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ALUMINUM BRONZE ALLOY HAVING IMPROVED RESISTANCE TO INTERGRANULAR OXIDATION BY THE ADDITION OF TIN

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5 Claims. (Cl. 75—154)

This invention relates to an aluminum bronze alloy having increased resistance to intergranular oxidation when subjected to stress in a hot water or steam atmosphere.

It is well known that almost all copper-base alloys, when stressed in certain corrosive media, are subject to a particular species of oxidation, generally termed intergranular oxidation by those skilled in the art. The term intergranular oxidation refers to the spontaneous cracking of a metal subjected to contact with a corrodent and to residual or applied stresses. This invention involves an improvement of the characteristics of aluminum bronze alloys which have been found to be subject to intergranular oxidation when stressed in media containing hot water and steam.

It has been found that the oxidation or fracture of an aluminum bronze alloy under the action of stress and a hot water or steam corrodent, follow paths which are intergranular in nature. It is believed that the reason for this intergranular type of attack is that aluminum atoms are in a higher concentration in the grain boundary than in the grains. This is probably because aluminum atoms have a negative elastic energy interaction with dilated grain boundary regions; therefore the larger aluminum atoms become segregated in the grain boundary and are subject to oxidation.

The problem of intergranular attack is particularly serious in alpha-phase aluminum bronze alloys which contain about 5% to 8% aluminum. The alpha-phase alloy has excellent working properties and can be readily fabricated into tube, sheet, plate, etc., in which form the alloy is often subjected to stress in a hot water or steam atmosphere.

The present invention is based on the discovery that the resistance of an alpha-phase aluminum bronze alloy to intergranular oxidation when subjected to a steam or hot water atmosphere can be increased by the addition of small amounts of tin to the alloy. This is particularly true where the amount of tin is maintained within a specific ratio to the aluminum content in the alloy.

More specifically, the tin content in the alloy should be in the ratio of about one part by weight of tin to 25 parts by weight of aluminum. It has been found that an aluminum bronze alloy containing the above aluminum to tin ratio is much more resistant to intergranular oxidation when stressed in hot water and steam because tin atoms, being larger than aluminum atoms, have a larger negative elastic energy interaction with dilated grain boundary regions than aluminum atoms, and therefore tend to replace aluminum atoms in the grain boundary. Aluminum bronze alloys containing tin atoms segregated at the grain boundaries probably have increased resistance to grain boundary oxidation because of the low heats of formation of tin oxides.

It has been found that it is necessary to make the improved alloy with a certain critical ratio of tin to the aluminum content. This is believed due to the additional theoretical consideration of the configurational entropy

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contributed by the aluminum and tin atoms. In effect, this means that the smaller the concentration of a particular solute atom, the more difficult it will be for it to segregate to a grain boundary. The tin atoms compete with aluminum atoms for grain boundary locations and the ability to create a grain boundary resistant to oxidation depends on holding the aluminum to tin content in certain critical ratios.

Tin has been used as a major and minor addition to copper and copper-base alloys in a great number of instances. However, the use of tin in alloying with copper in the previous art has not specified or contemplated the use in aluminum bronze alloys in a definite ratio to the aluminum content to substantially prevent intergranular attack when stressed in hot water and steam environments. For instance, tin additions have been made to brasses along with nickel, and the tin in this case combines with the nickel to form a compound which through proper heat treatment can be made to substantially harden the brass. In contrast, this invention relates to an alpha-phase aluminum bronze alloy, with greatly improved resistance to intergranular oxidation when stressed in hot water and steam media through the use of a specific aluminum to tin ratio. In this invention, the mechanical properties of the alpha-phase aluminum bronze alloy are maintained, and at the same time the resistance of the alloy to intergranular attack is increased.

According to the invention the general composition range, by weight, of the alloy is as follows:

Aluminum	5.0 to 8.0%.
Iron	1.6 to 2.9%.
Tin	In a weight ratio of 1 part tin to 11 to 32 parts of aluminum.
Copper	Balance.

The controlled preferred compositional range of the alloy is as follows, in weight percent:

Aluminum	6.00 to 6.75%.
Iron	2.00 to 2.40%.
Tin	In a weight ratio of 1 part tin to 24 to 27 parts of aluminum.
Copper	Balance.

A specific illustration of an alloy falling within the above ranges is as follows, in weight percent:

Aluminum	Percent
Iron	6.25
Tin	2.08
Copper	0.25
	91.42

In addition to the ratio between the aluminum and tin, it has been found that the most desirable properties are obtained in the alloy when the iron content is maintained at a 1 to 3 ratio to the aluminum content. The iron is employed as a grain stabilizer and also acts to increase the strength of the alloy, particularly at higher temperatures. The 1 to 3 ratio between the iron and aluminum provides the alloy with the most desirable grain refining characteristics and mechanical properties.

In addition, other metals such as nickel and manganese may be employed in the alloy in amounts up to about 0.25% to add to the mechanical properties of the alloy, if desired.

Age hardening metals such as silicon, beryllium and the like should be avoided in the present alloy. Nickel, if used alone in the aluminum-iron copper alloy of the invention, will not age harden, but if a combination of metals such as silicon and nickel is used, the combination will age harden when the alloy is heated during welding, forming, or the like to form nickel silicides. The

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nickel silicides are extremely brittle and tend to precipitate and gather at the grain boundaries. This tendency of the age hardening precipitate to gather at the grain boundary is believed to prevent the tin from gathering at the grain boundaries and in effect destroys the purpose of the tin addition. The age hardening precipitates can be removed by a solution quench from over 1600° F., but a heat treatment such as this is not possible with a finished or fabricated article because the dimensional tolerances of the article will be destroyed by the heat treatment. Therefore, age hardening constituents should be avoided in the present alloy.

