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Encapsulation of Radioactive Noble Gas Waste in Amorphous Alloy

Public demand for the containment and safe storage of radioactive waste materials has caused the U.S. Government to require that, beginning in January 1983, most of the ^{85}Kr, which until now has been vented to the atmosphere during the reprocessing of nuclear fission fuel rods, will have to be captured and retained for several decades. The cost of accomplishing this with present compressed-gas technology is enough to increase the cost of nuclear generated electricity by an estimated 0.3%. However, materials developed for amorphous magnetic bubble memory devices have been found to be capable of storing large quantities of Kr (30 atomic percent) with great stability up to temperatures above 1070 K. The cost of ^{85}Kr storage in the magnetic bubble memory material appears to be less than 1% of that for present compressed-gas technology.

Introduction

The problem of safe disposal of radioactive wastes from nuclear fission power plants is a major obstacle to the continued and expanded use of fission reactors. Perhaps the most difficult radioactive fission product to capture and contain is an isotope of the noble gas Kr, ^{85}Kr, which has a half-life of 10.7 years and emits β-particles at energies up to 0.67 MeV and γ-rays at 0.5 MeV [1, 2]. Unlike most other fission products, it is neither solid (above 121 K) in its elemental form nor can it be reacted to a stable solid compound. Although heavier than air, it mixes thoroughly in the atmosphere; if released even in a deep mine shaft, it would quickly diffuse into the atmosphere. It also diffuses rapidly through water and earth. It is produced in about 0.3% of all ^{235}U fission events. This is about 6% of the Kr and 0.8% of the noble gas produced by fission of ^{235}U. (The other major noble gas produced is Xe.) Almost all processors of nuclear fuel around the world have allowed these radioactive gases to escape to the atmosphere. (It should be noted that essentially all the Kr is released in *reprocessing*; less than 1% is released from the reactor [3].) One exception is the Chemical Processing Plant at the Idaho National Engineering Laboratory, Idaho Falls, which is operated by Allied Chemical Corp. and which has developed several methods [1] to capture ^{85}Kr. The National Engineering Laboratory reprocesses only U.S. Navy nuclear fuels; there are no commercial reprocessing plants at present.

Figure 1 shows the increase in atmospheric ^{85}Kr measured at various geographic locations up to 1968, at which time there were about 56 million curies (56 MCi) or about 10^{27} atoms of ^{85}Kr in the atmosphere worldwide [4]. Almost all ^{85}Kr is introduced by man; of this only 5% is due to nuclear weapons testing. If the rate of expansion of nuclear power along with the concomitant increases in atmospheric ^{85}Kr experienced up to 1968 had continued, there would now be about 0.6 GCi or about 10^{28} atoms of ^{85}Kr in the atmosphere [4]. (The medical consequences of this dose are argued [4] to be slight.) The actual amount is much less due to slowed progress in bringing on nuclear fission power as a replacement for fossil fuels. The rate of release has also been limited by the fact that spent fuel from power reactors is not being reprocessed at present. Spent fuel is stored on-site in deep pools, an unsatisfactory procedure for long-term storage. If nuclear fission power were to provide the projected fraction of our energy needs and if simple venting were to continue, the atmospheric burden would level out at well over 1 GCi. It might also be noted that 1 GCi of ^{85}Kr produces 4 MW of power, which might be put to some practical use if it could be safely handled; admittedly, this is an almost negligible amount compared to the total power that would be produced by the reactors.

