

# Dependence of Mechanical Properties of Platinum-Rhodium Binary Alloys on Valence Electron Parameters

## Increasing valence electron ratio improves alloy properties

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Dependence of mechanical properties of binary platinum-rhodium alloys on valence electron

ratio (VER), number of valence electrons ( $e_v$ ) and average atomic number of the alloys ( $Z$ ) are investigated. The alloys have a high number of valence electrons ( $9 \leq e_v \leq 10$ ) and a wide range of average atomic numbers ( $Z = 45-78$ ). Clear correlations between VER of the alloys and their mechanical properties are found. By increasing the VER of the alloy from 0.13 to 0.20 following the increase of rhodium content in the composition, the hardness, elastic modulus and ultimate tensile strength (UTS) of the alloy increases. Creep rates of the selected alloys clearly decrease with increasing VER at high temperatures (1500–1700°C), while stress rupture time at different temperatures consistently increases because of higher rhodium content in the alloy solid solution chemistry. Dependence of mechanical properties on valence electron parameters is discussed with reference to the atomic bonding.

## 1. Introduction

Platinum alloys attract considerable attention as new areas of application are found and continuously explored. Platinum alloys are used to produce jewellery (1, 2), thermocouples (3), resistance thermometers (4), resistance temperature detector sensors (5), catalysts (6) and glass fabrication equipment (7–9) due to their high strength, good workability and corrosion resistance at high temperatures (10, 11). The analysis of Gavin clearly indicates increased attention to platinum alloys (12). Hu *et al.* have recently presented research into platinum-based superalloys for high-temperature applications (13). Solid solution strengthened platinum-based alloys have been

the subject of research and development for some time. All transition group elements have considerable solid solubility in platinum. The elements near platinum in the periodic table form a continuous solid solution with platinum and have different degrees of solid solution strengthening effect on the platinum matrix. Platinum-based alloys are also being developed for high-temperature structural applications with the aim of replacing some of the currently used nickel-based superalloys (NBSAs) (13). Platinum-based superalloys have a similar structure to NBSAs and can potentially be used at higher temperatures and in more aggressive environmental conditions. Platinum-based superalloys show strong chemical stability and excellent mechanical properties such as high creep strength and ductility and can be used for next generation gas turbine materials and in chemical, glass and space technologies (14–21).

Of all platinum alloys, currently the most widely used are alloys of platinum with rhodium. Their range of application temperature is extended up to 1600°C (10). Luyten *et al.* have examined the platinum-rhodium, platinum-palladium, palladium-rhodium and platinum-palladium-rhodium phase diagrams using the Monte Carlo simulation method in combination with the modified embedded atom method (MEAM) and optimised parameters (22). Thermodynamics and phase equilibria of the binary platinum-rhodium system were studied by Jacob *et al.* (23) and Okamoto (24). To adjust the properties of these intermetallics, rhodium as the alloying element is added to platinum. Platinum and rhodium are completely miscible and form a single-phase solid solution at all concentrations of rhodium as shown by Okamoto (24).

New materials based on dispersion strengthening of platinum-rhodium alloys have been developed for high-temperature applications (25–27). Microstructural analyses of selected platinum alloys used in industry and jewellery (including PtRh10 and PtRh30) were performed by Battaini (28). Fischer *et al.* (29) studied the stress-rupture strength and creep behaviour of platinum, platinum-rhodium and platinum-iridium alloys, dispersion hardened platinum materials, rhodium and iridium. Investigations of elastic properties (Young's modulus, modulus of rigidity and Poisson's ratio) for platinum, platinum alloys, rhodium and iridium at high temperatures using a resonance technique have been carried out by Merker *et al.* (30). They observed that Young's modulus and the modulus of rigidity of platinum, rhodium, iridium and various platinum alloys in the as-cast condition decrease

