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RADIOACTIVE ALLOY

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3 Claims. (Cl. 75-170)

This application is a division of my co-pending application Serial No. 404,040, filed July 25, 1941, which is, in turn, a division of my application Serial No. 348,390, filed July 30, 1940, and a continuation in part of my application Serial No. 312,441, filed January 4, 1940.

This invention relates to radioactive alloys and relates especially to a radioactive alloy comprising a small but definite amount of a strongly ionizing radioactive metal.

Heretofore, radioactive alloys have not been commercially produced. A commercial use of radioactive alloys has only recently been suggested, making the development of a practical and effective alloy desirable.

A primary object of the present invention is to provide a practical and effective radioactive alloy.

Another object is to provide a radioactive alloy containing a small but definite proportion of a strongly ionizing radioactive metal.

Another object is to provide a practical polonium alloy.

Further objects will be manifest in the description of the invention, which follows.

Broadly, the invention includes a new radioactive alloy, particularly one which comprises a relatively small proportion of a strongly ionizing radioactive metal. The term "strongly ionizing radioactive metal" may be defined for the purposes of this invention as a radioactive metal which emits strong alpha rays at a practical rate and has a half life sufficiently long to allow its practical utilization. The following naturally occurring substances are examples of strongly ionizing radioactive metals contemplated by the invention: polonium, radium, ionium, uranium II, protoactinium, radioactinium, actinium X, radiothorium and thorium X. Equivalents of the named naturally occurring substances, such as a metal having a strongly induced radioactivity of the alpha-emission type, are also contemplated for use in the invention.

The new radioactive alloy preferably contains a minor proportion of a strongly ionizing radioactive metal and a larger proportion of a non-radioactive metal from the group of elements having atomic numbers in the range of 22 to 28, inclusive. That is, the radioactive alloy contains a smaller proportion of the radioactive metal and a larger proportion of titanium, vanadium, chromium, manganese, iron, cobalt, nickel, or a mixture of these metals. A preferred embodiment of the invention is a radioactive alloy comprising polonium and nickel suitable for spark plug electrodes. This embodiment is, in effect, a conventional spark plug electrode alloy containing nickel 90 to 97 per cent, manganese 2 to 6 per cent and silicon 0.75 to 1.25 per cent, but containing additionally a small amount of the radioactive metal distributed therein.

In general, radioactive metals are very expensive

because they are extracted with great difficulty from their ores, which are quite rare. However, it has been discovered that an alloy containing an exceedingly small proportion of a strongly ionizing radioactive metal possesses radioactive properties adequate for many purposes.

All radioactive materials give off one or more of three types of radiation called alpha, beta and gamma rays. The alpha rays, which are rapidly moving, doubly charged particles, are much more efficient for certain practical purposes, such as for ionizing gases, than either the beta rays (fast electrons) or gamma rays (electromagnetic radiation of extremely high frequency). Furthermore, the alpha rays are not dangerous to human beings, whereas gamma rays may be exceedingly harmful and require extreme safety precautions. The beta rays are also dangerous to some extent. Hence, it is quite apparent that a radioactive material emitting only alpha rays is much to be preferred for certain commercial purposes.

The relations between polonium (radium F), radium D, radium, and uranium are set forth in the following Table I, showing the uranium-radium radioactive series and adapted from Landolt-Börnstein Physikalisch-Chemische Tabellen, fifth edition, published by Julius Springer, Berlin.