To give perspective to the quantities involved, let us note that the fission of ^{235}U produces 200 MeV of thermal energy directly and, depending on design, approximately another 200 MeV of thermal energy by emitting neutrons that produce other fissionable isotopes, principally ^{239}Pu and ^{233}U, by transmutation. Thus, the complete fission of one gram of ^{235}U in a typical reactor would produce about 5.2×10^3 watt-years of heat. As nuclear power plants are about 32% efficient in converting heat to electricity, this one gram of ^{235}U would provide about 1.7 kW of electricity for a year. A typical nuclear power plant generates 1 GW of electricity. To run such a plant continuously for a year requires the complete fissioning of 0.6 Mg of ^{235}U. In a typical fueling cycle, 3% of the initial charge and 1% of the spent fuel is ^{235}U, so that fifty times as much material must be processed as is fissioned. At this rate of production, the alternative of storing spent-fuel bundles on-site is untenable. Thus a typical plant would require 30 Mg of fuel to be reprocessed each year of continuous operation. Of this mass, about 390 g would be ^{85}Kr, about 5×10^{24} atoms or 2.8×10^5 Ci. If we project to the year 2000 and assume that each of 3×10^8 Americans is to be provided electric energy totally supplied by nuclear fission at the present average consumption rate of 2 kW, *i.e.*, 600 GW for the nation, then 600 standard 1-GW plants would be required for the U.S. alone. These would produce 2.3 Mg or 1.7×10^8 Ci of ^{85}Kr annually. If nuclear power were to provide only a fraction of this energy need or if the average electric consumption were to decrease, the ^{85}Kr release would be correspondingly reduced. World production of ^{85}Kr would be at least three times this figure.

U.S. Federal regulations to take effect January 1983 [5] will limit the amount of ^{85}Kr that may be vented to 5×10^4 Ci/GW of electricity generated for one year, for fuel irradiated in 1983 or thereafter. [*Editor's note*: The global body dose rate per capita from the release of all of the ^{85}Kr generated in continuous operation of a 1-GW (electricity) reactor is $\approx 2 \times 10^{-5}$ mrem/year (rem = roentgen equivalent man). This dose rate is about 2×10^{-7} times the average background dose rate; see Reference [3].] Reprocessing with unrestricted venting would result in a release rate about seven times higher than this. The fuel reprocessing plants would be responsible for keeping the ^{85}Kr release down to this level. (A standard reprocessing plant handles about 2 Gg of spent fuel per year, which is the amount produced by 67 standard nuclear power plants, each producing 1 GW of electricity [2].)

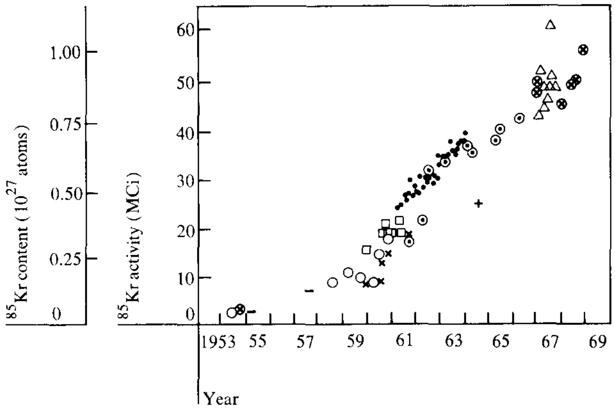

Figure 1 Atmospheric ^{85}Kr as a function of time up to 1968. Data taken from Ref. [3].

Where the Kr has been captured, the only technology available for storing it is to compress it into cylinders [2]; 133 cylinders 23 cm in diameter would be required to contain the noble gas released each year at each fuel reprocessing plant. There are several problems with this method of containment. Rubidium, the decay product of ^{85}Kr, causes a deterioration of ferrous alloys; so there is doubt about the long-term integrity of the cylinders. There is also the danger that the cylinders might burst due to some accident in handling and transport or due to corrosion- and radiation-induced damage over long periods of time. Because the radioactive gas is present in large quantity and under pressure, such an accident could easily be fatal to those nearby unless some means of secondary confinement of the gas is provided. The cost of meeting federally imposed safety standards with the compressed gas technology is rather high [2]. The estimated cost of a facility to contain on a 40-year cycle the compressed gas produced by a single reprocessing plant is \$208.5 million. For a 30-year loan at an 11.5% interest rate, this would require an annual payment of more than \$24 million. The cost of compressing the gas, of purchasing and transporting the cylinders, and of salaries and energy would be additional. The warehouse cost alone would run to more than \$200 million per year for the U.S. by the year 2000. In other terms, this would add \$0.00006 to the cost of generating a kWh of electricity, which would be an increase of about 0.3%.

