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HANDBOOK OF PRECIOUS METALS

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English Edition Editor
A. Prince, F. Eng.

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PREFACE

The scientists and practicing engineers who are to tackle a number of problems of crucial importance to science and technology and to find out the most promising lines of activity, have, among other things, to ensure that the precious metals—the national wealth of this country—be properly used and saved. These metals—gold, silver, ruthenium, osmium, rhodium, iridium, palladium, and platinum—are unequalled for a variety of physical and chemical properties and have found wide use as catalysts accelerating chemical reactions, corrosion-resistant and high-temperature structural materials, sensors generating electric signals, high-frequency reflectors, etc.

In recent decades the precious metals, their alloys, and compounds have been coming into increasing use in industry, and hence gold, silver, platinum, and iridium are now in short supply and have to be used more efficiently and replaced, where possible, by less scarce materials.

Information on the precious metals is found, both in the USSR and elsewhere, in a scattered literature sometimes containing conflicting opinions, which presents difficulties concerning the application of the precious metals and the designing of new precious-metal-based

alloys. This book is the first attempt to collect and systematize the most comprehensive and reliable data on the precious metals, their alloys, and compounds. The body of data varies considerably from metal to metal because different metals have been studied to varying degrees.

The book covers, in succession, the historical background, mineral resources, and the prospective applications of the precious metals, their physical and chemical properties in the liquid and solid state, and the constitution diagrams of binary, ternary, and multicomponent systems. Adequate space is given to the phase equilibria in precious-metal-based systems. The characteristics and service properties of the alloys that have the greatest potential are discussed. Material has been drawn from unclassified publications by Soviet and other authors.

This book will undoubtedly help in a more rational use of the precious metals.

E. M. Savitskii

NOMENCLATURE

a, b, c	lattice parameters (nm)
a	thermodynamic activity; thermal diffusivity ($\text{m}^2 \text{s}^{-1}$)
B	magnetic flux density (T)
B_d	magnetic flux density in working gap at maximum magnetic energy (T)
B_r	residual flux density (T)
$(BH)_{\text{max}}$	maximum magnetic energy (J m^{-3})
c	concentration (wt. %, at. %)
C	molar heat capacity ($\text{J mol}^{-1} \text{K}^{-1}$)
C_p	specific heat ($\text{J kg}^{-1} \text{K}^{-1}$)
d	density (kg m^{-3}); film thickness (nm)
D	diffusion coefficient ($\text{kg m}^{-2} \text{s}^{-1}$); bulk modulus of elasticity (Pa)
DPHN, BHN, RHN	diamond-pyramid (Vickers), Brinell, and Rockwell hardness numbers, respectively (Pa)

E	interaction energy (J); modulus of elasticity (Pa); activation energy for compound formation (eV); potential (V); electrical field strength (V m^{-1}); thermal electromotive force (thermo emf) (V)
E_{pm}	primary electron energy at σ_{\max} (eV)
E_z	zero-charge potential (V)
$E_{z.t.c.}$	zero total charge potential (V)
E_0	standard electrode potential (V)
F	Lorentz force
G	Gibbs free energy (J); shear modulus (Pa)
H	enthalpy (J); magnetic field strength (A m^{-1})
H_c	coercive force (kA m^{-1})
H_d	coercive force at maximum magnetic energy (kA m^{-1})
H_μ	microhardness (Pa)
i_0	exchange current (A m^{-2})
I	current (A)
j	current density for thermionic emission (A m^{-2})
k	index of absorption
L	Lorentz number
M	magnetic moment (μ_B)
n	index of refraction; concentration of charge carriers (m^{-3})
N	mole fraction
p	pressure; vapor pressure (Pa)
Q	internal energy (J)
Q_m	heat of fusion (J mol^{-1})
Q_s	heat of sublimation (J mol^{-1})
r	residual electrical resistance
R	reflectance
R_H	Hall constant ($\text{m}^3 \text{C}^{-1}$)
S	entropy (J K^{-1}); absolute thermoelectric power (V K^{-1}); sensitivity of strain gauge
T, t	temperature (K, $^{\circ}\text{C}$, respectively)
T_c	critical temperature for superconducting transition (K)
T_c, θ_c	Curie temperature (K)
U_c^1	first critical potential (V)
W	radiant heat flux density (W m^{-2})
W^0	radiant heat flux density from a perfect black body (W m^{-2})
α	linear (volume) thermal expansion coefficient (K^{-1}); temperature coefficient of resistance (TCR) (K^{-1})
γ	coefficient of thermodynamic activity; stacking fault energy (eV)
δ, El, e_1	elongation (%)
ϵ	integral emissivity
ϵ_n	normal emissivity
$\epsilon(\lambda)$	spectral emissivity
$\dot{\epsilon}$	creep rate
η	dynamic viscosity (Pa s)
η_{\max}	maximum coefficient of inelastic electron reflection
λ	thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$); wavelength (m)
μ	the Poisson ratio

