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PGMs: A cornucopia of possible applications

by L.A. Cornish

Synopsis

South Africa has been mining PGMs for many years and has a vested interest in the usefulness of these metals, although originally, there was little thought about downstream beneficiation and some of the products were later purchased, in effect repurchasing the raw materials. However, for about 20 years, it has been realised that in order to build wealth sustainably, at least some of the beneficiation and development has to be done in South Africa. This is because for each stage of beneficiation, the value of the commodity increases and so to buy the PGMs back in a later state is uneconomical. Also, the industries need to be established here in South Africa, so that South Africans can work in them and become part of the workforce and generate their own wealth and sustainability for their families. This is part of the rationale of the AMI.

In order to drive this, as well as a viable commercial strategy, with viable and wanted products, there has to be something different in the production mixture, for example, the product has to be better, new or cheaper. This is where research is used to try and develop new capabilities, or to improve them.

Keywords
PGMs, application, product development.

Introduction

South Africa has been mining PGMs for many years and has been the leading supplier of PGMs because of the richness of the Bushveld Complex. Originally, there was little thought about downstream beneficiation, with more emphasis being placed on exporting the metals. Some of the products were later purchased, which is uneconomical, because for each stage of beneficiation, the value of the commodity increases. However, for about 20 years, it has been realised that in order to build wealth sustainably, South Africa must have a vested interest in the usefulness of these metals and that at least some of the beneficiation and development has to be done in locally. Also, the industries need to be established here in South African order to create emploment oportunities, so that South Africans can generate their own wealth and sustainability for their families. This is part of the rationale of the Advanced Metals Initiative (AMI).

In order to drive this, as well as a viable commercial strategy, with viable and wanted products, there has to be something different in the production mixture. For example, the product has to be novel, better, or cheaper than what is available elsewhere. This is where research is used to try and develop new capabilities, or to improve them, or to develop better production routes and methods. Thus the research is important, although it must be remembered that while 'blue skies' research can lead to useful developments, the aim of the AMI is to remain focused on building local industry, so the research must have a perceived and needed impact.

Figure 1 shows some of the applications for which PGMs have been used over the years, some being very long-standing, together with some of the areas that have been researched. Thus, this diagram represents a partial overview of some older applications, as well as some later developments. There is a wide variety of properties and products and it is the basis for a discussion of past projects and the identification of future research areas that could lead to new industries.

Different applications for PGMs

Glass industry

Platinum-based alloys possess high melting points, good thermal stability and thermal shock resistance and good corrosion and oxidation resistance and different approaches to strengthening. The alloys were established by the 1980s (Hammer and Kaufmann, 1982; Heywood, 1988; Heraeus, 2011). There are several applications where the high electrical

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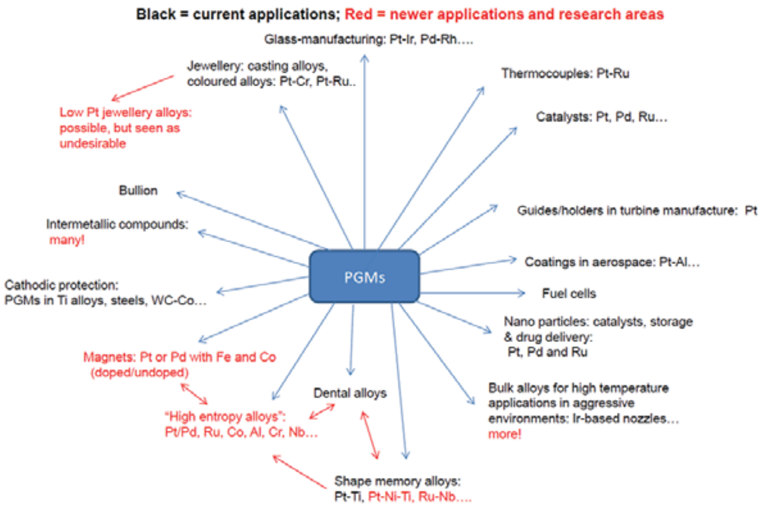