Proposed alternate methods of storage have included incorporation into zeolite lattice pores by high temperature-pressure diffusion and by incorporation into crystalline [2a] and amorphous [2b] metals. The zeolite method

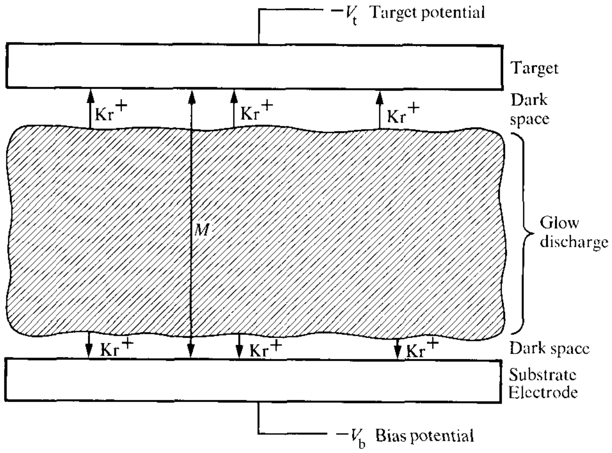

Figure 2 Schematic representation of the bias sputtering process.

suffers from the fact that if water gets to the material, it reacts and rapidly releases the gas. In crystalline metals the Kr forms small bubbles. At high concentration the pressure in these bubbles is sufficient to cause mechanical failure, a phenomenon known as blistering, in which the gas escapes. Furthermore, these bubbles tend to collect at grain boundaries and microcracks along which they diffuse at significant rates even at room temperature. Moreover, due to the power produced by the decay of the ^{85}Kr, the containment material will be self-heated to a temperature dependent on the size of the individual container; the larger the container, the larger the maximal temperature and the more severe the thermal diffusion and degradation. For most storage schemes the volumes of containment material required are substantial. Each reprocessing plant would require [2] the following volumes per year for the various proposed methods: compressed gas cylinders, 6.5 m^3; zeolite, 7.3 m^3; Ni, 1.3 m^3; Al, 1.6 m^3; glass, >190 m^3.

Storage in bias-sputtered amorphous metallic alloy

In the course of development of amorphous materials for magnetic bubble memory devices [6], we have come upon a method for the storage of Kr, Xe and other noble gases, whether or not radioactive, which seems capable of containing the radioactive waste from one of these reprocessing plants in just 0.2 m^3 of material, and of retaining it stably up to temperatures as high as 1070 K. We estimate the cost of storing the ^{85}Kr by this method as well under 1% that of storage in the compressed-gas cylinders, *i.e.*, less than \$0.24 million per reprocessing plant.

The containment materials in question are formed by bias-sputter deposition [7]. This process is illustrated in Fig. 2. A low-pressure discharge is established in a sputtering gas between two electrodes, one of which is known as the target and the other as the substrate electrode. The sputtering gas is normally chosen to be one of the noble gases, He, Ne, Ar, Kr, or Xe, to avoid chemical reactions with the target and substrate materials. In practice, Ar is usually chosen on the basis of cost and sputtering rate. The Kr and Xe sputter as rapidly in most applications but are more expensive. The discharge converts the noble gas to a positive ionization state, *e.g.*, Kr^+. These ions are accelerated toward the target electrode, which is biased negative with respect to the plasma by the *target potential*. The plasma is in turn biased from ground by a small *plasma potential*. When the noble gas ions reach the target surface they penetrate several atomic layers, producing a process known as a collision cascade in which the energy of an incident ion is transferred to many atoms of the target material. Several of these atoms are subsequently emitted from the target surface in a manner similar to the "break" at the start of a game of billiards. The target atoms are generally neutral and travel by virtue of their kinetic energy through the intervening space between the target and the substrate, perhaps suffering a few collisions with the sputtering gas on the way. For normal choices of substrate temperature and materials, virtually all of the target atoms reaching the substrate stick there. As normally practiced, this results in the growth of a polycrystalline film on the substrate. However, Nowick and Mader [8] discovered that when two or more elements are deposited simultaneously and the radii of their atoms are sufficiently different, the resultant films are not polycrystalline but amorphous. (This means that they are microscopically disordered but macroscopically homogeneous as contrasted to the polycrystalline films, which are microscopically ordered but macroscopically disordered.) It is also possible to obtain amorphous films with atoms all the same size if one deposits faster than a critical rate, this rate being a function of substrate temperature [9].

