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(54) **NICKEL-BASE ALLOY FOR THE ELECTRO-WELDING OF NICKEL ALLOYS AND STEELS, WELDING WIRE AND USE**

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(57) **ABSTRACT**

The alloy contains, by weight, less than 0.05% of carbon, from 0.015% to 0.5% of silicon, from 0.4% to 1.4% of manganese, from 28% to 31.5% of chromium, from 8% to 12% of iron, from 2% to 7% of molybdenum, from 0% to 0.75% of aluminium, from 0% to 0.8% of titanium, from 0.6% to 2% in total of niobium and tantalum, the ratio of percentages of niobium plus tantalum and of silicon being at least 4, less than 0.04% of nitrogen, from 0.0008% to 0.0120% of zirconium, from 0.0010% to 0.0100% of boron, less than 0.01% of sulphur, less than 0.020% of phosphorus, less than 0.30% of copper, less than 0.15% of cobalt and less than 0.10% of tungsten, the remainder of the alloy, with the exception of unavoidable impurities of which the total content is at most 0.5%, consisting of nickel. The alloy is used, in particular, for the production of wires for the electro-gas welding of units or components of nuclear reactors and, more particularly, of pressurised water-cooled nuclear reactors.

NICKEL-BASE ALLOY FOR THE ELECTRO-WELDING OF NICKEL ALLOYS AND STEELS, WELDING WIRE AND USE

TECHNICAL FIELD

[0001] The invention relates to a nickel-based alloy for the electro-welding of nickel alloys and steels, in particular non-alloyed or sparingly alloyed steels and stainless steels.

[0002] The invention also relates to wires and electrodes for the electro-welding of parts made of nickel alloy and/or steel, in particular in the field of the construction, assembly and repair of components of nuclear reactors.

BACKGROUND TO THE INVENTION

[0003] It is known to use chromium-containing nickel-based alloys for the production of certain components or units of nuclear reactors.

[0004] In particular, a nickel alloy containing approximately 15% of chromium, known as alloy 600, has been used for the production of units or components of pressurised water-cooled nuclear reactors.

[0005] To improve the corrosion resistance of the units or components of pressurised water-cooled nuclear reactors, there is a tendency to replace the alloy 600 containing approximately 15% of chromium by an alloy 690 containing approximately 30% of chromium and approximately 10% of iron.

[0006] Electro-welding wires or electrodes of nickel alloy of which the composition is adapted to the welding of the alloy 600 or the alloy 690 are used to produce welds on these nickel alloy units or components.

[0007] Table 1 below shows typical compositions of commercially available wires for the welding of the alloy 690 and for the welding of the alloy 600 (alloy 52 or alloy 82).

[0008] The first four columns of Table 1 show the compositions of alloy 52 wires known by the trade name Inconel 52 from the American company, Special Metals, produced on the basis of four separate castings and used to weld the alloy 690.

[0009] The last column of the table gives a typical analysis of an alloy 82 wire with the trade name Phyweld 82 made by Sprint Metal, for welding the alloy 600.

[0010] Alloy 52 wires or 82 may be used, in particular, for the inert gas electro-welding of the alloy 690 or the alloy 600.

TABLE 1

Analysis of wires for welding the alloys 600 and 690

Casting	Alloy 52 wire 1	Alloy 52 wire 2	Alloy 52 wire 3	Alloy 52 wire 4	Comparison example CF 52 wire	Comparison example CF 52 wire	Alloy 82 wire
C	0.022	0.020	0.020	0.020	0.022	0.020	0.030
S	0.001	0.001	0.001	0.001	0.002	0.001	0.002
P	0.004	0.004	0.003	0.004	<0.003	0.003	0.003
Si	0.150	0.140	0.140	0.170	0.020	0.03	0.170
Mn	0.25	0.24	0.25	0.25	0.88	0.92	3.04
Ni	61.13	60.46	60.40	59.13	58.20	60.10	70.54
Cr	29.00	28.97	28.91	28.94	30.93	30.13	20.99
Cu	0.010	0.010	0.010	0.010		0.03	0.005
Co	0.040	0.010	0.010	0.010			0.010
Mo	0.010	0.010	0.010	0.010	0.012	0.02	
Nb	0.021	0.010	0.010	0.010	0.918	0.93	2.287
Al	0.660	0.690	0.670	0.680	0.065	0.08	
Ti	0.560	0.580	0.560	0.530	0.193	0.22	0.200
Fe	8.14	8.86	9.03	10.25	9.12	8.50	2.72
Zr					0.0013	0.006	
B					0.0028	0.0040	
Nb/Si	0.14	0.07	0.00	0.00	45.90	31.0	13.45