Figure 1—Some areas of applications and research in PGMs

and thermal conductivity of Pt are important (Fischer, 1992). Mechanically, Pt alloyed with rhodium or iridium combine high ductility with adequate creep strength and these properties give the alloys potential for applications in the chemical, space technology and glass industries (Fischer, 2001; Whalen, 1988; Lupton, 1990). In the spacecraft industry, PGMs are used to increase the heat resistance of rocket engine nozzles. A very important application is the manufacture of high-purity optical glasses and glass fibres by using platinum-containing tank furnaces, stirrers and feeders to withstand the high temperatures, mechanical loads and corrosive attack under the glass manufacturing and shaping conditions. In glasses, outstanding purity, homogeneity and the absence of bubble inclusions can be achieved only by using platinum alloys. If ceramic melting vessels were used, ceramic particles would be loosened by erosion, which would contaminate the glass melts and so compromise the desired optical properties such as transmittance. The alloys are expensive, but there are very few other materials that could replace platinum without risk of compromising the product. Thus, this is a valuable and safe use for platinum and some of its alloys.

Thermocouples

Another 'standard use' of PGMs is in thermocouples and this arises because of their high electrical and thermal conductivity, as well as stability due to their high corrosion resistance. Typical alloys are Pt-Rh and Pt-Ru. The thermocouples do eventually degrade in aggressive environments and become brittle, but usually without much impairment of the electrical and conductive properties.

Catalysts

A long-standing use of PGMs is in catalysts and South Africa has established an autocatalyst manufacturing and refurbishment plant for automobiles. The alloys work very well and can be based on Pt or Pd, but are expensive. Competitors would like to replace these expensive alloys with something cheaper, but so far, nothing has been found that matches the good catalytic properties and robustness of the PGMs. There is also added flexibility because of work done with nanoparticles, which has the potential to increase the effectiveness of the catalysts, as the surface areas are increased. Much work has been done in South Africa on catalysts for different applications, both for PGMs, as well as for gold under the Autek project administered by Mintek. However, there is always a challenge with catalysis, because competitors are always looking for cheaper catalysts and there is a risk that the cheapness of other catalysts will offset the greater effectiveness of the PGM catalysts.

The knowledge that palladium could be a ductile permeable storage medium for hydrogen dates from the 1970s (Knapton, 1977) and there is still work being done to optimise the properties, although it is used in hydrogen fuel cells.

Jewellery

Platinum has been used for jewellery for many years. Most of the alloys are 95 wt% Pt, with the other components usually being added to improve the hardness, since platinum is relatively soft. Various methods of surface hardening have been tried (Weber *et al.*, 1996; McGill and Lucas, 1987). Other additions have been made to improve the flowability and hence castability. The most common alloying elements are copper, palladium, cobalt, gallium, iridium, ruthenium and indium. Copper is commonly added to make a general-purpose alloy which casts well and is easy to work, and titanium has also been identified as a possible addition (Biggs *et al.*, 2005). There is a 950 (95 wt%) hallmark for platinum alloys, which means that only 5 wt% is available for any additions. This hallmark is also synonymous with 'quality' and closely guarded by the platinum industry. This situation is different from gold, where there are hallmarks for a wide range of alloys and gold contents. This means that there is much less flexibility in the platinum jewellery alloys, because they have to be 95 wt% Pt to be considered for hallmarking.

Under the Innovation Fund, the Hot Platinum project was established. The aim was two-fold: to develop casting alloys for jewellery applications and also to develop small furnaces suitable for jewellers to use. Although some alloys were successfully developed, it was the furnaces that became more successful and these are still being sold under the Hot Platinum brand, in large and small sizes. This is an example of a successful project.