In bias sputtering, a *substrate bias* is also applied between the plasma and the substrate. This has the effect of accelerating noble gas ions toward the surface of the growing film as well as toward the target. The ion bombardment of the film during growth has a number of useful effects. In the first place, it introduces anisotropies in the properties of the film. In the development of amorphous magnetic bubble materials, it was necessary to use this effect to induce a perpendicular easy axis of magnetization. In the second place, it allows one to eliminate many types of impurities that are not as well bound as host atoms. This is done by inducing a collision cascade in the substrate that is not sufficiently violent to remove host atoms. A third effect, which was discovered by

Cuomo and Gambino [10], is that high concentrations of noble gas ions tend to remain stably in the growing amorphous metallic films. Sputtered gas ions are also known to become incorporated in polycrystalline films formed by bias sputtering, but only in concentrations much lower than those observed with the amorphous-alloy materials. As noted above, this method has previously been suggested as a means of storing ^{85}Kr in polycrystalline Ni or Al material.

In order to understand the difference between the noble gas containment properties of amorphous and polycrystalline bias-sputtered materials, we need to understand the atomic structure of the two classes of solids.

In the polycrystalline phase of these alloys, the atoms assume a close-packed structure within each crystalline grain, as they would in an elemental film. In the close-packed structure foreign atoms can be accommodated only on substitutional sites, where they replace a host atom, or in one of the interstitial spaces between host atoms in their regular crystalline array. The noble elements do not form substitutional impurities because these elements are much less chemically reactive than the host atoms they would replace. Moreover, the interstitial spaces in the crystalline structure are large enough to accommodate only He, the smallest of the noble gases. Therefore, although bias sputtering causes the larger noble gases to be incorporated into crystalline alloys, they are found in grain boundaries, dislocations, and, at large concentrations, in macroscopic bubbles [11]. Such bubbles of noble gas are found both in the grain boundaries and in the bulk of the crystallites and, as noted above, tend to destabilize the structure and to diffuse out of the material.

The structure of the type of amorphous alloy films we are concerned with has been most successfully described by the dense-random-packing-of-hard-spheres (drphs) model [12]. In particular, the drphs model has been very successful in explaining the observed radial-distribution function of amorphous alloys. Some materials, such as elemental Se, form stable amorphous phases due to directional covalent bonding between atoms that tend to favor chain or ring structures. In the amorphous phase of these materials, cross-linking between chains and/or rings tangles the covalent network, preventing it from assuming an ordered array. In our case, the individual atoms behave much more like hard spheres than do atoms of Se and other "glass-forming" elements. Our materials are stabilized in the microscopically disordered amorphous phase by the mismatch in size of the atoms of the two or more elements present. The microscopic disorder of such amorphous alloys introduces a large number of interstitial