[0011] The alloy 52 welding wires are used, in particular, in the nuclear field, to produce welds in zones of the nuclear reactor components in contact with the primary fluid, which is water at a high temperature (approximately 310° C.) and under high pressure (approximately 155 bars), in the case of pressurised water-cooled nuclear reactors.

[0012] Alloy 52 is used for the homogeneous welding of alloy 690 parts and for producing heterogeneous welds. These heterogeneous welds may be, for example, welds on an alloy 600 containing 15% of chromium in solid form or deposited on a base metal, wherein the chromium content of the deposited metal may be from 15% to 20%.

[0013] Another application for alloy 52 in heterogeneous welding is the coating of sparingly alloyed steels such as the steels 16MND5, 18MND5 or 20MND5 or the welding of sparingly alloyed steels to austenitic or austeno-ferritic stainless steels.

[0014] The alloy 52 may also be used to repair zones of nuclear reactor units or components consisting of various metals such as minimally alloyed steels (for example of the type 18MND5 according to the French standard), stainless steels of the type 304L (for example in solid form), of the type 308L (in deposited form) or else 316L (in solid or deposited form) according to the American standards. These zones may comprise a plurality of these materials on which heterogeneous welds made of alloy 52 are produced.

[0015] Certain defects, mainly in the form of small fissures, have been demonstrated when using commercial alloys 52 such as the alloys 52 from Special Metals.

[0016] In particular, when the molten welding wire is deposited on a layer consisting of a nickel alloy deposited by welding, fissuring in heat was observed and may be due to one of the following phenomena: solidification, liquation, reassignment or else lack of ductility in heat. It was noted that a single type or a plurality of types of fissure may be found in a weld. Small-dimension fissures formed in these conditions will be called type 1 fissures.

[0017] Tests were carried out on welding wires of different compositions under variable welding conditions, in particular by fusing these wires on various base metals such as: nickel alloys as mentioned above and stainless steels, in the form of solid metals or of layers pre-deposited by welding.

[0018] During these tests, it was demonstrated that commercially available wires, in particular alloys 52 for the welding of nickel 690 alloys, gave poor results when they were deposited on nickel alloys containing 15% or 30% of chromium or else stainless steels deposited by welding, in the form of a coating of sparingly alloyed steel components.

[0019] In addition to small type 1 fissures, other larger fissures, which will be called type 2 heat fissures, were observed in certain cases.

[0020] Type 2 fissures were observed, in particular, in the zones of pronounced dilution of the welding alloy (in the metal deposited during the first welding passes or in the region of the parts to be joined) or more generally in the case of the welding of stainless steels.

[0021] Commercial welding wires of which the grades had been modified to improve the resistance to fissuring in heat and the resistance to oxidation were also used during these tests.

[0022] The modified composition of these commercially available wires is given in columns 5 and 6 of Table 1. Grades modified to improve the resistance to fissuring in heat and defects due to oxidation have a substantially higher niobium content (higher than 0.9%) and substantially lower aluminium and titanium contents than unmodified commercial grades.

[0023] In these improved grades, the niobium to silicon ratio is high (higher than 30 or even 45). Finally, these grades contain boron and zirconium as complementary elements.

[0024] It has been found that the improved commercially available alloys gave good results in the diluted zones during the welding of nickel alloys containing 15% or 30% of chromium, these zones being virtually exempt from fissuring in heat, but poor results in diluted zones in the case of welding on stainless steels, the type 2 heat fissures being detected in these diluted zones.

[0025] The tests carried out showed that there are no commercially available wires for producing homogeneous or heterogeneous electro-welds, which are exempt from fissuring and oxidation, on nickel alloys and on steels.