Cathodic protection

In 1911, Monnartz (1911) found that the corrosion of iron-chromium alloys was much reduced by wrapping platinum wire about the samples, or by alloying with platinum. Subsequently, researchers added PGMs to various alloys to improve the corrosion resistance: Stern and Wissenberg (1959) added palladium to titanium; Tomashov (1958, 1967, 1970) added palladium to steels; Greene (1961) worked on chromium and high-chromium nickel-chromium alloys in sulphuric acids; Biefer (1970) added PGMs to type 430 stainless steel (and found Ru performed better than Pd); Tomashov (1976) added ruthenium to titanium and titanium-nickel alloys; Streicher (1977) added PGMs to stainless steels. Hoar (1960) identified platinum, palladium, rhodium and ruthenium as the best PGMs for cathodic medication, but as platinum and rhodium are expensive, ruthenium is likely to be the PGM of choice in South Africa. Green *et al.* (1961) gave a ranking of iridium, rhodium, ruthenium, platinum and palladium being more effective than osmium, gold or rhenium in maintaining a stable passivity. Although the ranking is not always obeyed, a discernable ranking of the alloying elements for improving corrosion is: Ir > Rh > Ru > Pt > Pd > Os > Au > Re, by comparing the corrosion rates and this was used by Potgieter (1990, 1991) in his discussion of the cathododic modification effect of the PGMs. Tomashov (1980) also looked at additions of 0.1 to 0.4 wt% Ru, Os, Ir, Pt or Pd and Chernova (1980) studied 0.2-0.4 wt% Ru. Tomashov *et al.* (1984) reported that Ru could block lattice defects, as well as enhancing the corrosion resistance in steels. Tomashov *et al.* (1969) added 0.1–0.4 wt% osmium, iridium and ruthenium to chromium alloys and found that the rate of dissolution in chromium-ruthenium alloys in the active state decreased with increasing Ru content. Tomashov *et al.* (1976) found that 0.2 %Ru addition improved the corrosion resistance of Ti-Ni alloys. Streicher (1977) found that Ru additions of 0.2 wt% conferred better corrosion resistance than the same amount of Pd. More recently, work in this area has increased, for example, Zhang. (2009) and Olaseinde *et al.* (2012), Potgieter (1995), to explain the effect, Van der Lingen and Sandenbergh (2001), and there is also work on ion implantation of PGMs in an attempt to retard stress corrosion cracking. Shing *et al.* (2001) added ruthenium to WC-Co alloys as a strengthener, with improved results and this also has potential for improving the corrosion resistance.

Bulk alloys for potential application for turbines and the aerospace industry

Nickel-based superalloys (NBSAs) have excellent mechanical properties due to precipitation strengthening of small, stable, ordered particles and have long been the 'standard' for aerospace alloys, although now lighter titanium alloys are being targeted for some of the components at lower temperatures. Platinum has similar chemistry to nickel and so reacts similarly with the alloying elements and has been investigated as a potential substitute in higher temperature alloys in even more aggressive environments (Coupland *et al.*, 1980; Fischer *et al.*, 1997, 1999a, 1999b, 2001; Völkl *et al.*, 2000; Wolff and Hill, 2000). The potential of using an fcc PGM analogue was mooted several times (Bard *et al.*, 1994), in NIMS, Japan owing to the good properties of iridium-based and rhodium-based alloys (Yamabe *et al.*, 1996, 1997, 1998a, 1998b, 1999; Yu *et al.*, 2000), Pt-Ir alloys (Yamabe-Miterai *et al.*, 2003), Pt-Al-Nb and Pt-Al-Ir-Nb alloys (Huang *et al.*, 2004) and platinum-based alloys in South Africa (Wolff and Hill, 2000). The PGMs and nickel have similar structures (mostly fcc) and similar chemistries for the formation of similar phases. Advantages of PGM-based alloys over the NBSAs are the increased melting temperatures (*e.g.* 2443°C for iridium, 1769°C for platinum compared to 1455°C for nickel) and the excellent corrosion properties. Although platinum-based alloys would have been unlikely to replace all NBSAs on account of their higher price and higher density (Pt has a density of 21.5 g.cm^{-3}, compared to nickel's density of 8.9 g.cm^{-3}), they were identified as having potential for use in components subjected to the highest temperatures. For Pt-based alloys, an increase in application temperature of at least 200°C could be gained (and more for Ir-based alloys). Although changes in engine design could be necessary, the higher application temperatures could offset the increased density and expense and the alloys could be recycled.