linearly with increasing test temperature. Platinum-rhodium alloys have more stable properties: an increase in rhodium content leads to higher temperature durability, extended creep life and decreased creep rate (13, 29). At higher rhodium content, however, the machinability of the alloys may be negatively affected (13). Darling presented the high-temperature and room-temperature properties of platinum-rhodium alloys as a function of rhodium content and service temperature in earlier work (31), while the work of Ackmen on the mechanical properties of platinum-rhodium was the first published report available in the literature (32). Rdzawski and Stobrawa studied the microstructural evolution and mechanical properties of yttrium- and boron-alloyed platinum-rhodium (33). Garbacz *et al.* studied the microstructure and mechanical properties of a platinum-rhodium alloy produced by powder metallurgy and subjected the alloy to plastic working (34).

One of the research goals in this area is to develop materials with better high-temperature mechanical properties (for example, tensile, fracture, creep and thermomechanical fatigue properties) and environmental stability (for example, resistance to high-temperature oxidation and hot corrosion) than NBSAs (13, 35). Platinum and its alloys represent a major challenge because of the high cost of materials and need for special experimental conditions (10). Finding fundamental correlations between structural factors of these alloys and their mechanical properties can help reduce the cost with less trial-and-error and difficulty.

To understand the factors that control the elastic and mechanical properties and transformation temperatures in alloy systems, Zarinejad suggested the need to acquire in-depth knowledge of how the atomic bonding strength of the solid solutions can be influenced by the chemical composition of the alloys (36). Apart from microstructural features such as precipitates and grain boundaries, the mechanical properties of the matrix crystals of solid solution alloys in several alloy systems such as nickel-titanium (37–41), titanium-palladium, titanium-platinum (42), titanium-gold titanium-iridium (43) and many other alloy systems (44) were shown to be influenced by the alloy chemistry and how the metallic bonding changes with chemical composition change. Hence, the dependence of mechanical properties of solid solutions on electron variables in alloys was introduced (36–44). It was shown that the mechanical properties (elastic moduli and plastic deformation characteristics) of the solid solution crystals are influenced by

changes in the chemical composition. Knowing that in metallic materials, the delocalised valence electrons dominate the bond strengths and elastic properties (45, 46), it is of particular importance to study the electronic parameters of alloys. In this study, the investigation is extended to the platinum-rhodium binary alloys to help understand the dependence of the mechanical properties on alloy chemistry.

The aim of this study is to show the dependence of mechanical properties of binary platinum-rhodium alloys on the VER of the alloys, which is related to the chemical composition, or the atomic fraction of platinum and rhodium in the alloys. The study focuses on existing data on hardness, tensile strength, creep rate and rupture time for the platinum-rhodium binary alloys. We reveal and explain the dependence of the mechanical properties of platinum-rhodium binary alloys on these parameters with a focus on the correlation of the mechanical properties to VER of the alloys. The correlations found in this study will provide important understanding towards effective alloy design.

## 2. Materials and Methods

Mechanical properties data including creep rate, rupture time, tensile strength and hardness of single-phase platinum-rhodium binary alloys (without precipitate or mechanical work) made to date were investigated. The data were extracted from the relevant literature especially the works of Trumic *et al.* (10, 11), Fischer *et al.* (29), Darling (31), Acken (32), Hu *et al.* (47) and other sources (48, 49) as listed in tables or presented in graphs. For the analyses in this study, three simple parameters were considered: (a) the average atomic number of the alloys; (b) the number of valence electrons and (c) VER of the alloys.

The average atomic numbers of the alloys were calculated based on the atomic fractions and atomic numbers of the elements comprising the alloys as follows, Equation (i):

$$Z = f_{Pt}Z_{Pt} + f_{Rh}Z_{Rh} \quad (i)$$

where  $Z$  is the fraction-averaged atomic number of the alloy.  $Z_{Pt}$  and  $Z_{Rh}$  are the atomic numbers of platinum and rhodium, respectively.  $f_{Pt}$  and  $f_{Rh}$  are the corresponding atomic fractions of platinum and rhodium in the alloy. To study the dependence of the transformation temperatures on the chemistry of alloys, the basic electron configurations of the alloys are analysed in the following section.

