Evolution of Native Gold under Exogenous Conditions

Z.S. Nikiforova^{a, \vee,} Yu.A. Kalinin^{b,c}, V.A. Makarov^d

^aDiamond and Precious Metal Geology Institute, Siberian Branch of the Russian Academy of Sciences, pr. Lenina 39, Yakutsk, 677980, Russia

^bV.S. Sobolev Institute of Geology and Mineralogy, Siberian Branch of the Russian Academy of Sciences, pr. Akademika Koptyuga 3, Novosibirsk, 630090, Russia

°Novosibirsk State University, ul. Pirogova 1, Novosibirsk, 630090, Russia

^dSiberian Federal University, pr. Svobodnyi 79, Krasnoyarsk, 660041, Russia

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Abstract—A long-term study of the typomorphism of native gold under exogenous conditions gave an insight into its evolution in time and space. The morphology, internal structure, and chemical composition of native gold change depending on the duration of its occurrence under near-surface conditions and on the thermodynamic parameters of the environment. Along with the known facts of gold transformation in the exogenous environment, we consider some of the first identified aspects of the evolution of native gold in weathering crusts and the hydrodynamic, eolian, and anthropogenic environments. Additional attention is given to the transformation of gold in ancient goldbearing conglomerates under lithostatic pressure and in metamorphic strata depending on the P-T environmental conditions. The paper is based on the results of field work, experiments, and analytical studies of the mineralogy of native gold. The objects of study were gold placers of the eastern Siberian Platform, Tuva, and Mongolia, gold-bearing conglomerates of the Timan Ridge, anthropogenic gold placers of the Yenisei Ridge, kaolinite and laterite weathering crusts of Salair, Kazakhstan, and the Republic of Guinea, and the collections of placer gold from the A.E. Fersman Museum (Moscow), the Central Research Institute of Geological Prospecting for Base and Precious Metals (Moscow), the Moscow Mining Academy, and ZAO VNESHMET (Moscow). In the course of experimental studies, we investigated for the fist time the mechanical transformation of gold particles under the impact of sand-air flow in the eolian conditions and under the lithostatic pressure of the overlying strata on ancient gold-bearing conglomerates. We also used a number of mineralogical and geochemical methods to study the typomorphic features of native gold. The evolution of gold under exogenous conditions depends on the ambient environment. Examination of weathering crust has revealed gold nano- and microparticles resulted from the decomposition of sulfides, tellurides, and other unstable gold-containing compounds. Newly formed gold nano- and microparticles in the form of finest crystals, dendrites, and globules are deposited on the surface of primary endogenous gold. The gold formed in weathering crust is spongy and nodular; the fineness of primary endogenous gold increases, the content of impurity elements in it decreases, and a high-fineness porous shell forms at the edges of the gold particles. In the hydrodynamic environment, placer gold, independently of its form (hemihedral, euhedral, interstitial, etc.), flattens and undergoes a simple deformation, but its chemical composition and internal structure change little; they depend on the stage of ore formation and on the mineragenic type of the gold ore source. We have established that the chemical composition and internal structure of gold change during its long occurrence in the environment and under its repeated redeposition from ancient (Precambrian) to younger (Quaternary) deposits. Based on the obtained results of experimental and mineralogical studies, we have proved that eolian processes change not only the shape of native gold but also its chemical composition and microhardness. In the eolian environment, placer gold of different shapes tends to become a globule with a film-fibrous surface. The change in the shape of gold is accompanied by an increase in its fineness, a decrease in the content of impurity elements, and, as a result, decrease in the gold microhardness. In ancient conglomerates (ancient fossil placers), placer gold subjected to the lithostatic pressure of overlying deposits transforms into pseudo-ore gold. In metamorphic strata with constant temperatures and pressures, gold becomes refined. The identified indicators of placer gold of different exogenous environments make it possible to reconstruct the geologic and geomorphologic conditions of gold placer formation, namely, to determine the genetic type of placers (related to weathering crusts, alluvial, eolian, etc.) and to define the source areas (intermediate or primary sources). This helps to find a more correct technique for the search for gold placer and ore deposits.

Keywords: gold evolution; exogenous processes; typomorphism; mineralogical and geochemical features; native (hypergene, eolian, pseudo-ore, and metamorphogenic) placer gold

INTRODUCTION

A long-term study of the typomorphism of native gold under exogenous conditions gave an insight into its evolution in

 $^{\boxtimes}$ Corresponding author.

time and space. The morphology, internal structure, and chemical composition of native gold change depending on the duration of its occurrence under near-surface conditions and on the thermodynamic parameters of the environment.

