No Drawing. Application August 28, 1947, Serial No. 771,146

12 Claims. (Cl. 49—92.5)

1

This invention relates to iron-nickel-columbium alloys.

It is an object of this invention to produce glass-sealing alloys of the class hereinafter described.

It is a further object to produce substantially gas-free alloys for glass-sealing purposes.

It is a further object to produce an improvement in glass-to-metal seals by eliminating the evolution of gas at the glass-to-metal interface.

As is well known in the art, iron-nickel alloys show a remarkable range of expansions, depending upon nickel content. This property of obtaining a given expansion depending upon the nickel content is used to obtain alloys of iron and nickel for effecting glass-to-metal seals.

At the present time the 42% and 49% nickel seals find wide application in glass-to-metal seals, the glasses being of the borosilicate and lead-bearing types. In making iron-nickel alloys for effecting glass-to-metal seals, the nickel can be chosen to yield the desired expansion and carbon is generally held to .07% maximum, manganese .40 to 1.25%, silicon .25/.50%, phosphorus .02% maximum, sulphur .02% maximum.

In making glass-to-metal seals, the metal and glass are chosen on the basis of metal and glass expansion from the "strain point" or "setting point" of the glass. These consideration are well known to those skilled in the art and will not be discussed further here.

The metallic parts to be sealed to the glass are formed into cups, pins, tubes, etc., and then cleaned carefully to remove oil, grease and other extraneous impurities. Following this the parts are then generally given a series of costly firing treatments at temperatures of 1900° F. to 2200° F. in dry or wet hydrogen. This high temperature firing treatment can be used to "degas" the metallic parts, or to put a proper scale on the metallic parts so that the glass will "wet" the part during the glass-sealing operation.

Under the best of firing techniques, there is some evolution of gas at the glass-to-metal interface in the resulting glass-to-metal seal. This is evidenced by the dull gray appearance of the glass-metal interface. It has long been apparent to those skilled in the art that what is needed to correct this defect is an alloy which is itself substantially gas-free at the glass-to-metal interface during the glass sealing operation. In this way, the firing technique can be directed entirely to preparing the proper scale on the surface for glass-wetting properties, and the amount of firing for "degassing" can either be dispensed with, or reduced to a minimum.

2

We have discovered that the addition of 0.50 to 2.00 per cent by weight of columbium to certain iron nickel alloys creates new alloys which are substantially gas-free during glass-to-metal sealing operations. The new alloys may be identified as to composition by the following analysis:

	Percent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	30 to 60
Columbium	0.50 to 2.00

All or substantially all of the balance is iron and incidental impurities. The incidental impurities may be illustrated by sulfur e. g. 0.020 maximum and phosphorus 0.020 maximum. Certain specific ranges of nickel content falling within the above mentioned range of 30 to 60 per cent by weight are for example 40.5 to 42.0 and 47.0 to 50.0 respectively.

Theoretically, the evolution of gas during the glass-to-metal sealing operation at temperatures of 1700° F. to 2000° F. can be ascribed to a chemical surface reaction between the glass and the metallic surface, or it can be ascribed equally as well to a diffusion of some gas, such as nitrogen, hydrogen, or oxygen from out of the metal to the glass-metal interface. While we are not prepared to state which of these viewpoints is correct, the fact remains that the alloys of the present invention are substantially gas-free at the glass-to-metal interface without the necessity of a costly "degassing" firing operation.

To further illustrate the invention attention is directed to tests made upon an iron nickel alloy having the following composition:

	Per cent
Carbon	.062
Manganese	.99
Silicon	.31
Phosphorus	.010
Sulphur	.004
Chromium	.14
Nickel	41.13

and the balance substantially all iron with the exception of incidental impurities.

The test used to study the amount of gas evolved at glass-sealing temperatures is as follows: A strip of the metal to be studied is supported on spaced blocks of refractory material resting on the hearth of a furnace capable of reaching 2000° F. A piece of plate glass is laid on the specimen of metal and the temperature of the electrically heated furnace is then raised

to known temperatures, in the range 1500° F. to 2000° F. The appearance of the glass-metal interface is examined carefully for gas bubbles after being held at a given temperature for 15 minutes. A gassy-metal will show bubbles at the interface a sixteenth of an inch in diameter at all temperatures above 1500° F. Pure electrolytic copper will show no bubbling at temperatures up to 1900° F.