The number of valence electrons in a transition metal atom is usually considered as the number of  $d$  and  $s$  electrons. The valence electrons per atom of platinum-rhodium binary alloys can be calculated based on the atomic fractions of the elements in the alloy by Equation (ii):

$$\frac{e_v}{a} = f_{Pt}e_v^{Pt} + f_{Rh}e_v^{Rh} \quad (ii)$$

where  $f_{Pt}$  and  $f_{Rh}$  represent the atomic fractions of platinum and rhodium in the alloy, respectively, and  $e_v^{Pt}$  and  $e_v^{Rh}$  are the corresponding numbers of valence electrons of elements platinum and rhodium.

The VERs of the alloys were calculated. VER is defined as the ratio of the number of valence electrons to the total number of electrons of the alloy,  $VER = (e_v/e_t)$ , which can be simply calculated by Equation (iii):

$$VER = \frac{e_v}{e_t} = \frac{f_{Pt}e_v^{Pt} + f_{Rh}e_v^{Rh}}{f_{Pt}Z_{Pt} + f_{Rh}Z_{Rh}} \quad (iii)$$

where, following the earlier Equations (i) and (ii),  $Z_{Pt}$  and  $Z_{Rh}$  represent the atomic numbers of platinum and rhodium, respectively, whilst  $f_{Pt}$  and  $f_{Rh}$  represent their corresponding atomic fractions in the alloy.

## 3. Results and Discussion

### 3.1. Average Atomic Number

**Table I** presents eighteen binary platinum-rhodium alloy compositions (31, 32, 48, 49), together with their available mechanical properties including hardness, modulus of elasticity ( $E$ ) and UTS in ascending order as reported in the literature and from the present study. The maximum hardness in the fully annealed alloys (130 HV) corresponds to pure rhodium (maximum rhodium content percentage in the alloy). The lowest hardness corresponds to pure platinum (45 HV). Platinum ( $Z = 78$ ) has a higher atomic number than rhodium ( $Z = 45$ ). This means that by increasing the rhodium content of the alloy, the average atomic number of the binary alloy decreases whereas the hardness increases (**Table I**, **Figure 1**). Similarly, for the available data on  $E$  and UTS of these binary alloys, the decrease in average atomic number of the alloy with increasing rhodium content results in higher values (**Table I**). The values of  $E$  and UTS of platinum with no rhodium content and atomic number  $Z = 78$  increase from  $E = 151.7$  GPa and UTS = 144.7 MPa to  $E = 221$  GPa and UTS = 413.7 MPa, respectively, in a binary alloy

**Table I Number of Valence Electrons ( $e_v$ ), Average Atomic Number ( $Z$ ), Valence Electron Ratio and Mechanical Properties of Binary Platinum-Rhodium Alloys (31, 32, 48, 49)**

Pt-Rh, wt%	Pt-Rh, at%	$e_v$	$Z$	VER	Hardness, HV	E, GPa	UTS, MPa	Reference		
								Hardness	E	UTS
0	0	10	78	0.128	45	151.7	144.7	(31)	(31)	(31)
5	9.1	9.909	74.997	0.132	63	172.4	206.8	(31)	(31)	(31)
10	17.4	9.826	72.258	0.136	79	191.7	292.3	(48)	(31)	(48)
10.01	17.5	9.825	72.225	0.136	80	-	-	(49)	-	-
13	22.1	9.779	70.707	0.138	82	196.5	318.5	(31)	(31)	(31)
15	25.1	9.749	69.717	0.140	86	201.3	343.3	(48)	(31)	(48)
18.1	29.5	9.705	68.265	0.142	88	206.8	355	(48)	(31)	(48)
20	32.2	9.678	67.374	0.144	90	213.7	372.3	(31)	(31)	(31)
25	38.8	9.612	65.196	0.147	97	218	386.1	(31)	(31)	(31)
30	44.9	9.551	63.183	0.151	102	221	413.7	(31)	(31)	(31)
35	50.6	9.494	61.302	0.155	105	-	-		(31, 32)	
40	55.9	9.441	59.553	0.159	108	-	-		(31, 32)	
50	65.5	9.345	56.385	0.166	112	-	-		(31, 32)	
60	74.1	9.259	53.547	0.173	118	-	-		(31, 32)	
70	81.6	9.184	51.072	0.180	122	-	-		(31, 32)	
80	88.4	9.116	48.828	0.187	124	-	-		(31, 32)	
90	94.5	9.055	46.815	0.193	127	-	-		(31, 32)	
100	100	9	45	0.2	130	-	-		(31, 32)	