Table I

Element	Atomic		Half life	Strong rays	Weak rays
	Wt.	No.			
Uranium I.....	238	92	4.4×10 ⁹ yrs.	Alpha.....	Gamma.
Uranium X ₁	234	90	24.5 days.....	Beta.....	
Uranium X ₂	234	91	1.17 min.....	Beta.....	
Uranium Z.....	234	91	6.7 hrs.....	Beta.....	
Uranium II.....	234	92	1.7×10 ⁸ yrs.	Alpha.....	
Ionium.....	230	90	85,000 yrs.....	Alpha.....	Beta and gamma.
Radium.....	226	88	1,690 yrs.....	Alpha.....	
Radon.....	222	86	3.83 days.....	Alpha.....	Gamma.
Radium A.....	218	84	3.05 min.....	Alpha.....	
Radium B.....	214	82	26.8 min.....	Beta.....	
Radium C.....	214	83	19.7 min.....	Beta and gamma	
Radium C'.....	214	84	0.0002 sec.....	Alpha.....	Beta and gamma.
Radium C''.....	210	81	1.3 min.....	Beta.....	
Radium D.....	210	82	22.3 yrs.....	Alpha.....	
Radium E.....	210	83	5.0 days.....	Beta.....	Gamma.
Radium F (polonium).....	210	84	138 days.....	Alpha.....	
Radium G (lead).....	206	82	Stable.....		

From Table I it is seen that polonium, sometimes known as radium F, occurs among the end transformation products of radium. Radon tubes, which are no longer suited for their original therapeutic functions, constitute a convenient source of polonium. Radon tubes are small ampules of glass or gold originally charged with the gaseous radon evolved by radium salts. The radon changes into short-lived radium C, which gives off powerful gamma rays and makes the radon tubes medicinally useful. After a few days of usefulness by virtue of the gamma ray

emission the radon tubes are medicinally "spent," since they contain practically no radon or radium C. They now contain mainly the relatively stable solid radium D, which exists as a minute deposit on the inner walls of the tubes. After several months of aging an appreciable amount of polonium is contained in the tubes, formed from the radium D.

Another source of greater commercial importance is the lead residue remaining after the extraction of radium from its ore. The lead residue is preferably treated to form the lead chloride salt. The resulting lead chloride contains all radio-active materials below radium C' in the above table, being rich in radium D (an isotope of lead), which slowly changes into radium E, and containing appreciable radium F, or polonium. Radium E rapidly changes into polonium, as shown in Table I.

Polonium is of especial interest because it is the only known radioactive metal emitting the desirable alpha rays almost exclusively and at a practical rate and giving rise to no other radioactive element emitting undesired beta or gamma rays. The immediate transformation product of polonium, radium G, is an inactive isotope of ordinary lead. Thus no dangerous products develop from polonium, in contrast with radium, certain transformation products of the latter emitting strong beta and gamma radiation. Table I indicates that polonium emits feeble gamma rays in addition to strong alpha rays. It is well known that polonium emits only a few quanta of gamma rays for each million alpha rays (see H. C. Webster, Chemical Abstracts, 32, 1565-6 (1938)). Practically speaking, therefore, polonium emits only alpha rays.

In selecting a radioactive metal for use in preparing a practical radioactive alloy the radiation activity of the metal must be considered. A radioactive element changes into another element according to an exponential law governed by the activity (number of rays per second) of the element. The selection of a metal of very low activity, such as uranium, would be unwise, because, even though it has the long half life (time required for one mass unit of the substance to be reduced to one-half mass unit) of approximately five billion years and emits only the desirable alpha rays, the activity of even the pure metal is too low to be of any practical value. On the other hand, radium A emits only alpha rays very copiously but has a half life of only three minutes.

Polonium emits alpha rays at a rate in excess of 4500 times the alpha emission rate of pure radium, and has the reasonable half life of 138 days, which period of time is sufficient in many cases for the utilization of the strong alpha radiation of the substance. It is practicable to add sufficient polonium to an alloy such that the alloy has useful radioactivity for many purposes several years after it is produced. Only a very minute quantity of polonium need be present in an alloy in order to render same of practical value. For example, a spark plug electrode alloy initially containing as little as two one-hundred-millionths of one per cent of polonium has been found to be effective in ionizing the gas in the gap of a spark plug for an internal combustion engine, at the end of two years, when the polonium content of the alloy has fallen to less than one-billionth of one per cent.

Although polonium is the preferred radioactive metal for use in the present invention for the reasons given above, certain other substances

listed in Table I are strongly radioactive and are suitable for incorporating in the novel alloy. These additional substances are radium, ionium and uranium II. The three additional substances emit alpha rays at effective rates and have sufficiently long half-life periods to render them especially useful in alloys. However, in view of the fact that these substances or their transformation products also emit beta and gamma rays, it is necessary to handle an alloy containing one of them with proper precautions. Radium, ionium and uranium II occur in certain pitchblende deposits and each is extracted therefrom in a substantially pure form by known methods.