μ_e, μ_h	Hall mobilities of electrons and holes, respectively
μ_{rev}	reversible permeability
ν	kinematic viscosity ($m^2 s^{-1}$)
Π	Peltier coefficient (V)
ρ	electrical resistivity (Ohm m)
ρ_c	contact resistivity (Ohm cm^2)
σ	surface tension ($J m^{-2}$); electrical conductivity ($S m^{-1}$) (S, siemens)
σ_a	cross-sectional area for thermal neutron absorption (b)
$\sigma_b(S)$	cross-sectional area for thermal neutron scattering (b)
σ_{max}	maximum coefficient of secondary electron emission
σ_T	Thomson coefficient ($V K^{-1}$)
σ_u	ultimate tensile strength (UTS) (Pa)
σ_y	yield stress (yield strength, yield point) (Pa)
$\sigma_{0.2}$	0.2% proof stress (Pa)
$\sigma_{0.001}$	0.001% proof stress (Pa)
φ	electron work function (eV)
φ_b	potential energy barrier (eV)
χ_m	specific magnetic susceptibility ($m^3 kg^{-1}$)
χ_v	volumetric magnetic susceptibility
ψ, q	reduction of area (%)

HISTORICAL BACKGROUND

GOLD

At the dawn of human history, when man, who subsisted chiefly by gathering and hunting, made a cult of objects and attributed magical potency to them, he chose gold as an important object of worship. Bright and shining, so different from common stones, native gold appeared to be the sunlight incarnate. This image of gold was embodied in the names given to the metal by different peoples [1-6].

The first workings of gold date back to the late Stone Age; all the earliest civilizations were familiar with gold. Thus, gold objects were known to the ancient Egyptians twelve-thousand years before Christ. The ancient Egyptians, Greeks, and Romans were highly skilled in dressing gold ores, extracting gold from its ore, and refining and working the metal [1-6]. Interestingly enough, the technique of producing gold leaf (0.01 to 0.001 mm thick), used by metallurgists of ancient Egypt, was not redeveloped in Europe until the 18th century A.D. In Egypt under Thutmose II, in the middle of the second millennium B.C., the output of gold was up to 50 tons a year [2, 4].

Beginning in the third millennium B.C., placer deposits were worked for gold on a substantial scale, not only in Egypt, but also in South and West Africa, which nowadays is still the leading producer of gold.

Another important gold-producing region in the ancient world was Western Europe. In the first millennium B.C., the Romans mined up to eight tons of gold a year in their Iberian colonies. Gold coins became the principal instrument of money circulation in Europe. In the sixth century B.C., gold placers were exploited on the Balkan Peninsula. By the fifth century B.C., the gold deposits in Europe were practically worked out.

In Asia, evidence of the mining of gold in ancient times has been found in Iran, in the coastal area along the Persian Gulf, and in Arabia. Beginning in the third millennium B.C., gold was mined in considerable amounts in India and Tibet. In China, coins were minted from gold as early as the second millennium B.C. In the first millennium B.C., placer mining was started in Baluchistan (Pakistan), Kashgaria (China), Western Siberia, the Near-Amuria, Central Asia, and Asia Minor.