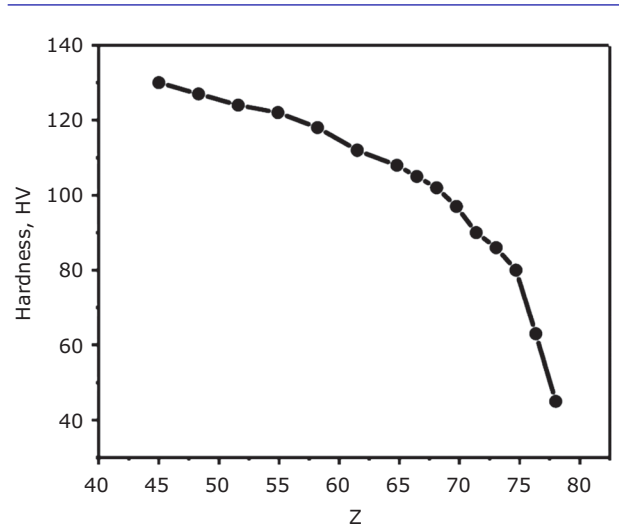


Fig. 1. Alloy atomic number: variations of hardness with average atomic number of binary platinum-rhodium alloys ( $Z$ )

with 44.9 at% rhodium and average  $Z = 63.183$ . Furthermore, the hardness increases from 45 HV to 102 HV by the reduction of average  $Z$  due to increasing rhodium content of the alloy and higher hardness is achieved by increasing rhodium content.

The average atomic numbers of the alloys in this study are tabulated in **Table I**. The mechanical

properties (hardness, E and UTS) of these alloys clearly increase with decreasing average  $Z$  as shown in **Figure 1**. In transition metals and intermetallics, it is known that when the average atomic number is halfway through the transition metal rows of the Periodic Table, i.e., either  $Z = 25-26$ , or  $Z = 43-44$ ,  $Z = 75-76$  and so on, the elastic properties and hardness of the solid solution crystal can be improved to some extent as the properties hit maxima at or around these atomic numbers (46) as a consequence of optimal orbital occupancy. This effect is not observed in platinum-rhodium binary alloys, as in most cases the average atomic number of the alloy is not close to the aforementioned  $Z$  numbers (**Table I**). As shown in the table, the existing platinum-rhodium alloys can have higher hardness, E and UTS with decreasing average atomic number. In other words, the mechanical properties of the alloys improve with decreasing total number of electrons (average atomic number) of the alloy. Non-valence electrons do not contribute to bonding and together with the protons comprise the ion kernels in the metallic bonding of transition metal alloys. Decreasing the non-valence electrons or the size of the kernels at constant (or nearly constant) valence electron numbers leads to stronger elastic bonding (45, 46).

This, in turn, means that the resistance to atomic displacement, crystal shape and volume change will be higher (at least in some crystallographic directions) resulting in the improved mechanical properties. The presence of rhodium ( $Z = 45$ ) compared to platinum ( $Z = 78$ ) in the alloys causes the total number of non-valence electrons to extensively decrease due to its lower  $Z$  value (Table I).

