

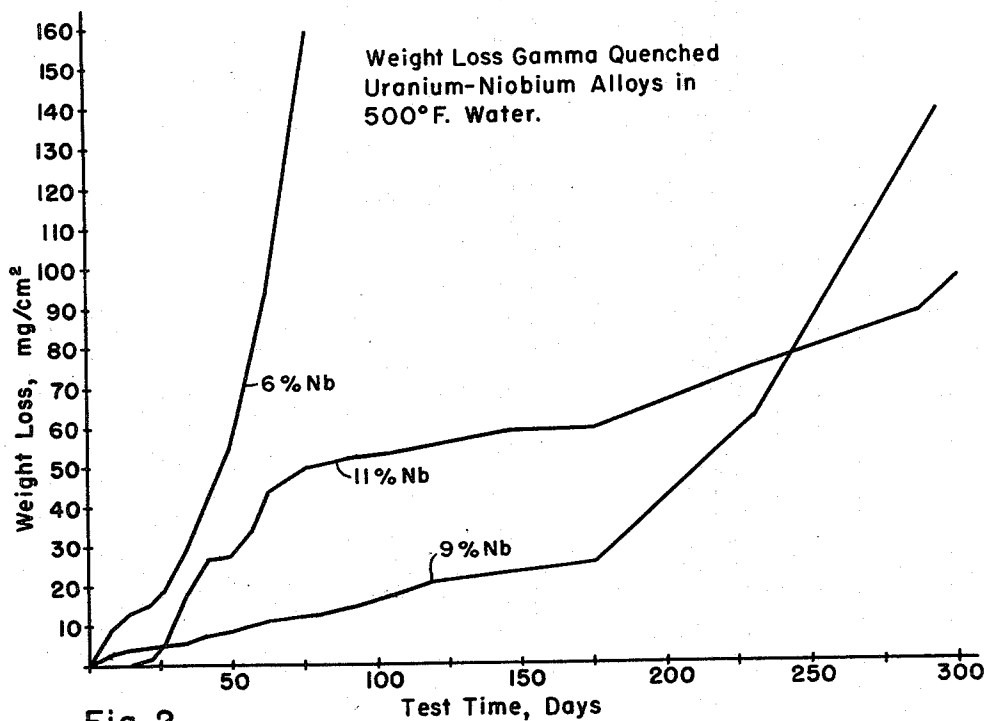
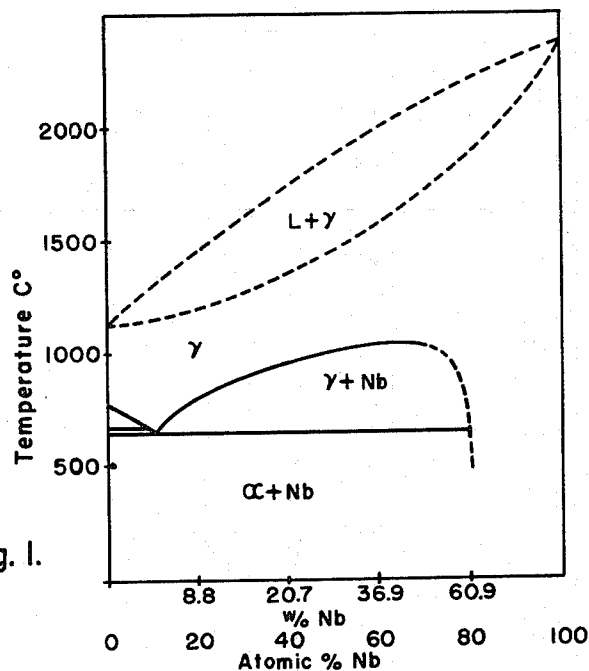
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2,914,433

HEAT TREATED U-Nb ALLOYS

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HEAT TREATED U-Nb ALLOYS

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3 Claims. (Cl. 148—11.5)

This invention relates to binary alloys of niobium and uranium and members prepared therefrom, particularly for use as fuel elements in nuclear reactors.

The use of uranium members as fuel elements in certain types of nuclear reactors employing water as a moderating medium and for other purposes results in unsatisfactory operation because of the high rate of corrosion of the uranium when in contact with hot water. In many cases, uranium members of substantial thickness will disintegrate completely in less than one day when in contact with hot water at a temperature of 600° F. While it has been proposed to apply a protective cladding material to the uranium fuel elements, there are substantial possibilities for voids, fissures, cracks and the like to be present which will permit the hot water or steam to reach the uranium and thereby cause corrosion, swelling and other undesirable results which may cause improper function of the reactor.

While it has been proposed to alloy uranium with various metals in order to reduce corrosion resistance when in contact with water at elevated temperatures, it has been discovered that many alloying elements do not improve the corrosion resistance, and, in some cases, they may even accelerate corrosion.