Archeological findings suggest that the main gold mining areas and sites in America were in the territories occupied now by Mexico, Guatemala, Colombia, Panama, Peru, and Cuba, with a total of about 200 tons of gold produced in pre-Columbian times [2-4]. Of the approximately 80 thousand tons of gold produced throughout the ages of human history, some 15% was extracted in ancient times. By comparison, from the fifth to the 15th century, i.e., before the discovery of America, the amount of gold production did not exceed 2500 tons. Later (after the 15th century), the bulk of the world's gold supply was contributed by America.

Within the present borders of the USSR, gold mining was also started in extreme antiquity. There is archeological evidence that gold deposits were worked in the Altai in the middle of the fourth millennium B.C. Gold objects were known to the Caucasian peoples three-thousand years B.C. Gold articles of the Bronze Age have been found in Kazakhstan, Siberia, the Urals, and the Trans-Volga region.

No gold mining existed in Kievan Russia, and gold for coinage and jewelry was bought in the Byzantine Empire. Gold mining in Russia is reckoned from the early 18th century. Gold was first extracted as a by-product from auriferous silver ores at the Nerchinsk works in the Trans-Baikal region and at the Kolyvan-Voskresensk works in the Altai. In 1745, a gold placer was found by the peasant Erofei Markov not far from Ekaterinburg (Sverdlovsk). By the late 18th century, several dozen gold lodes were discovered in the Ural, Altai, and Trans-Baikal territories. Before 1814, as little as 21 tons of gold was produced in Russia.

In 1814, the discovery, by L. I. Brunsitsyn, of the method for concentrating the gold content of auriferous gravels and the repeal of the state monopoly gave a big impetus to gold-mining activities all over the Urals, Western Siberia (1827), the Altai (1830), Yakutia (1840), the Trans-Baikal region (1832), Amuria (1850), and the Primorski Territory in the south of the Pacific coast of Russia (1870). Russia became the leading gold-producing country [4]. After 1850, the contribution of Russia to the world's gold supply decreased sharply following the discovery and development of the California gold fields. Next, gold was found in Australia and America (Colorado, North Dakota, and Nevada). In the late 19th century, South Africa took the lead and has since been the principal producer of gold among the capitalist countries. It should be noted that, during the 20th century, the world output of gold was markedly affected by the overall economic situation in the capitalist countries. By 1940, the capitalist countries produced 1140 tons of gold a year. This figure exceeded 1380 tons in 1970 and decreased perceptibly during the ensuing years.

SILVER [6-8]

Of the precious metals, silver is the most abundant in nature and constitutes $1 \times 10^{-5}\%$ (by mass) of the earth's crust, i.e., 20 times as much as gold and about as much as the total of the platinum metals. Unlike gold, silver rarely occurs natively, but is most often contained in sulfides of nonferrous metals (zinc, lead, etc.) present in metallic ores.

A brilliant white metal that could be kept for a long time was undoubtedly attractive. While the ancients associated the yellow color of gold with the Sun, the brilliant white color of silver reminded them of the Moon, which was reflected in the names given to this metal. The late Latin name for silver, *argentum*, is derived from the Latin root meaning white, brilliant. Silver was one of the first metals to be used by man, as were gold and copper. Since antiquity, silverware has been handed down in Southern Europe, and the Near and Middle East. Silver bullion and cut pieces of the metal were used as an instrument of exchange.

The earliest large-scale workings of silver were in Eastern Asia Minor some time in the fourth millennium B.C. Later, silver deposits were exploited in Armenia and Bactria (along the upper and middle course of the Amu Darya). Around 500 B.C., Athens began to develop the famous deposits of silver-lead ores at Laurion (Greece), which brought her over 77 thousand tons of silver and were the basis of her economic power for a few centuries. In more recent times (the eighth to 15th century), silver was mined in Transylvania (Romania) and in Bohemia (Czechia).