### 3.2. Number of Valence Electrons

Platinum-rhodium alloys with 0 to 100 at% rhodium were included in this study. In transition metal alloys, a wide range of number of valence electrons can be observed ( $3 \leq e_v \leq 12$ ) as categorised by Zarinejad and Liu (36, 44). The alloys were divided into low ( $e_v < 5$ ), medium ( $5 \leq e_v \leq 7.50$ ) and high ( $e_v > 7.50$ ) valence electron groups. Following this categorisation, platinum-rhodium binary alloys all belong to the high valence electron group with  $e_v > 7.50$  in a range between 9 and 10 (Table I, Figure 2).

By increasing rhodium content percentage in the binary alloys, the number of valence electrons of the alloy decreases as the number of valence electrons of platinum ( $s + d$  orbital electrons) is 10 and that of rhodium is 9. By decreasing the number of valence electrons in the alloys, the hardness,  $E$  and UTS are consistently increased (Table I). This indicates the dependence of the mechanical properties on number of valence electrons in the

binary solid solution alloy and that although  $e_v$  is a factor in bonding, its effect must be studied along with other influencing factors such as  $Z$ . Increasing the rhodium content of binary alloys accompanied by lowering  $e_v$  results in a lower average  $Z$  for the alloy crystal. These two opposing trends not only affect the number of valence and non-valence electrons of the alloys but also the dynamics of metallic bonding. Decreasing the number of non-valence electrons because of a decrease in the average atomic number of the alloys, even when accompanied by slight reductions in the number of valence electrons, affects the interatomic bonding that keeps the ion kernels in the metallic bonds together as it affects the size and density of the kernels (41). The higher numbers of non-valence electrons and protons in this condition reduce the bonding effects of the valence electrons and therefore reduce the mechanical properties of the solid solution crystal at least in some crystallographic directions.

### 3.3. Valence Electron Ratio

#### 3.3.1 Hardness

Depending on the atomic fractions of rhodium in the platinum-rhodium alloys, different VER values have resulted. The values for the alloys examined are tabulated in Table I. The variations of hardness vs. VER and vs. atomic percentage of rhodium in the alloy are plotted in Figure 3. A trend can

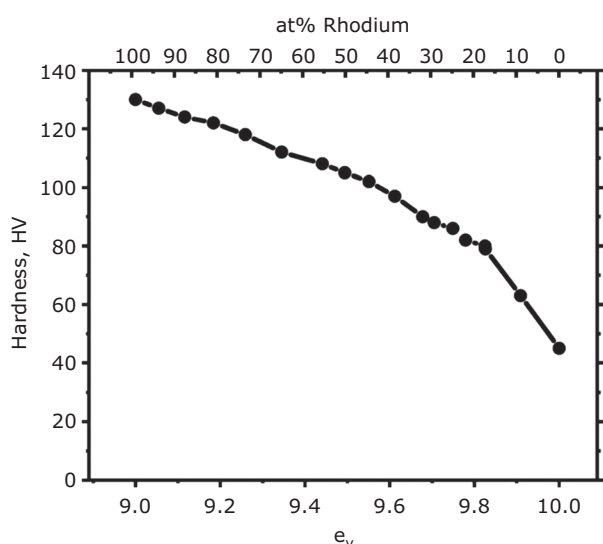


Fig. 2. Hardness vs. number of valence electrons: variations of hardness with number of valence electrons of binary platinum-rhodium alloys ( $e_v$ ) and rhodium content of the alloy (at% rhodium)

