CLADDING AND OTHER STRUCTURAL MATERIALS

12. CLADDING MATERIALS FOR SPACE ISOTOPIC HEAT SOURCES

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The purpose of this program is to develop new cladding materials for containment of radioisotopes in power systems for use in space. We are attempting to develop a single alloy with an optimum combination of strength at high temperature, fabricability, environmental stability, and resistance to oxidation in air to take the place of the layered combination of strength member, diffusion barrier, and oxidation resistant cladding used in present devices.

Development of Improved Alloys

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The material for fuel capsules for space thermoelectric systems should (1) be useful in the range 1000 to 1600°C; (2) be resistant to oxidation, evaporation, and corrosion by soil and seawater; (3) have a high melting point and high impact resistance; (4) be compatible with the fuel, ablative materials, and specialized coatings; and (5) be fabricable and weldable. The purpose of this task is to develop, in light of these requirements, new cladding alloys specifically for isotopic heat sources for space.

Noble Alloys (C. T. Liu)

Tensile tests of the experimental Pt-base alloys were conducted at elevated temperature in a newly designed apparatus in which a temperature of about 1300°C can be attained at a vacuum of 1×10^{-5} torr. The tensile properties at both room temperature and elevated temperatures are given in Table 12.1. For comparison, the data for Pt–30% Rh and TZM are also included.[1] The ternary alloys that contain small amounts of Ti or Hf are much stronger than the binary alloys Pt–5% Mo and Pt–30% Rh at high temperatures. They are as strong as TZM at 1093°C but weaker at 1290°C.

Table 12.1. Tensile Properties of Noble Alloys at Room Temperatures and High Temperatures

Alloy	Ultimate Tensile Strength (psi)	Elongation (%)	Reduction in Area (%)
	Room Temperature		
Pt–5% Mo	94,000	22	70
Pt–3% Mo–1.76% Ti	89,000[a]		
Pt–6% W–1% Hf	118,000	23	75
	1093°C		
Pt–5% Mo	18,400	20.6	37
Pt–3% Mo–1.76% Ti	27,000	29	51
Pt–6% W–1% Hf	33,300[b]	2.6[b]	
Pt–30% Rh[c]	22,000		
TZM[c]	33,000	24	
	1290°C		
Pt–3% Mo–1.76% Ti	12,700	48	100
Pt–6% W–1% Hf	13,500	19	23
$Pt_3(Cr_{0.86},W_{0.14})$	19,500	6	
Pt–30% Rh[c]	8,000[d]		
TZM[c]	22,000[d]	30[d]	

[a]Specimen fractured within the elastic limit.

[b]Tested at 1000°C.

[c]Multi-Hundred Watt, Radioisotope Thermoelectric Generator Program, GESP-7034 (1970).

[d]Tested at 1316°C.

The Pt-Mo alloy looses ductility at room temperature when alloyed with 1.76% Ti, but Ti enhances the ductility of the Pt–3% Mo–1.76% Ti alloy at high temperatures (e.g., 48% elongation and 100% reduction of area at 1290°C). The Pt–6% W–1% Hf alloy has good ductility at room temperature but reaches a minimum in ductility at about 1000°C. In order to understand this, we analyzed the interstitial content of the alloy and characterized its fracture mode by electron microscopy. Chemical analyses indicated that this alloy did not contain interstitial levels higher than the other two alloys. Electron micrographs of fracture surfaces showed no evidence that gas bubbles had caused the low ductility of this alloy. The binary Pt-W alloy behaved similarly.[2]

Thus, we believe that this brittleness at intermediate temperatures is probably associated with the segregation on the grain boundaries of substitutional impurities that have low melting points when Pt and W are alloyed.

We studied the oxidation of the Pt–5% Mo–0.5% Hf alloy in air at 1000°C. Although the molybdenum oxide is very volatile at this temperature, the alloy lost weight at an average rate of 1.7×10^{-3} mg cm^{-2} hr^{-1}, which is only a factor of 2.5 higher than that for pure Pt. The microstructures of the Pt–5% Mo–0.5% Hf and Pt–6% W–1% Hf alloys after exposure to air at 1000°C for 492 and 270 hr, respectively, were examined metallographically. A small degree of internal porosity was observed in the Pt–5% Mo–0.5% Hf alloy. The formation of the porosity at the grain boundaries in this alloy was presumably due to the diffusion of Mo along the grain boundary from the interior to the surface, where it was removed by oxidation. No porosity was observed in the Pt–5% W–1% Hf alloy. We also studied the effect of oxidation on fabricability. We found that both oxidized specimens could be cold rolled 50% or more without cracking. Hence, we concluded that oxidation in air did not impair the ductility of these two alloys at room temperature.