The goal of this research was to examine the main tendencies of the evolution of native gold under different exogenous conditions: hypergene, hydrodynamic, anthropogen-

E-mail address: znikiforova@yandex.ru (Z.S. Nikiforova)

1245

ic, etc. Along with the known facts of gold transformation in the exogenous environment, we consider some of the first identified aspects of the evolution of native gold under the impact of eolian processes and lithostatic pressure on ancient gold-bearing conglomerates, as well as gold transformation in metamorphic strata depending on the P-T conditions.

METHODS

The paper is based on the results of field work, experiments, and analytical studies of the mineral composition of native gold from different areas. The objects of study were gold placers of the eastern Siberian Platform, Tuva, and Mongolia, gold-bearing conglomerates of the Timan Ridge, anthropogenic gold placers of the Partizanskoe and South Yenisei placer districts of the Yenisei Ridge, kaolinite and laterite weathering crusts of Salair, Kazakhstan, and the Republic of Guinea, and the collections of placer gold from the A.E. Fersman Museum (Moscow), the Central Research Institute of Geological Prospecting for Base and Precious Metals (Moscow), the Moscow Mining Academy, and ZAO VNESHMET (Moscow).

In the course of experimental studies (Filippov and Nikiforova, 1988; Nikiforova and Filippov, 1990), we have first investigated the mechanical transformation of gold shapes under the impact of sand–air flow in the eolian conditions and under the lithostatic pressure of overlying strata on ancient gold-bearing conglomerates (intermediate sources).

Gold of anthropogenic placers was studied by a technique including enrichment of large-volume samples on a Knelson-7.5 concentrator, recovery of each fraction of the concentrate in laboratory, and a detailed study of the typomorphic features of gold by a number of mineralogical and geochemical analytical methods.

All analyses were carried out in the Laboratory of Physicochemical Methods of Analysis of the Diamond and Precious Metal Geology Institute, Yakutsk, and at the Analytical Center for Multi-Elemental and Isotope Research SB RAS, Novosibirsk. The morphology, surface structures, and internal structure of gold particles were studied using a JEOL JSM-6480LV scanning electron microscope, a LEI-CA MZ6 stereoscopic microscope, and a JENAVERT SL 100 ore microscope. The trace element composition of gold was analyzed on a JSA-50A, JSM-6480LV microprobe. The contents of trace elements were determined by atomic-emission spectroscopy. Microinclusions were identified using a JEOL JSM- 6480LV scanning electron microscope equipped with an Energy 350 (Oxford Instruments) energy-dispersive spectrometer. Quantitative analysis and processing of the results were carried out by the XPP method, using INCA Energy Software. The internal structures of native gold were examined by the generally accepted technique (Petrovskaya et al., 1980).

NATIVE GOLD IN A WEATHERING CRUST

The evolution history of native gold in the exogenous environment begins from the moment when endogenous goldbearing parageneses came out to the day surface. The beginning of the formation of a weathering profile (oxidation zone) can be considered the zero point of reference in this history. At present, a weathering crust is understood to be a geologic eluvial body being an integral part of a single hypergene cover permanently forming on land under the effect of numerous factors (tectonic, geomorphologic, climatic, including paleoclimatic, etc.). The activity of these factors governs the formation of eluvial deposits of weathering crusts. The following destruction, transition, and redeposition of weathering crust products leads to the formation of placers and to a relative or absolute enrichment of rock deposits with gold and other stable minerals.

The world's scientific literature concerned with weathering profiles of Australia, the Amazon region, Africa, and southeastern Asia considers "mature" laterite weathering crusts and the behavior of gold in them. The performed studies proved the high mobility of gold in weathering crusts (Mann, 1984; Lawrance and Griffin, 1994; Freyssinet et al., 2000; Townley et al., 2003; Anand and Butt, 2010; Reith et al., 2012; Kalinin et al., 2019).

The novelty of this research is identification of new facts of gold mobility during the formation of both laterite and kaolinite weathering crusts. The studied gold deposits of laterite (West Africa, Republic of Guinea) and kaolinite (Salair, Kuznetsk Alatau, Altai, Kazakhstan, and the Urals) weathering crusts show the presence of not only endogenous primary Au but also hypergene native gold. The latter occurs as newly formed crystals, drusey aggregates, and finest growth



Fig. 1. Morphology of gold from the hypergenesis zone: a, Mustard, b, nodular, c, porous high-fineness shell.



Fig. 2. Morphology of hypergene gold from weathering crusts of the Suzdal'skoe (a-f) and Raigorodok (g-i) deposits (Kazakhstan): *a*, Elongate crystal with newly formed gold on the surface (growth crystal); *b*, intergrowths of isometric crystals; *c*, isometric crystal with numerous growth forms on the surface; *d*, fragment of the surface of gold nanoparticle; *e*, *f*, fragments of crystals (growth faces); *g*, isometric crystal with step growth faces; *h*, scepter-like crystal with growth faces; *i*, intergrowths of isometric crystals.

forms on the surface of earlier formed gold particles (Kalinin et al., 2006, 2009). Hypergene gold in the form of faceted crystals with a smooth shiny surface is described in detail elsewhere (Nesterenko et al., 1985; Colin and Vieillard, 1991; Craw and MacKenzie, 2015).