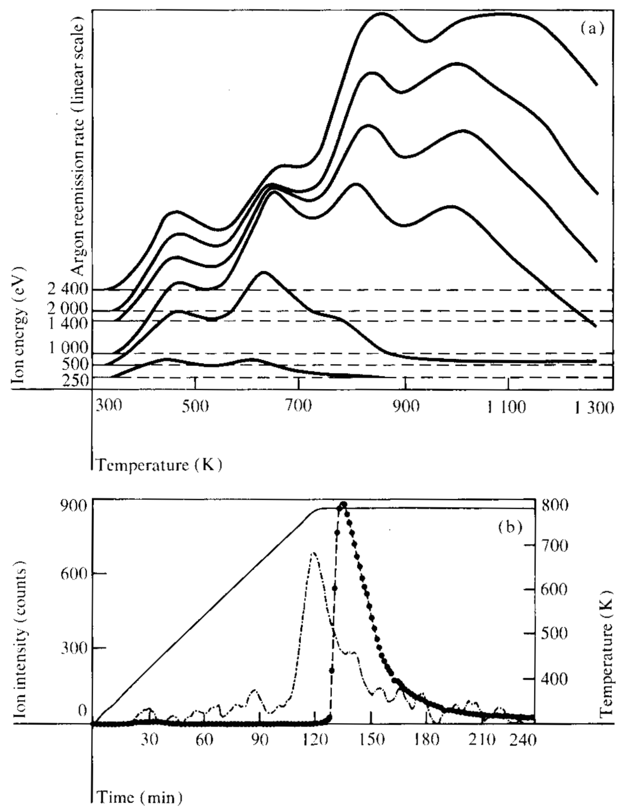

Figure 3 (a) Argon release from a nickel single crystal; data of Rantanen *et al.* [13a, his Fig. 6]. (b) Krypton release from amorphous GdCo (dotted line and data points). The solid line is a plot of temperature as a function of time (right-hand scale). Evolution of H_2 is shown by the dot-dash line [14].

spaces that are much larger than those in the crystalline phase. In favorable cases these interstitial spaces are large enough to accommodate a large noble gas atom such as Kr.

This difference in the distribution of the noble gas in the amorphous film as compared to the crystalline film has a profound effect on the stability of containment of the gas and on the kinetics of the process by which that gas may be liberated [13a, b; 14a]. For example, Rantanen *et al.* [13a] have studied the rate of evolution of noble gases from crystalline Ni. Their data for Ar release from a ⟨100⟩ oriented Ni single crystal are shown in Fig. 3(a). The samples were bombarded with 3.86×10^{14} ions/cm^2 at ambient temperature with the ion energies indicated in the figure. The samples were then heated at a linear rate of 100 K/min to obtain the thermal re-emission spectra shown. Note that there is a detectable evolution of Ar at all temperatures above the implantation temperature, 300 K. Also, there is considerable structure to the evolution curves, with peaks occurring at 478, 630, 794, and

990 K. The authors associate these peaks with mechanisms having activation energies of 1.31, 1.74, 2.21, and 2.78 eV, respectively.

Rantanen *et al.* [13a] also studied the thermal re-emission spectra of Kr from polycrystalline Ni. They reported activation energies of 1.18, 1.36, 1.50, and 1.71 eV for this case. They also pointed out that these activation energies are probably associated with interstitial migration (1.03-1.09 eV), vacancy formation (1.35 eV), vacancy migration (1.55 eV), and surface diffusion (1.68 eV).

The above results for Kr in polycrystalline Ni should be compared with the thermal release of Kr from amorphous GdCo and GdCoMo alloy films by Frisch and Reuter [14]; see Fig. 3(b). The method used to study the amorphous film was similar to that of Rantanen *et al.*, except that the heating rate was 10 K/min and a high-sensitivity mass spectrometer was used. Extensive measurements have been made on a large number of these bias-sputtered amorphous GdCo and GdCoMo alloy films. All of the thermal re-emission spectra for *unoxidized* films have the character shown in Fig. 3(b). Oxidation lowers the temperature at which Kr release occurs [14b]. In the amorphous alloy films no detectable rate of noble gas evolution was observed until the film began to crystallize [14a]. At the crystallization temperature the gas was evolved very rapidly. In this case the kinetics of gas liberation are determined by the kinetics of the crystallization, which is a nucleation-and-growth process. An activation energy of 4 eV has been estimated for the migration of Kr in amorphous GdCo alloy [14a]. This implies that the mean time to diffuse one atomic site would be about 10^{14} years at 570 K; at 1070 K, the Kr would diffuse about 10 nm in the 40 years required for the radioactivity to decay to 3% of its original value.