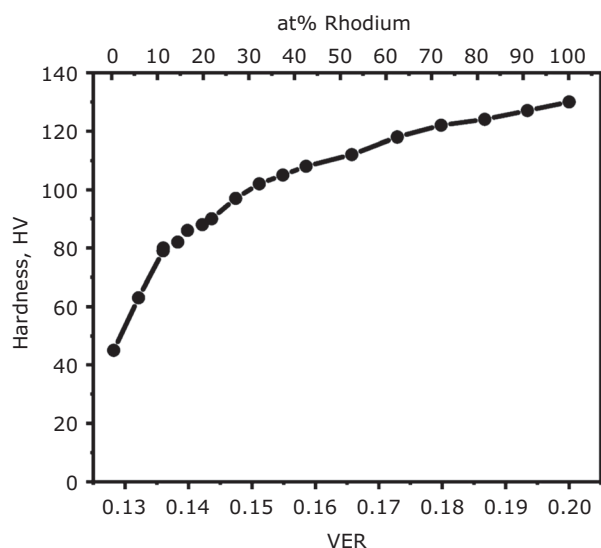


Fig. 3. Hardness vs. VER and rhodium content: variations of hardness with VER and rhodium content of the of binary platinum-rhodium alloys (at% rhodium)

be observed with increasing VER. By increasing rhodium content, the hardness of the binary alloy increases consistently from values as low as 45 HV with VER 0.13 to values as high as 130 HV with VER 0.20. The general trend in the variation of hardness with VER and rhodium content is identical (Figure 3). It is evident that the varying direction of hardness with VER is ascending. VER represents the compound effect of both  $e_v$  and Z.

### 3.3.2 Tensile Strength and Elastic Modulus

Figure 4 shows the variation of E and UTS of the alloys as a function of rhodium content and VER. It is evident that both E and UTS increase with VER, which is brought about by increasing rhodium content of the binary alloys. The gradient of UTS with increasing VER and rhodium content is more pronounced than the gradient of E, although in both cases the variations with VER as it represents chemical composition are remarkably high (Figure 4). It should be noted that microstructural factors including grain boundary characteristics

and defect populations can have a major influence especially on UTS. However, the intrinsic properties of the solid solution are dictated by bonding and valence electron parameters.

### 3.3.3 Creep Rate and Rupture Time

Table II presents the four binary platinum-rhodium alloy compositions and their creep rate characteristics at 1500°C, 1600°C and 1700°C (10, 11). Table III presents the seven binary platinum-rhodium alloy compositions and their stress rupture time characteristics at various test temperatures from 1200°C to 1700°C (10, 11, 29). With reference to the tables, it is evident that the creep rate of the binary alloys at all tested temperatures decreases with VER. This means that with the presence of more rhodium in the binary alloy the creep resistance of the alloy is improved (Figure 5). Moreover, the stress rupture time of the binary alloys at any temperature of creep testing (1200°C to 1700°C) increases with VER and rhodium content (Figure 6). In all the variations of mechanical properties, VER follows the change in

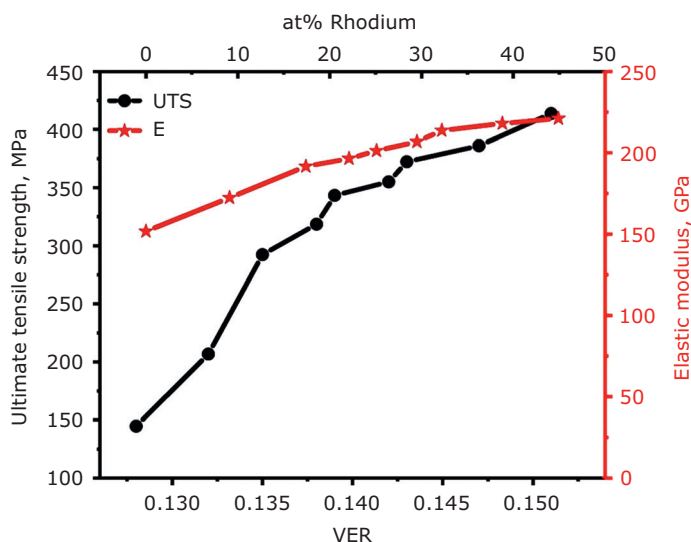


Fig. 4. UTS and E vs. VER and rhodium content: variations of UTS and E with VER and rhodium content of the binary platinum-rhodium alloys (at% rhodium)