The object of this invention is to provide members suitable for use as fuel elements in a nuclear reactor comprising an alloy composed of from 7% to 20% by weight of niobium and the balance being uranium, either natural or enriched, the alloy being substantially entirely in the gamma phase.

Other objects of the invention will, in part, be obvious and will, in part, appear hereinafter. For a better understanding of the nature and objects of the invention, reference should be had to the following detailed description and drawing, in which:

Figure 1 is a phase diagram of uranium-niobium alloys; and

Fig. 2 is a graph plotting corrosion resistance for three different uranium-niobium alloys in 500° F. water.

Referring to Fig. 1 of the drawing, there is illustrated a phase diagram of the alloys of niobium and uranium. It will be noted that the solid alloys at a temperature of about 1000° C. are essentially all of the gamma phase structure. However, on being cooled to a temperature of below 625° C., the alloys tend to transform into the alpha phase of uranium in which niobium has substantially no solid solubility, the rate of transformation depending on the niobium content. The alpha uranium is characterized by poor corrosion resistance when in contact with hot water, for example, at 600° F. The pres-

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ence of particles of undissolved niobium in alpha uranium confers no improved corrosion resistance and may cause accelerated corrosion.

We have found that alloys of uranium containing from 7% to 20% by weight of niobium can be heated to a temperature in the gamma field of the phase diagram and quenched from this temperature, for instance approximately 900° C., at which they are essentially all of the gamma phase, to room temperature where they will retain the gamma structure. Alloys having 6% and less of niobium transform rapidly from the gamma to the alpha phase. Such transformation in the low niobium content alloys is initiated rapidly and complete transformation occurs more rapidly as the alloy is heated to temperatures above 300° C. By contrast, alloys containing 7% to 20% of niobium in the gamma phase have been heated for many days at temperatures of up to 650° F. (343° C.) without showing any appreciable amount of transformation to the alpha phase.

Members prepared from alloys composed of from 7% to 20% niobium and the balance being uranium, when quenched from a temperature of approximately 900° C. to room temperature to retain the gamma phase, exhibit high corrosion resistance to hot water at elevated temperatures. We prepared a series of binary uranium alloys comprising (a) 9% by weight of niobium, (b) 11½% by weight of niobium, and (c) 20% by weight of niobium, which alloys had been heated for 24 hours at 900° C. and then water quenched to room temperature. Specimens of these alloys were tested in an autoclave filled with hot water at temperatures of 500° F., 575° F. and 650° F. Our tests showed that the average corrosion rate for all the specimens in 500° F. water was 0.008 milligram per square centimeter per hour. In 575° F. water, the average corrosion rate was 0.02 milligram per square centimeter per hour. When tested in 650° F. water, the average corrosion rate of the three alloys was 0.045 milligram per square centimeter per hour. By comparison, alloys containing from 2% to 4% niobium prepared by annealing and quenching under the same conditions disintegrated completely in a matter of hours in hot water under each of these test conditions.

The alloys of the present invention may be prepared by melting the desired proportions of niobium and uranium in an induction furnace comprising a graphite crucible coated with a wash of zirconium oxide. The uranium may comprise natural uranium containing not over a few hundredths of 1% of impurities. The uranium may be entirely natural uranium or natural uranium that has been enriched with, for example, 10% of uranium 235. The molten alloy is poured into a graphite crucible to produce an ingot. In some instances the melt may be cast within a precision mold into the desired final form of member. We have found it ordinarily preferable to hot work the cast ingot by forging, extrusion, rolling and the like into the desired shape of member. The ingot may be heated in a salt bath to a temperature of from 1700° to 2100° F., and will then be readily extrudable into strips, bars or other desired structures. The alloy may be hot forged when heated to temperatures of 1700° F. In some instances, we have secured ingots of improved homogeneity and structure by employing the original cast ingot as the consumable electrode in an arc furnace. The arc furnace may be operated as set forth

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in copending application, Serial No. 367,524, assigned to the assignee of the present invention. The arc cast ingot may be readily extruded, rolled or forged to produce members of suitable size and shape.

The following examples are illustrative of the practice of the invention:

Example I

Into a crucible of an induction furnace there was placed 9 parts by weight of niobium and 91 parts by weight of natural uranium having the following impurities:

Element:	P.p.m.
Carbon -----	26 to 36
Zirconium -----	50 maximum
Nitrogen -----	20 maximum
Iron -----	40 to 55
Silicon -----	36 to 46
Nickel -----	10 to 12
Manganese -----	5 to 7
Chromium -----	4 to 6
Magnesium -----	3 to 5
Other elements -----	Less than 2

The alloy after having been melted was then poured into a graphite mold and cast into an ingot of a diameter of 2.4 inches. The ingot was then extruded into a bar. Specimens of a thickness of $\frac{1}{4}$ inch having a surface area of 5 square centimeters were cut from the extrusion. The members were homogenized by annealing at 900° C. for 24 hours and quenched in water to retain the gamma phase throughout. The specimens were placed in an autoclave containing water at 500° F. and tested for a prolonged period of time of 275 days. For the first 150 days, the total corrosion on the specimens comprised 22 milligrams per square centimeter. The corrosion rate increased thereafter so that at the end of 275 days the total weight loss comprised 135 milligrams per square centimeter.