A further benefit of an amorphous structure for a material to contain ^{85}Kr is that the disorder improves the ability of the material to tolerate radiation damage and impurities. Even if the containment material were pure to begin with, it would not remain so because the ^{85}Kr transmutes to Rb by radioactive decay. The stability of a crystalline host material would be adversely affected by the simultaneous effects of irradiation, which generally enhances atomic diffusion, and of the incorporation of the daughter isotope, which is chemically incompatible with the crystal lattice of the proposed host materials. This would cause embrittlement of a crystalline host material and would accelerate mechanical failure by such mechanisms as blistering. However, those amorphous alloys which are stabilized by atomic size mismatch and a highly disordered drphs structure are much less sensitive to the chemical nature of minor impurity constituents and can exist over a broad range of composition. The amorphous alloys in question will contain about 30 at% Kr or Xe, but, as noted above, only 6% of the total Kr released at the reprocessing plant would be radioactive ^{85}Kr. Let us assume that the Xe is separated out by distillation so that only Kr is stored. This would seem to be economically desirable, although one could also easily store the Xe by expanding the size of the sputtering unit. Eventually, 1.8 at% Rb will be contained in the storage material. This would be enough to affect many crystalline hosts substantially but would have a negligible effect on a drphs-amorphous host. Such host materials are also less susceptible to radiation damage because the currents produced by ionizing radiation do not persist as long and because the resultant atomic diffusion does not have as much effect on a structure that is already disordered.

The selection of the most practical composition from which to form the encapsulating host material requires the consideration of four factors: gas-incorporation capacity, thermal stability, chemical stability, and cost. Let us start with the amorphous magnetic bubble memory material, GdCoMo, for which the incorporation of large quantities of noble gas was first discovered. This material can incorporate more than 50 at% Ar and more than 30 at% Kr and Xe when the three bias voltages of the system are adjusted properly. This large noble gas incorporation capacity occurs because the rare earth element Gd has an atomic radius much larger than the first-series transition element Co. The second-series transition element Mo is intermediate in size and serves to further disorder the drphs structure so that these mixtures will condense in an amorphous phase over a wide range of compositions and will have a relatively large number of interstitial spaces large enough to accommodate a Kr or Xe atom. However, the GdCoMo composition of the magnetic bubble memory would not be an attractive choice from the point of view of cost. Because the rare-earth elements (which in fact are not that rare) are all very similar in their chemical behavior, they are expensive in their pure elemental form. A typical price for pure Gd would be $500/kg. If one instead purchases the rare earth elements in an unseparated form, called mischmetal or RMM [15], the price is much less, typically $10/kg, and the chemical behavior as it affects Kr storage in amorphous alloys is no worse. One can also replace Co with Fe without affecting the containment properties significantly. With respect to thermal stability, it has been shown that GdCoMo and GdCoCr ternary alloys are much more stable than binary alloys like GdCo or even ternary alloys containing Au or Cu, *e.g.*, GdCoAu or GdCoCu. For example, 15 to 20 at% Mo increases the crystallization temperature from 770 K for GdCo to more