It is at this stage of the exogenous history that primary gold transforms maximally both in morphology and in composition. The main aspect is not the degree of transformation of primary endogenous gold in a weathering crust but the scale of this process. In regions with global laterite weathering, the hypergenesis zone is often enriched in gold and the gold particles are large. For example, in southwestern India, the average size of gold particles increases from 0.2 mm in quartz veins to 0.5 mm in the overlying laterite crust, and the gold content increases almost five times (Santosh and Omana, 1991). In addition, both the dissolution and the growth of gold particles and the formation of dendritic nuggets are observed (Nair et al., 1987). Coarsening of gold particles is also observed in the weathering crust in the Posse deposit (central Brazil) (Porto and Hall, 1995).

Changes in the morphology and chemical composition of primary gold in a weathering crust depend on the minera-

genic type of endogenous mineralization. In weathering crusts developed after Au-Te and Au-Sb ores, "secondary" mustard and nodular gold forms (Fig. 1a, b). Newly formed gold particles measuring up to few tens of microns and an increase in the gold fineness are observed in the oxidation zones of pyrite and polymetallic ores (Petrovskaya, 1973; Nikolaeva et al., 2015). The newly formed gold, defined as authigenic, is often present as finest (few microns) globules and lamellar crystals (Amosov et al., 1988; Genkin et al., 1994). The change in the morphology of gold particles involves an increase in the gold fineness (on average, up to 1000‰) and the removal of impurity elements (primarily Cu, Pb, Hg, Bi, etc.) from the edge of the gold grains. As a result, a high-fineness gold shell forms, which has a porous structure and differs strongly from the high-fineness dense shell formed under eolian conditions (Fig. 1c).

Study of the morphology of native gold from weathering crusts revealed signs of the growth of late gold on primary gold and traces of dissolution and spiral growth (Fig. 2) (Kalinin et al., 2009, 2018). It was established that "hypergene" gold can have various morphologic forms: crystalline gold particles with clay and Fe hydroxide spots, dendrites,



Fig. 3. Morphology of gold: *a*–*d*, gold from laterite weathering crust (West Africa): *a*, Spherical, *b*, crystal with smooth faces, *c*, spongy gold grown on a massive spherical gold particle, *d*, brain-like gold on an elongate gold crystal; *e*, *f*, high-fineness gold spherules from kaolinite weathering crust (Salair Ridge).

and flaky and wire-like segregations, which are found in an essentially clayey matrix. Films and drop-like segregations of secondary Au are detected in colloform and pisolitic goe-thite-kaolinite aggregates.

The performed microscopic studies showed that gold from the upper parts of the weathering profile (cuirass, spotted zone) has a number of morphologic forms: rounded crystals with smoothed edges (Fig. 3), isometric grains, and ideal globules larger (0.2-0.3 mm) than those in the underlying horizons of the clayey weathering crust and in saprolite (Zhmodik et al., 2012).

During the formation of a gold-bearing weathering crust, sulfides, tellurides, and other minerals containing finely dispersed and nanosized gold dissolve, and the separated gold is deposited. Gold passes into mobile complex compounds, migrates, and is reduced at geochemical barriers (alkaline, reducing, sorptive, etc.). It is deposited on seeds, e.g., carbon particles, sulfides, and Fe and Mn hydroxides, and on primary native gold grains, thus forming "hypergene" gold (Kalinin et al., 2009, 2018). This newly formed gold often constitutes a significant portion of the total gold in a weathering crust.

Thus, the micro- and nanoscale studies of gold from different zones of weathering crusts made it possible to prove the earlier hypotheses by factual material. These studies revealed crystal dissolution and growth structures on the surface of primary endogenous gold at the macro- and nanoscale levels as well as an increase in the gold fineness and a decrease in the content of impurity elements at the edges of gold particles and the formation of a high-fineness (up to 1000‰) shell. In a weathering crust, gold is separated from gold-containing sulfides, tellurides, iron oxides, and other minerals and is deposited at certain geochemical barriers. Gold passes from unstable compounds into a stable dispersed nanosized mineral form, as evidenced by numerous newly formed gold microcrystals on the surface of various seeds and by intergrowths of newly formed gold and chemogenic minerals: limonite, jarosite, goethite, Mn hydroxides, opal, gypsum, clay minerals, calcite, etc.