SUMMARY OF THE INVENTION

[0026] The object of the invention is therefore to propose a nickel-based alloy for the electro-welding of alloys of nickel steels, in particular stainless steels, which allow the production of homogeneous or heterogeneous welds on these materials, which are exempt from fissuring in heat and from traces of oxidation.

[0027] Accordingly, the alloy according to the invention contains, by weight, less than 0.05% of carbon, from 0.015% to 0.5% of silicon, from 0.4% to 1.4% of manganese, from 28% to 31.5% of chromium, from 8% to 12% of iron, from 2% to 7% of molybdenum, from 0% to 0.8% of titanium, from 0.6% to 2% in total of niobium and tantalum, the ratio of percentages of niobium plus tantalum and of silicon being at least 4, from 0% to 0.75% of aluminium, less than 0.04% of nitrogen, from 0.0008% to 0.0120% of zirconium, from 0.0010% to 0.010% of boron, less than 0.01% of sulphur, less than 0.020% of phosphorus, less than 0.30% of copper, less than 0.15% of cobalt and less than 0.10% of tungsten, the remainder of the alloy, with the exception of unavoidable impurities of which the total content is at most 0.5%, consisting of nickel.

[0028] The invention also relates to an alloy preferably containing less than 0.20% silicon, about 4% of molybdenum, 0.006% of zirconium and 0.004% of boron.

[0029] The invention also relates to a welding wire for the electro-gas welding of nickel-base alloy according to the invention.

[0030] The invention additionally relates to the application of the alloy and of the electro-welding wire for the welding of units or components of nuclear reactors, in particular pressurised water-cooled nuclear reactors, for the production of assemblies during the construction of nuclear reactors, the coating of components by metal deposition and for making repairs, wherein these welding operations may be operations for the homogeneous or heterogeneous welding of any nickel alloy or steel component.

DESCRIPTION OF PREFERRED EMBODIMENTS

[0031] To assist understanding of the invention, a plurality of grades of alloy according to the invention used for the production of wires will be described by way of examples which have been used during homogeneous and heterogeneous welding tests on nickel alloys and stainless steels.

[0032] In Table 2, column 1 shows the minimum contents of the various elements of the alloy, column 2 the maximum contents of these elements and column 3 the preferred contents.

[0033] Columns 4 and 5 of the table give alloy compositions according to two embodiments which will be described hereinafter.

[0034] The effect of the various elements of the alloy and the reasons for the claimed ranges or the limitations of these elements will be explained hereinafter.

TABLE 2

Elements in the alloy	Mini	Maxi	Preferred	Example 1	Example 2
C	/	0.05		0.020	0.015
S	/	0.010		0.001	0.001
P	/	0.020		0.003	0.003

TABLE 2-continued

Elements in the alloy	Mini	Maxi	Preferred	Example 1	Example 2
Si	0.015	0.5	<0.20	0.025	0.15
Mn	0.4	1.4		1	1
Ni	balance	balance	balance	balance	balance
Cr	28	31.5		30.0	29.0
Cu	/	0.30		0.020	0.020
Co	/	0.15		0.01	0.01
Mo	2	7	4	4.00	5.50
Nb (+Ta)	0.6	2		0.80	1.2
Al	0	0.75	0.15	0.07	0.25
Ti	0	0.8	0.30	0.35	0.20
Zr	0.0008	0.012	0.006	0.0015	0.006
B	0.001	0.010	0.004	0.003	0.004
N	/	0.040		0.01	0.01
W	/	0.10		0.01	0.01
Fe	8	12		9.00	8.5
Nb/Si	4			32	8

[0035] Carbon, Sulphur, Phosphorus

[0036] These elements are residual elements of which the contents have to be limited as far as possible and, in any case, fixed below 0.05% in the case of carbon, 0.010% in the case of sulphur and 0.020% in the case of phosphorus. Depending on the methods of production and the starting products for preparation of the alloys, the effective contents of carbon, sulphur and phosphorus may be substantially lower than the given maximum limits. As shown in columns 4 and 5 relating the examples 1 and 2, the effective carbon, sulphur and phosphorus contents of the castings carried out are substantially lower than the maximum values given hereinbefore.