**Table II Valence Electron Ratio and Creep Rate of Binary Platinum-Rhodium Alloys at Different Temperatures (10, 11)**

Pt-Rh, wt% Rh	Pt-Rh, at% Rh	VER	Creep rate $\xi$ , % h <sup>-1</sup>			Reference
			1500°C	1600°C	1700°C	
15	25.1	0.140	2.5	5.5	12	(10, 11)
20	32.2	0.144	2	4	9.5	(10, 11)
30	44.9	0.151	0.8	2.5	6	(10, 11)
40	55.9	0.158	0.1	1.3	5	(10, 11)

**Table III Valence Electron Ratio and Stress Rupture Time of Binary Platinum-Rhodium Alloys at Different Temperatures (10, 11, 29)**

Pt-Rh, wt% Rh	Pt-Rh, at% Rh	VER	Rupture time, h						Reference
			1200°C	1300°C	1400°C	1500°C	1600°C	1700°C	
0	0	0.128	1.5	0.2	0.1	–	–	–	(10, 11, 29)
7	12.5	0.133	10	6	3.5	–	–	–	(10, 11)
10	17.4	0.136	26	12	6	–	2	–	(10, 11, 29)
15	25.1	0.140	31	21	11	6	3.5	2.5	(10, 11)
20	32.2	0.144	–	–	–	9	4	2.8	(10, 11)
30	44.9	0.151	–	–	–	17	5	3.5	(10, 11)
40	55.9	0.158	–	–	–	–	6	3.7	(10, 11)

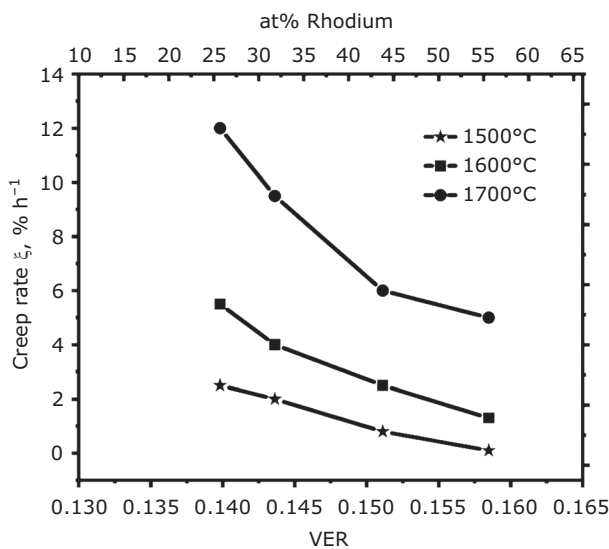


Fig. 5. Creep rate vs. VER and rhodium content: variations of creep rate with VER and rhodium content of the binary platinum-rhodium alloys (at% rhodium) at different temperatures

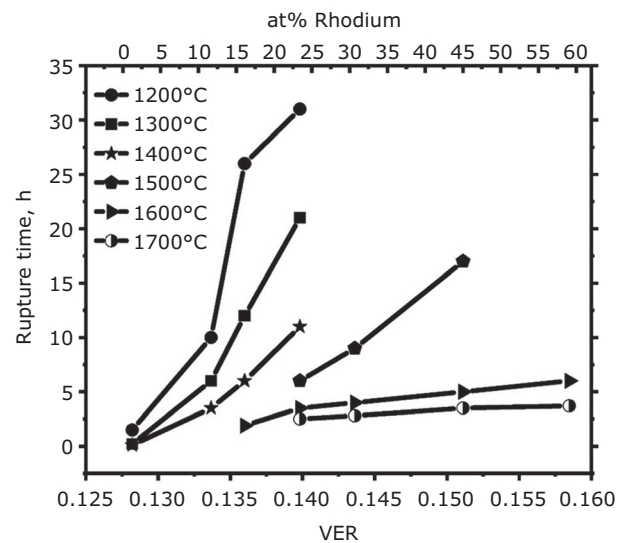


Fig. 6. Stress rupture time vs. VER and rhodium content: variations of stress rupture time with VER and rhodium content of the binary platinum-rhodium alloys (at% rhodium) at different temperatures

rhodium content. Hence, the relationship between VER and mechanical properties of platinum-rhodium alloy is discussed.