Most PGMs have the face-centred cubic (fcc) structure, apart from ruthenium, which is hexagonal close-packed (hcp or cph). Iridium has a higher melting point than platinum, but has the disadvantage of brittleness (Panfilov *et al.*, 2008) and is also in shorter supply. Thus, platinum is the preferred alloy base among the PGMs in the most extreme environments in terms of elevated temperatures, aggressive atmospheres and higher stresses (Wolff and Hill, 2000; Hill *et al.*, 2001a; Cornish *et al.*, 2003). Concerning coatings on these alloys, investigations have indicated that either no coatings would be necessary, or at least simpler coatings could be used than those currently used on nickel-based superalloys (Cornish *et al.*, 2009a, 2009b; Douglas *et al.*, 2009). Three major ranges of Pt-based alloys have been developed. One is based on Pt-Al-Cr-Ni (Hüller *et al.*, 2005; Wenderoth *et al.*, 2005, 2007; Rudnik *et al.*, 2008; Völkl *et al.*, 2005, 2009) and two are based on Pt-Al-Cr-Ru (Cornish *et al.*, 2009a, 2009b; Douglas *et al.*, 2009). For the Pt-Al-Cr-Ru alloys, one set was more malleable but less resistant to extreme chemical environments, whereas the other had higher chemical resistance, but was more difficult to form. The alloys showed very favourable mechanical properties (Süss *et al.*, 2002). No alloys were produced commercially and the major problems was to identify a suitable application where the density of the alloys would not compromise their use, as they were too dense for the current designs of turbine engines. Another disadvantage is that they are extremely expensive. However, within these restrictions, the alloys could have potential as coatings on other lighter, more affordable substrates.

Under the Platinum Development Initiative (PDI), much work was done to prove the potential of a Pt-based alloy and some alloys were developed that showed very good properties (Cornish *et al.*, 2003, 2009a, 2009b; Cornish and Chown, 2011; Douglas *et al.*, 2009; Potgieter *et al.*, 2010), which were mainly targeted for land-based turbines and were based on Pt-Al-Cr-Ru in South Africa, with German colleagues in the collaboration preferring to base their alloys on Pt-Al-Cr-Ni. The reason for their choice was a reluctance to use ruthenium because of its ability to form RuO_2, RuO_4 and RuO_3 (Caston, 1965) and it is the latter which has the highest partial vapour pressure, which was seen as a concern for long-term stability because of its volatility. The alloys performed well, but much better-supported parallel work outside of South Africa produced some lighter alloys which,.although they initially had stability problems, became better designed and were identified as having better potential in aerospace than the much denser Pt-based alloys, which would increase the momentum and hence the required strength in the moving parts of a turbine. Additionally, more work was done on the Ni-based superalloys which resulted in an increase the temperature range, although only by about 100°C. Most of the improved temperature range was due to the improved coatings (which provide a thermal barrier), as well as the design of turbines which employ forced cooling. The Pt-based alloys did have better properties at higher temperatures than the Ni-based superalloys and the volime fraction of the precipitates was increased during the course of the research to become closer to that of the NBSAs (Shongwe *et al.*, 2010). The PDI work also allowed collaboration with Cambridge University, UK (Fairbank *et al.*, 2000; Hill *et al.*, 2001a) and NIMS, Japan (Hill *et al.*, 2001b). However, this work was changed to focus on coatings and some interesting coatings with potential were developed. The bulk alloys remain and could be used for applications where density is not a problem, possibly in reactors for high temperature and corrosive environments.

One of the interesting outomes from the work was the understanding of why there were two phase diagrams available for the base Pt-Al system (McAlister and Kahan, 1986; Oya *et al.*, 1987). The diagrams differed in the shape of the platinum-based solid solution and well as the number and transformation temperatures of the Pt_3Al phases. These differences were significant because they would affect the stabilities of the strengthening Pt_3Al phases, as well as the amounts that could be produced. The German workers in the PDI collaboration (Fischer, 1992, 2001; Fischer *et al.* 1997 1999a, 1999b; Völkl *et al.*, 2000; 2005, 2009; Hüller *et al.*, 2005; Rudnik *et al.*, 2008; Vorberg *et al.*, 2004, 2005; Wenderoth *et al.*, 2007) preferred McAlister and Kahan's (1986) diagram, while the South African workers preferred the McAlister and Kahan (1986) diagram for most of the phase boundaries, but Oya *et al.*'s (1987) diagram for the temperatures of transformation of the Pt_3Al phases. The structure of one lower temperature phase was derived (Douglas *et al.*, 2007) and showed some similarities to the modelled phase of Chauke *et al.* (2010). The only difference was the purity of the raw materials: those of the Germans were much purer (Cornish and Chown, 2011). This was an important observation, because the purity differences in the were small (less than 0.1 at.%), but the affect was significant.