[0037] Silicon

[0038] Silicon is an element which is always present in the alloy but of which the content is to be limited to a low value, preferably lower than 0.20% for example 0.12% or 0.15%). In all cases, this content must be lower than 0.5% to limit fissuration of the welding metal in heat. However, the silicon must be present in a content of at least 0.015% to obtain good weldability on account of the fact that it influences the wetting and the viscosity of the bath during welding.

[0039] It will be seen hereinafter, with respect to niobium, that the important parameter for resistance to fissuration in heat is the ratio of the percentage by weight of niobium and silicon.

[0040] Manganese

[0041] The manganese must be at least 0.4% to achieve satisfactory conditions for the production of the alloy in the presence of sulphur (limited to the value of 0.01% mentioned hereinafter).

[0042] The manganese contributes to the resistance to fissuration in heat, but this effect is rapidly saturated as a function of the manganese content, and a manganese content limited to 1.4% leads to satisfactory results.

[0043] Chromium

[0044] The chromium must be close to the percentage of chromium in the alloy 690, and the composition range of 28% to 31.5%, which is also that of the alloys 52, has been found to be satisfactory in the case of homogeneous and heterogeneous welds employing the alloy 690 or stainless steels. This level of chromium is required for achieving good anti-corrosion behaviour in a primary PWR medium.

[0045] Copper

[0046] Copper must be strictly limited to less than 0.30% to avoid a deterioration in the properties of the alloy.

[0047] Cobalt

[0048] The cobalt must necessarily be limited to a value below 0.15%. In fact, this element, which is activated in the presence of radiation in a nuclear reactor, must be avoided as far as possible in any application to the construction or repair of nuclear reactors.

[0049] Molybdenum

[0050] Molybdenum is a particularly important element in the production of the alloys according to the invention, and this represents a significant difference relative to formerly known alloys (see Table 1) which only have very low molybdenum contents.

[0051] Tests carried out by the Applicant of the present patent application have shown that the molybdenum had a decisive influence on the resistance to fissuring of the metal deposited by fusion of a nickel alloy welding wire, in particular when the welding wire is deposited on stainless steels, for example steels containing 18% of chromium and 8% of nickel with or without an addition of molybdenum, in solid form or in the form of a deposit obtained by covered electrode or TIG electro-welding.

[0052] The tests carried out showed that:

- **[0053]** the formation of type 2 fissures is observed with low levels of molybdenum in the molten metal of the welding wire, typically lower than 0.5%, in particular when welding stainless steels,
- **[0054]** as the molybdenum content increases, for example to 1% in the molten metal, it is found that the fissuring resistance of the welding alloy is substantially improved if the alloy contains sufficient quantities of titanium (and/or of aluminium). A minimal titanium and/or aluminium content of 0.3% to 0.4%, with a molybdenum content of 1% in the alloy, limits the number of type 2 fissures in the welding metal to a low level, particularly in the case of the welding of stainless steels; these results suggest that the molybdenum, titanium and/or aluminium contents should be increased beyond these limits in order to limit or eliminate fissuring,
- **[0055]** when the molybdenum content increases to at least 2% in the molten metal, the tests show that the type 2 fissures completely disappear and that the titanium and/or the aluminium have less influence on the fissuring resistance than in the case of a molybdenum of about 1%.

[0056] The molybdenum contents have been fixed in the alloy welding wire or rod according to the invention while agreeing that, in the region of the defects, the dilution is high and may reach 50% but does not exceed much this limit which has been considered as respected in all the work forming the basis of the present patent application and corresponds to normal welding conditions.

[0057] As a result of the tests, it has been possible to establish that the molybdenum content has to be at least 2% in order to obtain, in all cases of use in welding, very high resistance to fissuring and, in particular, a total disappearance of type 2 fissures, while limiting the total titanium and aluminium content to a level at which oxidation of the welding metal is avoided.

[0058] A molybdenum content higher than 7% is possible, but not essential, in so far as the influence of the molybdenum on the fissuring resistance is saturated at a value of approximately 7%. A content higher than 7% increases the price of the alloy and may undesirably modify the properties of the welding metal.

[0059] The molybdenum should preferably be in the region of 4%.

[0060] Aluminium and Titanium

[0061] It is possible to provide a certain proportion of titanium and aluminium in the welding metal while however limiting the proportion of aluminium to a value at which the undesirable oxidation effects of the molten bath are avoided during welding.