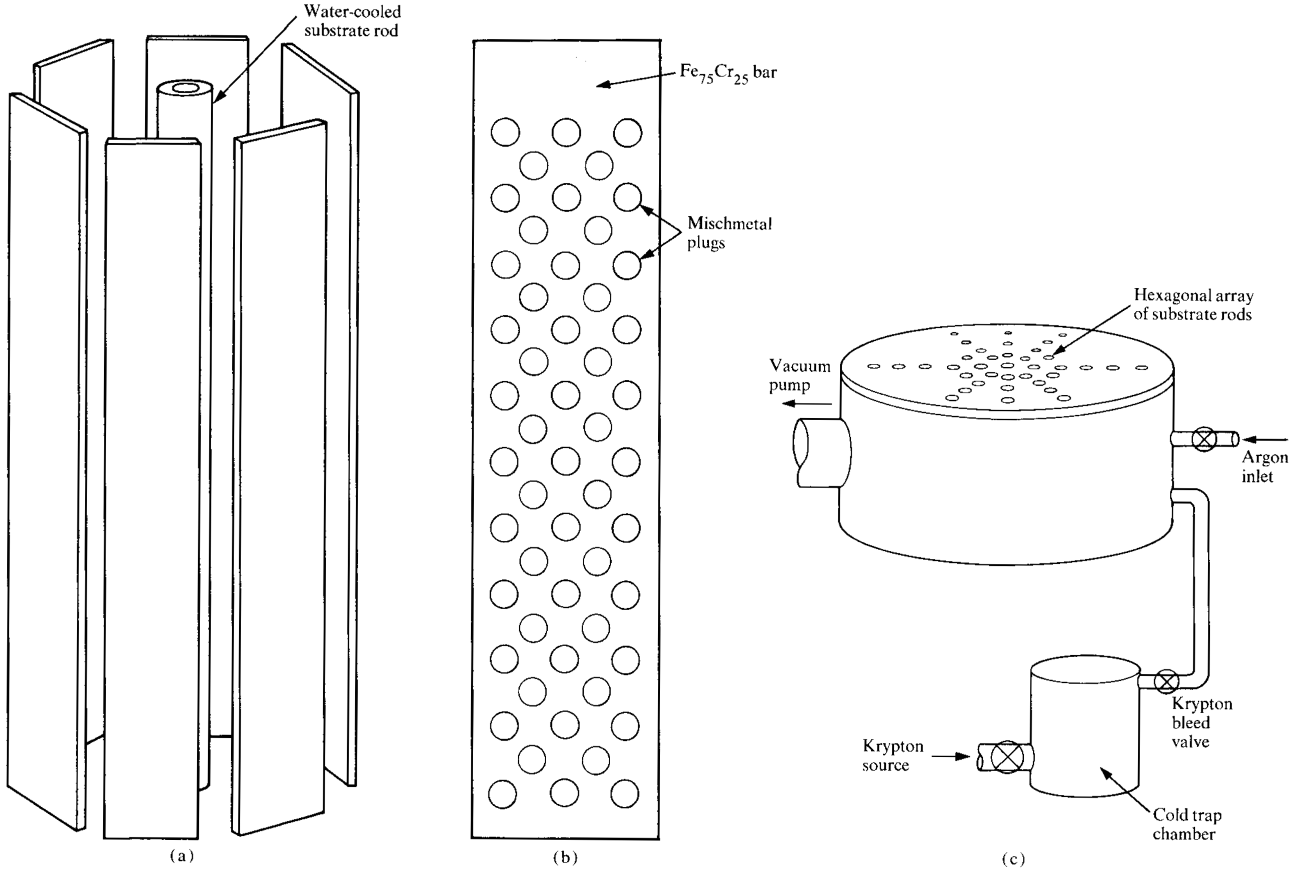

Figure 4 Proposed sputtering apparatus for incorporation of Kr into the amorphous alloy on a production scale. (a) Modular sputtering unit, (b) target bar assembly, and (c) sputtering chamber.

than 1070 K for the ternary alloys. From the point of view of chemical stability, the rare earth concentrations should be kept low because these materials oxidize (as well as cost more than the other constituents). Chromium, on the other hand, significantly improves the oxidation resistance and should be added at a concentration consistent with its cost. Therefore, an appropriate composition for the containment application would be (in atomic percent): RMM 20%, Fe 60%, and Cr 20%. The 2.1 Mg, or about 0.2 m^3, of this composition that would be required to store the Kr retrieved at each 2-Gg/year reprocessing plant would cost about $10 thousand. Of course, this material could be recovered and recycled every century or so as the level of Kr radioactivity from each charge decreases.

The process

At the reprocessing plant, spent fuel elements containing UO_2 ceramic pellets encased in metal are dissolved in nitric acid. At this point the Kr and Xe are released and bubble out of solution together with several other volatile species. The various volatile species can be separated and trapped in a cryogenic distillation tower [1]. The Kr and Xe would be trapped at the end of the distillation sequence in cold traps or on charcoal cooled to 77 K with liquid nitrogen.

The liquified noble gas is maintained at 77 K and transferred to the sputtering station for incorporation into the amorphous alloy; see Fig. 4. The vapor pressure of the Kr at this temperature is about 10^2 Pa (10^{-3} atm), which is enough to bleed through valving into the sputtering chamber but low enough that the danger of excessive leaks would be easily managed. The gas pressure in the sputtering chamber is about 10^{-2} Pa (10^{-7} atm). [Compare this situation with that of the compressed gas cylinders, which handle the gas at a pressure of about 10^7 Pa (10^2 atm).]