A second family of radioactive elements, the actinium series, is shown in Table II, and is adapted from the above-mentioned Landolt-Börnstein reference and Lange's Handbook of Chemistry, pages 60 and 61, third edition, published in 1939 by Handbook Publishers, Inc., Sandusky, Ohio.

Table II

Element	Atomic		Half life	Strong rays	Weak rays
	Wt.	No.			
Actino-uranium	235	92	4.0×10^8 yrs.	Alpha	
Uranium Y	231	90	24 hrs.	Beta	
Protoactinium	231	91	32,000 yrs.	Alpha	
Actinium	227	89	13.4 yrs.		Beta.
Radioactinium	227	90	19.0 days	Alpha	Beta.
Actinium X	223	88	11.2 days	Alpha	
Actinon	219	86	3.92 sec.	Alpha	
Actinium A	215	84	0.0015 sec.	Alpha	
Actinium B	211	82	36 min.		Beta and gamma.
Actinium C	211	83	2.16 min.	Alpha	Beta.
Actinium C'	211	84	0.005 sec.	Alpha	
Actinium C''	207	81	4.76 min.	Beta and gamma.	
Actinium D (lead)	207	82	Stable		

Three metals listed in Table II are strongly radioactive in that they emit strong alpha rays at practical rates and have half-life periods sufficiently long to render them useful in radioactive alloys. These metals are protoactinium, radioactinium and actinium X. Protoactinium is associated with uranium in pitchblende and carnotite, radioactinium and actinium X also being present in these ores. Each of the three active substances can be separated from one of the ores by known methods. Usually, protoactinium is separated first, and radioactinium and actinium X are separately removed from an aged protoactinium preparation by known procedures.

A third family of radioactive elements, the thorium series, is set out in Table III and is adapted from the aforementioned Landolt-Börnstein reference.

Table III

Element	Atomic		Half life	Strong rays	Weak rays
	Wt.	No.			
Thorium	232	90	1.3×10^{10} yrs.	Alpha	
Mesothorium 1	228	88	6.7 yrs.	Beta	
Mesothorium 2	228	89	6.13 hrs.	Beta and gamma.	
Radiothorium	228	90	1.9 yrs.	Alpha	Beta.
Thorium X	224	88	3.64 days	Alpha	
Thoron	220	86	54.5 sec.	Alpha	
Thorium A	216	84	0.14 sec.	Alpha	
Thorium B	212	82	10.2 hrs.	Beta and gamma.	
Thorium C	212	83	60.8 min.	Alpha and beta.	
Thorium C'	212	84	10^{-11} sec.	Alpha	
Thorium C''	208	81	3.1 min.	Beta and gamma.	
Thorium D (lead)	208	82	Stable		

Two of the substances shown in Table III, radiothorium and thorium X, are strongly radioactive metals, in that they emit strong alpha rays at relatively high rates and have half-life periods long enough to allow them to be successfully employed in radioactive alloys. Both of these substances occur in thorium ores and may be separated therefrom by known methods. A convenient scheme for separating them in useful form is first to separate mesothorium 1 and thorium X together, employing this mixture where thorium X is desired. Radiothorium is formed, according to Table III, from the mesothorium 1, when this mixture is allowed to age, and can be readily separated from the aged mixture by known methods. If a relatively pure thorium X is desired, it is obtained by known methods directly from an aged preparation of radiothorium free from mesothorium 1.

According to a preferred method of preparing the novel alloy, a strongly radioactive metal is first plated (by electrodeposition, electrochemical displacement, adsorption or vaporization) onto a base metal (in any convenient form such as, for example, plate, foil, wire, granules or powder). After determining the amount of plated radioactive metal, which amount can be relatively accurately predetermined, by any suitable measurement, as hereinafter described, the plated object is then melted with or without admixture of further base metal or other metal in order to produce the desired alloy or mixture of metals.