[0062] The titanium may be present in the alloy in a proportion of between 0% and 0.8% and, for example, close to 0.30%. A titanium content higher than 0.8% is not desired.

[0063] The aluminium may be present in the alloy in a proportion of between 0% and 0.75% and, for example, in the region of 0.15%. An Aluminium content higher than 0.75% is not desired.

[0064] Zirconium and Boron

[0065] When combined, these elements have a favourable influence on the fissuring resistance owing to the ductility dip cracking. However, these elements alone are not sufficient to solve the fissuring problems which the alloy according to the invention rectifies. Furthermore, zirconium, like aluminium, affects the oxidation of the molten bath during welding. The zirconium and boron must therefore be present in the alloy but in limited quantities.

[0066] The zirconium must be present in the alloy according to the invention in a proportion of between 0.0008% and 0.012% and preferably in a proportion of approximately 0.006%.

[0067] In relation to these proportions of zirconium, the boron must be between 0.001% and 0.010% and, preferably, in the region of 0.004%.

[0068] Niobium and Tantalum

[0069] Niobium affects the resistance to fissuring in heat. To avoid increasing the risks of fissuring in heat and undesirably modifying the characteristics of the deposited metal, this element must not be present in an excessively large quantity.

[0070] As a result, the proportion of niobium must be at least 0.6% to obtain the desirable effects of resistance to fissuring in heat and at most 2%.

[0071] The proportion of niobium within this range must be fixed at a value which is such that the ratio of the percentage of niobium to the percentage of silicon is higher than 4 to obtain a satisfactory effect on the resistance to fissuring in heat.

[0072] Iron

[0073] As in commercial alloys, the iron is fixed at a content of between 8% and 12% for providing a good structural stability, by avoiding any risk of formation of an ordered embritteling NiCr phase.

[0074] Nitrogen

[0075] Nitrogen, which is a residual element, is not necessary in the alloy. The nitrogen will be limited, in all cases, to a value of less than 0.040%.

[0076] Tungsten

[0077] Tungsten is an element which is not desired in the alloy, this residual element being limited to 0.10% in any case to avoid undesirable modification of the properties of the welding metal.

[0078] The alloy may contain small proportions of other residual elements; these elements may be, for example, tin, vanadium, lead, cadmium, magnesium, zinc, antimony, tellurium, calcium or cerium. These elements, which are in a very small quantity in the alloy, are in a total proportion with the other residual elements considered above (carbon, sulphur, phosphorus, copper, cobalt, nitrogen and tungsten) of less than 0.5% by weight.

[0079] Nickel

[0080] As a base alloy, it makes up the remainder of the composition to 100%.

[0081] The compositions of two welding alloys according to the invention are shown in Table 2 under columns 5 and 6 (example 1 and example 2).

EXAMPLE 1

[0082] In the case of example 1, the molybdenum content is at the optimum value (4%). The aluminium content is in the region of the lower limit of the range of aluminium and the titanium content has a value close to the typical value of 0.30%.

[0083] The silicon content of the alloy is low (0.025%) and is clearly below the preferred upper limit. Although the niobium content is only 0.80%, the niobium/silicon ratio is high and is approximately the same as in commercial alloys of the improved type shown in Table 1 (32). The value of this ratio is much higher than the lower limit imposed. The zirconium and boron contents lie towards the bottom of the claimed range.

EXAMPLE 2

[0084] In the case of example 2, the molybdenum content is higher than the mean content considered as preferred (4%). The aluminium content which is substantially higher than in the case of example 1 is fixed above the typical value of 0.15% and the titanium content is lower than the typical value, the entirety of the aluminium and titanium representing a percentage by weight which is substantially identical in the case of example 1 and in the case of example 2.

[0085] The boron content is higher than in the case of example 1 and corresponds to the preferred values.

[0086] The silicon content is substantially higher than in the case of example 1. The niobium content is also slightly higher than in the case of example 1. Owing to the presence of a fairly large quantity of silicon, the niobium to silicon ratio is substantially lower in the case of example 1.

[0087] However, this ratio is twice as high as the minimum required value.