In metallic bonding, valence electrons act like 'glue' to bond non-valence electrons and nuclei units together (45, 46), whereas non-valence electrons contribute to the total atomic volume of the alloy. Increasing VER could mean thickening of the 'glue' to bond the ionic kernels together. The elastic response coefficients are the most fundamental of all the properties of solid crystals. The most important subset of properties are the shear and bulk moduli. Bulk modulus is a measure of the resistance of a solid to volume change. Shear modulus is a measure of resistance to shape change. Shear moduli have the highest influence on the mechanical properties of crystals (45, 46).

Furthermore, stacking-fault energy, dislocation mobility and plastic deformation characteristics of crystals are also dictated by metallic bonding (41). Bonding type (metallic, covalent, ionic and molecular) and solidity index of a crystal (which is simply the ratio of the shear modulus to the bulk modulus, multiplied by a coefficient of the order of unity) have been used to classify crystals (46). Hardness, tensile properties and other mechanical properties at elevated temperatures are also dominated by the nature and change in the metallic bonding in transition metal alloys. Metallic bonding is in turn controlled by valence electrons, which are altered by the chemistry of the alloys. The chemical factors that affect the mechanical properties of platinum-rhodium-based alloys in this study are discussed as follows.

An established empirical relationship between the valence electron density and bulk modulus of metallic materials and intermetallic compounds is known. Higher VER usually results in higher bulk and therefore higher shear moduli (46). Increasing VER of the platinum-rhodium alloys, therefore, is expected to result in higher elastic and shear moduli, lower dislocation mobility and more difficult local plastic deformation at least in some crystallographic directions. The change of VER by altering the platinum:rhodium ratio in the alloy is accompanied by a change in the elastic properties, dislocation mobility and stacking-fault energy as the interatomic bonding is affected.

A key factor controlling bonding is the VER of the alloy. When the properties of a solid solution crystal of a platinum-rhodium alloy are enhanced because of higher VER, the crystal's resistance to shape or volume change, atomic displacement and dislocation mobility increases. In this way mechanical properties such as hardness, elastic modulus, tensile strength and creep rate are enhanced. More rhodium content in the alloy increases VER and contributes to improvement of the mechanical properties.

From the work of Okamoto (24), we know that platinum-rhodium alloys are complete solid solutions and second phase precipitation does not take place at any rhodium content. Therefore, all the rhodium contributes to the change in VER and mechanical properties. We assume that impurity trace elements at very low concentrations do not contribute significantly to grain boundary precipitations in the alloys. The comprehensive work of Termic *et al.* (50) has shown that impurity elements might influence the mechanical properties by the formation of grain boundary effects. However, addition of any other element to this binary system that may cause precipitation can change the influence of the VER and warrants further study. The VER dependence of mechanical properties discussed above is limited to platinum-rhodium binary alloys, although it might be extended to other solid solution alloy systems after careful future studies. The trends shown are general and indicative of the major underlying influence of VER on the intrinsic mechanical properties of platinum-rhodium alloy systems.

#### 4. Conclusions

The main correlations shown in this study reveal clear dependence of the intrinsic creep, tensile and hardness properties of binary platinum-rhodium

solid solution alloys on VER. Alloy chemistry influences the number and ratio of valence electrons and average atomic number of platinum-rhodium alloys and thereby changes the mechanical properties of the solid solution alloy crystals. All the mechanical properties improve with increasing VER of binary platinum-rhodium alloys. VER presents itself as a dominant intrinsic parameter that can be utilised to take steps toward successful alloy design to achieve the desired mechanical properties.

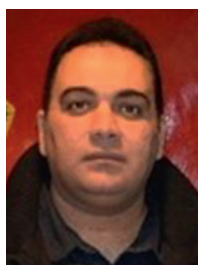
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