The discovery of America was followed by a particularly rapid upsurge in silver mining and production. Large and rich deposits of silver were found in Bolivia and Peru.

Within the present borders of the USSR, the ancient silver mining areas and sites were in Transcaucasia and Bashkiria. However, Russia did not have her own precious metal industry before the 18th century. It was not until the 18th century that the exploitation of large deposits of silver started in the Trans-Baikal region, the Altai, Central Asia, and the Caucasus. Russia accounted only for a small fraction (2.92% in 1912) of the world's silver output. In 1900, it was as little as 3240 kg, as compared to 1803 tons in the USA, 1746 tons in Mexico, and 355 tons in Bolivia. The amount of silver produced in tsarist Russia was even too small to meet the home demand.

As more silver was produced, the techniques for its extraction and purification were improved. A major contribution to the development of silver metallurgy was made by Russian scientists and craftsmen. Ivan Mokeyev, a foreman at the Moscow Mint, was the first to succeed in separating gold from silver. In 1721, medals were specially struck in gold and silver, using the metal produced by his method.

In 1752, Academician U. Kh. Sal'khov suggested that nitric acid be employed to separate gold and silver. Ivan Schlatter introduced the technique of separating silver and gold by pyrometallurgy followed by the hydrometallurgical treatment of the resulting alloy of gold with silver with nitric acid. This combined technique of refining was used at the St. Petersburg Mint until 1820.

The basic theory of the cyanide process was developed by P. R. Bagration, who showed that gold and silver are readily soluble in dilute aqueous solutions of alkali cyanides. He also found that air accelerates the dissolution of gold and silver and demonstrated that gold and silver can be precipitated from the cyanide solution by base metals.

In 1802, D'Arcet, a French investigator, devised a technique of refining silver with sulfuric acid. The crude silver is dissolved in boiling cupric sulphate, the gold precipitates to the bottom, and the silver is reduced to silver metal by copper or iron. This process was widely

used in the 19th century to give way subsequently to electrolysis, by which 99.9% pure silver can be obtained from an anode of crude silver.

THE PLATINUM METALS [6, 8-17]

Archeological evidence reveals that man first came across platinum millenia ago. Found along with gold, nuggets of platinum were known as "white gold" (Egypt, Spain, and Abyssinia), "frog's gold" (Borneo), or "white lead" (Eastern Siberia) [6]. Even in ancient times, mirrors, articles of jewelry, ornaments, and household utensils were made from platinum and alloys of platinum with gold (Egypt, 700 years B.C.; the Iberian colonies of Rome, one millenium B.C.; the Inca Empire in the north of Ecuador, Columbia).

Platinum had been forgotten by the beginning of our era and was not recognized until after the Spanish conquest of South America. In the European medieval writings, platinum was first referred to as a "bright, unmeltable" metal by J. C. Scaliger, an Italian author, in 1537. But is Dr. Wood who is acknowledged to be the discoverer of platinum (in 1749). The first scientific description of platinum was published by Watson in the Proceedings of the Royal Society (England) in 1749-1750.

In the late 18th century, the heavy white metal came to the increasing attention of chemists. Coincident with the detailed study of the metal, the development of methods for making malleable platinum was begun. Pure platinum was first obtained in 1803 by Wollaston, who also developed the first method of producing malleable platinum on a commercial scale [11]. A comprehensive study of the platinum ore enabled him to discover two more elements—palladium and rhodium—during the same year. In 1804, Tennant extracted iridium and osmium from the black sediment the platinum ore leaves on dissolution in aqua regia [12]. It was not until 40 years later that the remaining metal of the platinum group was extracted from the Uralian platinum ore by K. K. Klaus, Professor at the Kazan University, who chose for it the name ruthenium, derived from Ruthenia, which is the Latin for Russia.