[0088] Welding wires in the two grades corresponding to examples 1 and 2 were produced. The welding wires were used for diverse homogeneous or heterogeneous welding of nickel alloys containing 30% and 15% of chromium and stainless steels.

[0089] The total absence of type 2 fissures in the deposited metal was observed, even in the diluted zones of the weld.

[0090] The deposited metal is also virtually exempt from type 1 fissures in all cases.

[0091] No trace of oxidation which could lead to deterioration of the deposited metal was found.

[0092] It has never been possible to obtain results of this type in the case of alloy wires according to the prior art.

[0093] Considering the comparison examples in columns 5 and 6 of Table 1, it can be seen that the comparison alloy in column 5 (CF 52) has a silicon content comparable to that of example 1 according to the invention and a slightly higher niobium content, the niobium to silicon ratio being 50% higher than the niobium to silicon ratio of example 1. However, this alloy according to the prior art contains only a very small proportion of molybdenum (0.012%) whereas the alloys according to the invention contain more than 2% and generally 4% or more of molybdenum. Despite a higher niobium to silicon ratio and similar contents of aluminium and titanium, the alloy CF 52 does not result in a resistance to fissuring which is comparable to that of the alloys according to the invention.

[0094] In the case of the second comparison example (alloy 52 M) in column 6 of Table 2, the silicon and niobium contents and the niobium to silicon ratio are similar to those of the alloy of example 1. The aluminium and the titanium, on the other hand, are limited to values comparable to those of the examples according to the invention. The zirconium and boron contents of the comparison alloys are, moreover, similar to those of the alloys of examples 1 and 2 according to the invention respectively.

[0095] It is perfectly clear that a quasi-absence of molybdenum (0.02%) in the second comparison alloy explains the differences in welding behaviour and the good results achieved with the examples of alloy according to the invention and, in particular, example 1.

[0096] A comparison of the examples according to the invention and the examples of alloys according to the prior art therefore shows that a welding alloy having a molybdenum content of approximately 4% or slightly higher, an adequate niobium content to obtain a niobium to silicon ratio substantially higher than 4 and moderate aluminium and titanium contents solves the problems of welding nickel alloys containing approximately 15% and 30% of chromium to stainless steels.

[0097] The alloy according to the invention leads to electro-gas welding wires for the perfect homogeneous or heterogeneous welding of nickel alloys and stainless steels for the construction and repair of nuclear reactor components.

[0098] The invention is not strictly limited to the described embodiments.

[0099] The contents of the various elements of the alloys for electro-welding in the considered applications may be adapted within the claimed ranges to optimise the properties of the welding metal and the welding conditions.

[0100] The alloy according to the invention may be used not only in the form of electro-gas welding wires or rods but also in other forms, for example in the form of coated electrodes.

[0101] Although the alloy is intended, in particular, for applications in the field of the construction and repair of nuclear reactors, its use in other industries may be considered.

1. A nickel-based alloy for the electro-welding of nickel alloys and steels, the alloy comprising, by weight: less than 0.05% carbon, 0.015% to 0.5% silicon, 0.4% to 1.4% manganese, 28% to 31.5% chromium, 8% to 12% iron, 2% to 7% molybdenum, 0.6% to 2% in total of niobium and tantalum, the ratio of percentages of niobium plus tantalum and of silicon being at least 4, less than 0.04% nitrogen, 0.0008% to 0.0120% zirconium, 0.0010% to 0.0100% boron, 0% to 0.75% aluminium, 0% to 0.8% titanium, less than 0.01% sulphur, less than 0.020% phosphorus, less than 0.30% copper, less than 0.15% cobalt and less than 0.10% tungsten, the remainder of the alloy containing nickel, with the exception of impurities having a content less than or equal to 0.5%.

2. An alloy according to claim 1, containing less than 0.20% silicon, about 4% molybdenum, 0.006% zirconium and 0.004% boron.

3. Welding wire for the electro-gas welding of nickel alloys and steel, produced from the alloy according to claim 1.

4. Welding wire for the electro-gas welding of nickel alloys and steel produced from the alloy according to claim 2.

5. Use of an alloy according claim 1, for the welding of components of a nuclear reactor during construction, assembly or repair of a component.

6. Use of an alloy according claim 2, for the welding of components of a nuclear reactor during construction, assembly or repair of a component.

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