Using this test, the alloy having the actual analysis shown above when not "degassed" by a firing technique shows bubbles of about one sixteenth of an inch in diameter at the glass-to-metal interface, at a temperature of 1400° F. to 1700° F. and the bubbles increase in size at temperatures up to 2000° F. The same alloy base but containing .25% columbium shows no improvement in gas evolution. With a .50% columbium addition to the above base, a marked decrease in the amount of gas evolution takes place, and the specimens show no gas evolution until temperatures of 1800° F. are reached. With a .80% columbium addition the resulting alloy is substantially gas-free at temperatures up to 2000° F. Further increases in columbium can be used up to about 2.00% by weight so that the alloys will be substantially gas-free at long-time holds at temperatures of at least 2000° F.

Slight modifications of nickel content will be necessary to maintain the same expansion in the new alloy with columbium as was obtained in the old alloy without columbium. In general, it is necessary to decrease the nickel content .20% to .40% by weight for each 1.00% columbium added to maintain substantially the same expansion in the alloys with and without the columbium.

What is claimed is:

1. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	30 to 60
Columbium	0.50 to 2.00

and the balance iron and incidental impurities.

2. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	40.5 to 42
Columbium	0.50 to 2.00

and the balance iron and incidental impurities.

3. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	47 to 50
Columbium	0.50 to 2.00

and the balance iron and incidental impurities.

4. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	30 to 60
Columbium	0.50 to 2.00

and the balance iron and incidental impurities said alloy being substantially gas-free at a glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F.

5. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	40.5 to 42
Columbium	0.50 to 2.00

and the balance iron and incidental impurities said alloy being substantially gas-free at a glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F.

6. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	47 to 50
Columbium	0.50 to 2.00

and the balance iron and incidental impurities said alloy being substantially gas-free at a glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F.

7. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	30 to 60
Columbium	0.50 to 2.00

and the balance iron and incidental impurities said alloy being substantially gas-free at a glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F. without pre-firing in hydrogen at 1900° F. to 2200° F. to degasify the metal.

8. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	40.5 to 42
Columbium	0.50 to 2.00

and the balance iron and incidental impurities said alloy being substantially gas-free at a glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F. without pre-firing in hydrogen at 1900° F. to 2200° F. to degasify the metal.

9. An alloy having the composition

	Per cent by weight
Carbon (maximum)	0.07
Manganese	0.50 to 2.00
Silicon	0.25 to 1.00
Nickel	47 to 50
Columbium	0.50 to 2.00

and the balance iron and incidental impurities said alloy being substantially gas-free at a glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F. without pre-firing in hydrogen at 1900° F. to 2200° F. to degasify the metal.

10. A glass-to-metal seal between a glass and an alloy said alloy being substantially gas-free at the glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F. and having the composition carbon 0.07 per cent maximum, manganese 0.50 to 2.00 per cent, silicon 0.25 to 1.00 per cent, nickel 30 to 60 per cent, columbium 0.50 to 2.00 per cent

and the balance iron and incidental impurities.

11. A glass-to-metal seal between a glass and an alloy said alloy being substantially gas-free at the glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F. and having the composition carbon 0.07 per cent maximum, manganese 0.50 to 2.00 per cent, silicon 0.25 to 1.00 per cent, nickel 40.5 to 42.0 per cent, columbium 0.50 to 2.00 per cent and the balance iron and incidental impurities.

12. A glass-to-metal seal between a glass and an alloy said alloy being substantially gas-free at the glass-to-metal interface during glass sealing operations at temperatures in the range of 1700° F. to 2200° F. and having the composition carbon 0.07 per cent maximum, manganese 0.50 to 2.00 per cent, silicon 0.25 to 1.00 per cent, nickel 47 to 50 per cent, columbium 0.50 to 2.00 per cent and the balance iron and incidental impurities.

CARL B. POST.
WARREN S. EBERLY.

REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
2,162,489	Mathies et al.	June 13, 1939
2,239,423	Hull	Apr. 22, 1941
2,266,481	Talbot	Dec. 16, 1941

FOREIGN PATENTS

Number	Country	Date
679,794	Germany	Aug. 15, 1937