In Russia, platinum was first discovered in gold fields in 1819. The first alluvial deposit of platinum of commercial importance was found in the Urals in 1824 by the superintendent of the Goroblagodatskii Mines, N. R. Mamyshev. Also, his studies on the refining of platinum and the manufacture of articles from the metal were the first works on the subject in Russia. The bulk of the world output of the platinum metals had come from the Urals until a method was developed, in 1936, to extract these metals from the slimes resulting from electrolytic refining of copper or nickel.

Russian scientists have made a major contribution to the development of the platinum industry, and the chemistry, metallurgy, and physical chemistry of the platinum metals. Of great importance to the advancement of the precious metal industry was a fairly simple and safe procedure for refining platinum and making malleable platinum worked out by P. G. Sobolevsky [14]. His paper on the production of malleable platinum was the first learned treatise on powder metallurgy. A contemporary of Sobolevsky, K. K. Klaus, was the founder of the chemistry of the platinum metals (19th century); he devised methods for separating and refining the platinum metals. Pure platinum and rhodium were commercially produced by these methods up until 1929 [16]. Beginning in 1879, Ural platinum was refined in production quantities at the Tentelevskii Works in St. Petersburg. The refining operations were in the charge of F. V. Vilm, who also developed a method of obtaining palladium from palladous ammine chloride formed at an intermediate stage of the process. This process is still in general use [16].

After the Great October Socialist Revolution, staff members of the Platinum Research

Institute and, later, of the Institute of General and Inorganic Chemistry (V. V. Lebedinsky, N. K. Pshenitsyn, S. E. Krasikov, N. I. Podkopayev, O. E. Zvyagintsev, et al.), working in cooperation with industrial workers (Professor N. N. Baraboshkin, A. V. Mironov, N. F. Fedorov, et al.) under Academicians L. A. Chugayev and I. I. Chernyayev, dealt with problems of crucial importance in the refining and separation of the platinum metals and the production of spectroscopically pure gold, platinum, and palladium. With these problems solved, the home demand for the platinum metals was satisfied.

Starting early in 1900, a series of investigations of metallic alloys, specific aspects of their formation, and the influence of various factors on their structure and properties were carried out by N. S. Kurnakov and co-workers [17]. Research in this field was continued subsequently under the guidance of V. A. Nemilov, A. A. Rudnitsky, and later, E. M. Savitskii at the Academy of Sciences of the USSR and are going on now at the Academy, Moscow University, and other institutions in the USSR with the aim of studying the physical and chemical properties of the precious metals and their alloys, to construct the constitution and composition-property diagrams, and to reveal the features of the physicochemical interaction of the precious metals.

Elsewhere, the bulk of research work on the precious metals is done at the special laboratories of Johnson Matthey & Co., Ltd. (Great Britain), Engelhard Chemical Corporation (USA), and also at Forschungsinstitut für Edelmetalle und Metallchemie (West Germany). Among foreign scientists, those to be singled out are E. Raub, W. Obrowski and G. Zwingmann, R. Jaffee and A. Knapton, O. Loebich and T. Harris, K. Schubert, and A. Darling, the purpose of their research efforts being to examine the structure and the mechanical and physical properties of the precious metals, to construct constitution diagrams, and to study the effect of various factors on the brittleness and workability of the platinum metals.

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B1

PRINCIPAL ORES AND MINERALS. PRINCIPAL DEPOSITS.

1.1 GOLD

Three types of gold deposits can be differentiated according to their mode of formation. These are (i) primary (endogenetic) deposits formed due to some kind of plutonic processes, (ii) exogenetic deposits represented mainly by gold placers which have formed due to the destruction of primary deposits, and (iii) metamorphosed exogenetic deposits—gold-bearing conglomerates and sandstones which are the product of transformation of older placers.

Endogenetic Gold Deposits Endogenetic deposits have the most variety in composition. The numerous classifications of these [22] usually account for the depth of formation of the deposit, the influence of the “sulfideness” of the ores, and other features. We mention only two main types of gold deposits, i.e., gold-quartz and gold-quartz-sulfide deposits which have