Later work (Odera *et al.*, 2015) looked at reducing the Pt content without compromising the properties and substituted vanadium and/or niobium for some of the Pt, which could potentially increase the application temperature as well. An important outcome of this research was the development of a better method of etching the Pt-based alloys (Odera *et al.*, 2012), which allowed the microstructure to be better resolved. This has improved the understanding of the microstructure-property relationship for different samples and has helped to determine the phase diagrams.

Before the PDI work, Pt wires had been used to stabilize the position of components and Pt-Al coatings are used in the complex coatings of the nickel-based superalloys (Purvis and Warnes, 2001), because of their high temperature stability and strength. Some work continued on this using the experience derived from the bulk Pt-alloys.

Shape memory alloys

Shape memory alloys can change their shape at a specific temperature and are used in many applications: sensors, temperature-sensitive switches, force actuators, fire safety valves, orthodontic wires, fasteners and couplers. The most well-known example is NiTi, commercially known as Nitanol. Biggs *et al.* (2003) showed that the addition of platinum increases the transition temperature of the alloy, which would allow different applications. By varying the Pt content, the transition temperature for the shape memory alloy can be varied between room temperature and 1000°C and so alloys could be developed for niche applications at specific temperatures. There is potential for adding a ternary component to the Pt_3Al and PtTi phases and the $PtFe_3$ phase has been recognized for its potential for low temperature applications. Work has been done at Mintek, the University of the Witwatersrand and the CSIR, but no alloy has been commercialized yet from South Africa. It is likely that it would be better to identify the niche, derive the required properties and then select the most likely alloy.

PGM shape memory alloys could be used for dental applications, using the noble properties as well. In Germany, palladium is not permitted in dental alloys and so the target market must be assessed.

Magnetic materials

Magnetic materials are essential in many industries and there is a need to find substitutes for expensive rare earth metals. Historically, patents held by American and Japanese companies have been a barrier to entry to the magnet market, but China's establishment of research institutes in the 1950s and 1960s and the expiry of the American and Japanese patents in the 1980s, has enabled China to enter and dominate the magnet market. Currently, China has a monopoly on the magnet value chain, producing 90% of the world's permanent magnets. China also has most of the world's rare earth resources.

One of the new magnetic materials being investigated is the Fe-Co-Pd alloy, (Vokoun *et al.*, 2005) which has potential applications in biotechnology, electronics and green technologies (hybrid vehicles and direct drive wind turbines). Magnetic thin films and nanostructures that are magnetized perpendicular to their surface are essential to many developing technologies, including spintronics devices (Mangin *et al.*, 2006) and patterned media (Todorovic *et al.*, 1999; Ding and Adeyeye, 2013), especially because of the need to maintain thermal stability as device dimensions are shrinking increasingly into the nanoscale. Multilayer or superlattice structures consisting of alternating ferromagnetic and non-magnetic layers are highly suitable for these applications due to their tunable perpendicular magnetic anisotropy (PMA) and saturation magnetization (Terris and Thomson, 2005; Rippard *et al.*, 2010).

By combining the soft magnetic properties of iron and the hard magnetic properties of cobalt and palladium, an optimal magnetic material could be developed (Vokoun *et al.*, 2005). Fe-Co alloys have a B2 structure which makes them extremely brittle at room temperature (Matsuda *et al.*, 2016). The workability of this alloy can be improved by substitution of nickel or palladium. Substitution of palladium leads to a remarkably high tensile strength and elongation at room temperature (Matsuda *et al.*, 2016).

Apart from the mechanical properties of the Fe-Co-Pd alloy and research on the magnetic shape memory properties, little work has been done on the ternary phase diagram. In order to understand the potential alloys better, the ternary phase diagram is needed, so that the compositions can be tailored to give the optimum phases for the desired properties.

Conclusions

The PGMs have a wide range of uses. Some of these are well established, but others are new. Although the PGMs show excellent properties, because of their high cost there is always the risk of substitution, even by materials with slightly inferior properties. However, there are newer potential uses and these should be targeted, first by research and then if successful, more commercially. The additions of small amounts of PGMs to other materials is worth pursuing and probably one of the most exciting applications are magnets containing Pt and Pd.

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