Example II

Members comprising 11½% by weight of niobium and the balance being natural uranium were prepared by the process of Example I. The annealed and quenched specimens having substantially all gamma phase were placed in an autoclave and tested in contact with water at 500° F. for 300 days. The total corrosion of the specimens at the end of 275 days was approximately 88 milligrams per square centimeter which was somewhat less than that of the alloy of Example I. However, the initial corrosion rate was somewhat greater for the alloy of this example than was exhibited by the 9% niobium alloy.

Example III

Specimens were prepared from a 6% niobium-uranium alloy following the procedure of Example I. The specimens of the alloy were placed in an autoclave containing water at 500° F. The specimens were found to corrode at a much higher rate than the specimens of Examples I and II. Thus at the end of 75 days, the corrosion had caused a weight loss of 155 milligrams per square centimeter and complete disintegration and failure occurred soon thereafter.

The data obtained in the tests set forth under Examples I and II, along with the 6% niobium specimens, are plotted in Fig. 2 of the drawing. It will be observed that the niobium alloys of the present invention have a much greater resistance to corrosion as exemplified by the weight loss in hot water than alloys containing 6% of niobium. Alloys of uranium containing less than 6% niobium would have been completely disintegrated in a matter of a few days at most.

We have discovered outstanding corrosion resistance is exhibited by uranium alloys containing from 8% to 12% by weight of niobium.

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For use as fuel elements in nuclear reactors, it is ordinarily desirable to employ alloys containing the maximum proportion of uranium and the minimum amount of the alloying element, such as niobium. Consequently, alloys containing 8% to 12% of niobium will be preferred to the alloys containing, for example, 18% to 20% niobium.

We have secured good results with respect to corrosion resistance from alloys comprising 16% niobium and 20% niobium, the balance being natural uranium in each case. The gamma quenched alloys when placed in contact with water at 650° F. withstood more than 56 days in each case before complete corrosion failure took place.

It is desirable in all cases in preparing members from alloys comprising from 7% to 20% by weight of niobium, the balance being uranium, to homogenize the members at temperatures in the gamma field, ordinarily we use temperatures above 900° C., for at least several hours and to quench the members to retain the gamma phase at room temperature and under conditions in reactors. Water quenching has given good results. Cooling with inert gas or other means to reduce the temperature of the alloy from 900° C. to room temperature in an hour or two will also maintain the gamma phase.

It will be understood that the alloys of the present invention may be clad with protective coatings of materials such, for example, as the zirconium alloy set forth in copending application, Serial No. 416,396, now Patent No. 2,772,964 assigned to the assignee of the present invention.

It will be understood that fuel elements prepared from the alloys of the present invention when employed in nuclear reactors embodying water for cooling, moderating and for extracting the heat therefrom, should be operated at temperatures such that the fuel elements do not exceed a temperature of approximately 350° C. for any appreciable length of time.

Upon being tested by irradiation, specimens of the gamma phase alloys of this invention showed quite small isotropic changes in shape as compared to low niobium alloys which changed in shape irregularly and exhibited pronounced dimensional changes in certain directions.

It will be understood that the above description and drawing are only exemplary and not in limitation of the invention.

We claim as our invention:

1. A member suitable for use as a fuel element in a nuclear reactor comprising an alloy wrought to the shape of the member, the alloy composed of from 7% to 20% by weight of niobium and the balance being uranium, the alloy being substantially entirely in the gamma phase, the alloy having been produced by working an ingot of the alloy into the member, homogenizing the member by annealing it at a temperature in the gamma phase field and quenching the member to retain the gamma phase structure of the alloy.

2. A member suitable for use as a fuel element in a nuclear reactor comprising an alloy wrought to the shape of the member, the alloy composed of from 8% to 12% by weight of niobium and the balance being uranium, the alloy being substantially entirely in the gamma phase, the alloy having been produced by working an ingot of the alloy into the member, homogenizing the member by annealing it at a temperature in the gamma phase field and quenching the member to retain the gamma phase structure of the alloy.

3. In the method of preparing a member suitable for use as a fuel element in a nuclear reactor comprising an alloy composed of from 7% to 20% by weight of niobium and the balance being uranium, the steps comprising working an ingot of the alloy into the member, homogenizing the member by annealing at a temperature in the gamma phase field and quenching the member to retain the gamma phase structure of the alloy.

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