The alloy is formed by initially preparing a pre-alloy of copper, aluminum and iron. This pre-alloy is then alloyed with additional copper and tin and cast. It is preferred not to add the tin to the pre-alloy because a portion of the tin would be lost by oxidation during melting and additional quantities of tin would have to be employed to hold the concentration of the same within the desired limits.

The present alloy can be made to provide a yield strength from 20,000 p. s. i. to 90,000 p. s. i., a tensile strength of 40,000 p. s. i. to 120,000 p. s. i. and an elongation in 2 inches of 1% to 60%, depending on the alloy composition, the method of fabrication employed and the heat treatment used. For example, an alloy having 6.25% aluminum, 2.10% iron, 0.25% tin and the balance copper, in the form of ½ inch thick plate, can be hot rolled and annealed to give a tensile strength of 85,000 p. s. i., a yield strength of 42,000 p. s. i., an elongation in 2 inches of 32% and a Brinell hardness of 160.

The same alloy in the form of cold drawn 1 inch diameter rod has a tensile strength of 90,000 p. s. i., a yield strength of 70,000 p. s. i., an elongation in 2 inches of 30% and a Brinell hardness of 185.

As an example of the alloy as a weld deposit, the composition consisted of 7.0% aluminum, 2.4% iron, 0.25% tin and the balance copper. A sample of this alloy, taken through a butt welded joint, had a tensile strength of 75,000 p. s. i., a yield strength of 35,000 p. s. i., an elongation in 2 inches of 25% and a Brinell hardness of 150.

The alloy has high resistance to intergranular fracture resulting from oxidation at the grain boundaries. For example, the alloy will resist intergranular fracture for at least 2000 hours when stressed from 20,000 to 50,000 p. s. i. in a steam or hot water atmosphere at temperatures from 180° F. to 500° F. In some cases the alloy has been stressed for 8000 hours under the above conditions without failure.

As an example of the increased resistance of the alloy to intergranular oxidation when stressed in a hot water or steam atmosphere, a specimen of an alloy containing 6.00% aluminum, 2.20% iron, 0.24% tin and the balance copper was stressed beyond the yield strength by deflection bending in a media consisting of a 10% aqueous solution of sodium hydroxide at a temperature of 350° F. The specimen did not fail in 1000 hours of testing.

A second specimen having a similar composition, but without tin, was stressed under identical conditions and failed after 100 hours of testing.

The present invention provides an aluminum bronze alloy that has greatly improved resistance to intergranular oxidation when stressed in steam and water atmospheres. The term "steam" is intended to include gaseous mixtures of steam and other vapors and the term "hot water" is intended to include heated aqueous solutions of salts, acids or bases and other materials containing water or moisture as an ingredient.

Various modes of carrying out the invention are contemplated as being within the scope of the following claims particularly pointing out and distinctly claiming the subject matter which is regarded as the invention.

I claim:

1. An aluminum bronze alloy having increased resistance to intergranular oxidation when subjected to stress in a hot water or steam atmosphere, consisting essentially by weight of: about 5.0% to 8.0% aluminum; about 1.6% to 2.9% iron; tin in a weight ratio of 1 part tin to 11 to 32 parts of aluminum; and the balance substantially copper.

2. An aluminum bronze alloy consisting essentially by weight of: about 6.00% to 6.75% aluminum; about 2.00% to 2.40% iron; tin in a weight ratio of 1 part tin to 24 to 27 parts of aluminum; and the balance substantially copper, said alloy characterized by resistance to intergranular fracture for a period of at least 2000 hours when subjected to a stress in the range of 20,000 to 50,000 p. s. i. and a hot water or steam atmosphere at a temperature of 180° F. to 500° F.

3. An aluminum bronze alloy having increased resistance to intergranular oxidation when subjected to stress in a hot water or steam atmosphere, consisting essentially of, by weight: about 5.0% to 8.0% aluminum; iron in a weight ratio of about 1 part iron to about 3 parts aluminum; tin in a weight ratio of about 1 part tin to 11 to 32 parts of aluminum; and the balance substantially copper, said tin segregating in the grain boundaries of the alloy and restricting the oxidation of said alloy at said grain boundaries.

4. An aluminum bronze alloy having increased resistance to intergranular oxidation when subjected to stress in a medium containing hot water or steam, consisting essentially of, by weight: about 6.00% to 6.75% aluminum; iron in a weight ratio of about 1 part iron to about 3 parts aluminum; tin in a weight ratio of about 1 part tin to 24 to 27 parts of aluminum; and the balance substantially copper.

5. An aluminum bronze alloy, consisting essentially of, by weight: 6.25% aluminum, 2.08% iron, 0.25% tin; and the balance copper, said alloy having a high concentration of tin in the grain boundaries to thereby increase the resistance of the alloy to intergranular oxidation when subjected to stress in a medium containing hot water or steam.

References Cited in the file of this patent

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U. S. DEPARTMENT OF COMMERCE
PATENT OFFICE
CERTIFICATE OF CORRECTION

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It is hereby certified that error appears in the printed specification of the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 2, line 4, for "complete" read --compete--.

Signed and sealed this 27th day of May 1958.

(SEAL)

Attest:

KARL H. AXLINE

Attesting Officer

ROBERT C. WATSON
Commissioner of Patents