In order to determine the recrystallization temperature and softening behavior, the three ternary alloys (Pt–5% Mo–0.5% Hf, Pt–6% W–0.5% Hf, Pt–6% W–1% Hf) were cold rolled 35% and then aged 1 hr at 200 to 1200°C in evacuated quartz tubes. Figure 12.1 shows the plot of hardness as a function of annealing temperature. In general, the variation of hardness for these three alloys followed the same trend. Above 400°C, the hardness decreased steadily with increasing temperature. Hardness dropped sharply above 1000°C. Metallographic examination showed the formation of a few recrystallized grains at 1100°C. The annealing temperature for 50% recrystallization was about 1200°C; as a comparison, the annealing temperature for 50% recrystallization of unalloyed Pt is only 525°C (ref. 3).

The melting point of the Pt-base alloys we have studied is between 1750 and 1850°C. In order to develop alloys with higher melting points, we are now working on Pt–31% Rh-base alloys [Pt:Rh (atomic ratio) = 1] further alloyed with W and Re. Two alloy buttons with the nominal

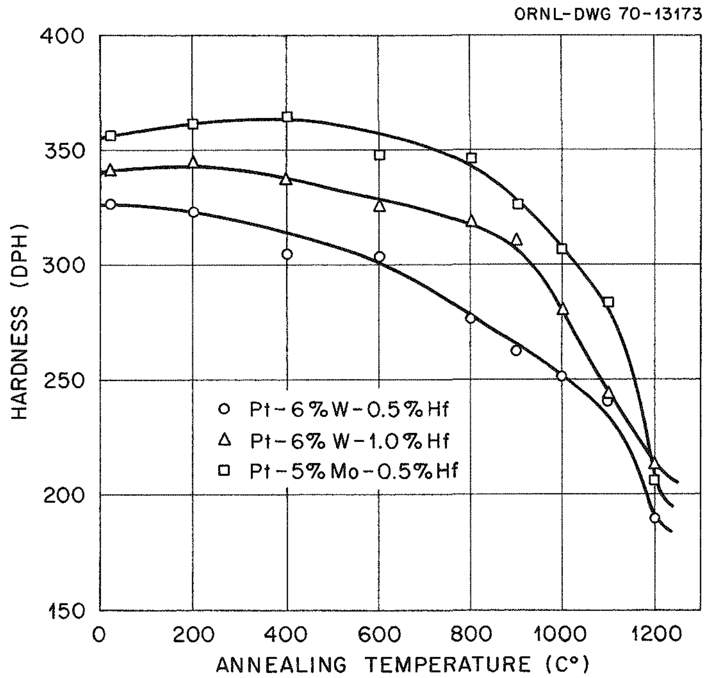

Fig. 12.1. Effect of Annealing Temperature on Hardness of Three Platinum-Base Alloys Cold Rolled 35% in Reduction.

composition listed in Table 12.2 were prepared by arc melting. Based on the binary alloy systems, the estimated melting points of these alloys are about 2000°C. In order to determine their melting points, we so modified the hot zone of a vacuum furnace that it can be heated to about 2200°C by induction. Preliminary determination shows that the melting points are both about 2060°C. The cold fabricability of the Pt-Rh-W alloy is good. The Pt-Rh-Re alloy is also fabricable cold, although it was cracked after rolling 18%.

Table 12.2. Experimental Platinum- and Rhodium-Base Alloys

Composition (%)	Melting Point (°C)	Rockwell Hardness (R_A)	Cold Fabricability
Pt–31.3 Rh–9.9 Re[a]	~ 2060	53	Fair
Pt–31.3 Rh–9.7 W[a]	~ 2060	44	Good

[a]Atomic ratio of Pt:Rh = 1.

Notes

1. Multi-Hundred Watt, Radioisotope Thermoelectric Generator Program, GESP-7034 (1970).

2. E. P. Sadowski, "Stress-Rupture Properties of Some Platinum and Palladium Alloys," pp. 465–482 in Refractory Metals and Alloys, vol. 11, ed. by M. Semchyshen and J. J. Harwood, Interscience, New York, 1961.

3. R. W. Douglass et al., High Temperature Properties and Alloying Behavior of the Refractory Platinum-Group Metals, NP-10939 (1961).