Unweighable quantities of the radioactive substance to be alloyed may be measured by a combination of an electroscope and an ionization chamber which has previously been calibrated against a known amount of a radioactive substance or an absolute measuring apparatus. An example of the latter type of apparatus is the so-called "Geiger" counter, such as was described by Rutherford and Geiger in The Proceedings of the Royal Society, volume 811A, page 141 (1908). Both of the above mentioned devices are usually responsive to alpha, beta and gamma rays to a greater or less extent, but may be made responsive only to alpha rays by selection of appropriate conditions, even though other rays are present. By the use of such modifications these devices may be used to determine accurately such an alpha ray emitting substance as polonium, even though radioactive substances which emit rays other than alpha are also present.

Examples disclosed in said application Serial No. 312,441 show the formation of alloys containing polonium in the range of approximately one-one hundred millionth of one per cent to one-one hundred thousandth of one per cent. Obviously an alloy containing even less polonium, such as one billionth of one per cent or less, can be prepared by melting the polonium-plated metal with an even larger proportion of the other metal. Likewise, an alloy containing a larger percentage of polonium than 0.00001 can be produced by melting a plated foil containing a larger proportion of polonium than shown by the foils described in said application Ser. No. 312,441. The invention contemplates radioactive alloys containing preferably less than 0.01 per cent of a strongly ionizing radioactive metal, since the use of an alloy containing more than this proportion of those radioactive metals emitting gamma rays is not practical from the standpoint of the well-being of the user. Moreover, an alloy containing

less than 0.01 per cent of a strongly radioactive alloy has been found to be commercially feasible, whereas the cost of producing one containing a higher proportion of a strongly radioactive metal would be prohibitive from a commercial standpoint.

Although the said examples show the preferred use of nickel or a nickel-manganese alloy as the non-radioactive component of the radioactive alloys produced, a similar alloy may be produced in a similar manner by using any metal having an atomic number in the range of 22 to 28. These non-radioactive metals may be used in a substantially pure form or mixed with each other to form the major part of the alloy. Also, one of these metals may be used in admixture with various other metals, so long as the alloy so produced is substantially equivalent to the alloys hereinabove described.

An important result of the invention is that a practical and relatively cheap radioactive alloy is for the first time provided, and is of such nature that it can be readily and cheaply produced commercially and can be advantageously employed commercially, especially for use in an electrode for a gaseous discharge device, such as a spark plug.

Although it is thought that the mixtures of metals herein described as alloys are properly so designated, it has not been found possible to prove that a true alloy is always produced by the practice of the preferred method. Therefore, it is to be understood in the specification and appended claims that "alloy" means any substantial mixture of two or more metals, such as the product of the preferred method. Although it is thought that the radioactive alloy produced by the preferred method is a substantially uniform dispersion of the radioactive metal throughout the other components of the alloy, it is possible that some of the radioactive metal is oxidized during the alloying process, so that the alloy may contain a substantially uniform dispersion of the oxidized radioactive metal.

The invention includes alloys of strongly ionizing radioactive metals no matter what may be their manner of production. Obvious chemical equivalents may be substituted and other obvious modifications may be made without departing from the nature and spirit of the invention as defined in the appended claims.

What is claimed is:

1. A radioactive alloy composed of one-billionth (0.000000001) per cent to one-hundredth (0.01) per cent of polonium, 2 to 6 per cent of manganese, 0.75 to 1.25 per cent of silicon, and the balance substantially all nickel.

2. A radioactive alloy composed of one-billionth (0.000000001) per cent to one-hundredth (0.01) per cent of radium, 2 to 6 per cent of manganese, 0.75 to 1.25 per cent of silicon, and the balance substantially all nickel.

3. A radioactive alloy composed of from one-billionth (0.000000001) per cent to one-hundredth (0.01) per cent of a strongly ionizing radioactive metal of the group consisting of polonium, radium, ionium, uranium II, protoactinium, radioactinium, actinium X, radiothorium and thorium X, about 2 to 6 per cent of manganese, about 0.75 to 1.25 per cent of silicon, and the balance substantially all nickel.

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