The rate at which material may be deposited by bias-sputter deposition varies from 1 μm/h for very simple diode systems to 30 μm/h for systems that use electron-injection or magnetic-field confinement of the plasma. We feel that the most practical arrangement would be modular and would consist of a hexagonal array of water-cooled substrate rods surrounded by bar-shaped target electrodes. With this arrangement a continuous deposition rate of 10 μm/h would be practical. In order to deposit the 0.2 m^3 of material per year required to contain the Kr retrieved at each 2-Gg/year reprocessing plant, the volume deposition rate will have to be 2.3×10^{-5} m^3/h, so that 2.3 m^2 of deposition area are needed. This can be accommodated with a system of 232 rods 2 cm in diameter and 30 cm long arrayed honeycomb fashion in a cylindrical vacuum chamber 1.5 m in diameter and 0.5 m high. Such sputtering systems sell commercially for about $80 thousand [16].

About 200 kW/m^2 input power would be required to sputter at the proposed rate of 10 μm/h [17]. Therefore, the sputtering station would consume about 460 kW of electrical power in order to capture the Kr retrieved at a 2-Gg/year fuel reprocessing plant. At $0.04/kWh the cost of this power would be $160 thousand per year. Perhaps another $10 thousand per year of electricity would be consumed running the vacuum, cooling and control systems.

Due to the inherent simplicity of the sputtering process itself, this could easily be automated or remotely controlled. The cost of special control equipment for the radioactive environment automated operation should not exceed $100 thousand. However, the deposited rods would have to be removed and replaced periodically. This could be accomplished by valving off the source of ^{85}Kr and of the cooling water, breaking the vacuum of the system, and pulling the top flange of the vacuum chamber with all the rods and the remains of the target electrodes attached to it out of the body of the vacuum chamber and removing it from the sputtering station. Operators could then attach a new top flange with substrate-rod assembly and target electrodes to the vacuum and cooling systems. This should be done about once a month after about 7 mm of material has been deposited on the rods.

The configuration of the target electrodes shown in Fig. 4 indicates that these consist of $Fe_{75}Cr_{25}$ bars with mischmetal plugs inserted into drilled holes. This configuration is recommended for easy handling of the mischmetal, which is hard and brittle. With this configuration one could also arrange to coat the deposited layer of amorphous metal with crystalline stainless steel in order to provide further protection from corrosion and abrasion, and to contain the beta particles emitted by the Kr. This would be done by continuing to sputter after the Kr source had been turned off and the mischmetal plugs nearly consumed, and the bias voltage would be increased to 250 V in order to increase the fraction of Fe and Cr in the deposited mixture.

For final storage one might wish to pot the entire top flange, rod and target remains assembly in cement and wrap it in lead. However, we feel that the amorphous alloy is so stable a method of storage that the material could be released for several practical applications (ranging from nuclear batteries to fire detectors, cold-cathode stabilizers, thickness monitors, and simple sources of heat) rather than simply putting it away in a deep salt mine.

Conclusion

The materials developed for the amorphous magnetic bubble memory system have been shown to provide a very stable medium for the long-term/high-temperature storage of the noble gases Kr and Xe. The radioactive isotope ^{85}Kr, produced in ^{235}U fission reactors, is difficult and expensive to contain by other means. Compared to the present technology of compressed-gas cylinder storage, which is estimated to cost $24 million per year per reprocessing plant for warehouse amortization alone, our process would cost approximately $180 thousand for capital equipment, which would be amortized at less than $40 thousand per year, plus $10 thousand per year for materials and $170 thousand per year for electricity. In our economic analysis we have not considered the cost of the building to contain the process; but since the process runs at high vacuum instead of at high pressure and since the product is quite stable to high temperatures, we feel the cost of this building should be minimal. In the high-pressure cylinder technology the cost of the building is a major part of the total expense. With our process the radioactive material is present only in small quantities before it is incorporated into the solid, and because of the stability of that solid, can be dispersed in practical applications afterwards.

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15. Mischmetal is the material from which cigarette lighter flints are made, and is available from Moly Corp., Inc., White Plains, NY.
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Received November 6, 1978; revised January 8, 1979

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