NATIVE GOLD IN THE HYDRODYNAMIC ENVIRONMENT

In the hydrodynamic environment, gold particles flatten independently of their initial shape. This identified mechanism was first proved experimentally by Tishchenko (1981). It was substantiated that gold supplied from a primary source is projected onto the valley thalweg and is differentiated by size and thickness, depending on its hydraulic size (Fig. 4a, b) (Trushkov, 1971; Filippov, 1991; Izbekov, 1995; Kopylov, 2002). Therefore, the heads of all typical placers bear euhedral, hemihedral, interstitial, and other gold particles of different shapes (Fig. 4a, b), and the tails contain only flattened ones.



Fig. 4. Scheme of placer gold transition: *a*, Into a riverbed, *b*, projection of gold onto the valley thalweg and its differentiation depending on its hydraulic size: *1*, orebody, *2*, angle of metal projection, *3*, coarse-grained, *4*, medium-grained, *5*, fine-grained gold.

The surface of gold particles in the hydrodynamic environment varies from coarse-pitted to finely shagreen (polished). The surface of authochthonous placer gold has imprints of ore minerals and growth faces specific to ore gold. The perfectly rounded alluvial placer gold has a smooth polished surface, but at high magnification (500x–2000x and more) one can see its loose porous structure (Fig. 5c). Globules of authigenic gold (auric chloride AuCl₃, formed by the reaction $3AuCl \rightarrow AuCl_3 + 2Au$) measuring few tens of microns are sometimes observed on the surface of some gold



Fig. 5. Morphology of gold from alluvial deposits: a, Lumpy gold, b, lamellar gold with simple deformation, c, porous surface of gold (500×), d, auric chloride globule deposited on a gold particle.

particles, e.g., flaky gold particles from the headwaters of the rivers of the Kempendyai region (eastern Siberian Platform), where brine solutions circulate (Fig. 5*d*).

The chemical composition and internal structure of placer gold change little in the hydrodynamic environment. They depend on the ore formation stage and the mineragenic type of the gold ore source (Nikiforova et al., 2013, 2018). We have revealed a regularity in the mineralogy of Precambrian placer gold. This gold is mostly fine-grained (0.10-0.25 mm), of high fineness (>900‰) varying over a narrow range of values, and poor in impurity elements (Ag, Cu, and Hg). This is due to the removal of silver and impurity elements from gold and its refinement under exogenous conditions during the repeated gold redeposition from Precambrian deposits into younger ones. As a result, the internal structure of the gold significantly changes, and recrystallization and regrowth structures, deformation lines, and thick (10–30 μ m) high-fineness shells form, which indicate the long occurrence of gold in various exogenous environments. Placer gold of the Mesozoic ore formation stage is less transformed. It occurs as both finely dispersed and coarse (up to 1–2 mm and larger) grains, is of low to high fineness, and contains a number of impurity elements. This gold is characterized mainly by aggregate grains and coarse- and medium-grained internal structures. The revealed indicative typomorphic features of placer gold of two ore formation stages do not change in the hydrodynamic environment (river flow, coastal-beach zones, etc.).

NATIVE GOLD IN ANTHROPOGENIC PLACERS

Native gold in anthropogenic placers and tailings of goldconcentrating plants undergoes certain changes. In recent years, this problem has become urgent because of the growth of practical interest in anthropogenic placers (Fel'dbarg and Zakharova, 1984; Parii and Amosov, 1998; Myazin and Tataurov, 2000; Makarov, 2001; Kovlekov, 2002; Naumov, 2010). Study of gold transformations in anthropogenic placers is of great scientific and practical importance for their repeated mining. The scale and rate of these transformations depend mainly on the applied technology of placer mining and the landscape and climatic conditions.

Gold of anthropogenic placers differs from gold of basic objects in morphology and grain size composition. Finegrained gold accumulates in crushed gold ore; gold in pebbles has the same grain size and weight characteristics as gold in pillar placers. It was also established that lamellar gold is predominant in anthropogenic placers (crushed gold ore) because of its lower hydraulic size and maximum washout from the sluices. For example, in the placer dumps of the Bol'shaya Penchenga River and the middle reaches of the Ederei River, the portion of lamellar gold is 90%, whereas in the original sands it does not exceed 20% (Makarov and Samorodskii, 2018). In the presence of sulfide minerals in placers, iron hydroxides form under the influence of meteoric water. Naumov (2010) described the processes of formation of up to 15 cm thick iron hydroxide crusts in the anthropogenic placers of the Urals as well as high-gold finegravel conglomerates cemented with iron hydroxides.

Under certain conditions, dump gold can be separated, grow, and be redistributed at geochemical barriers. There are hypotheses of the possible rapid natural regeneration of mined-out placers (Freise, 1931; Smirnov, 1955), which is contributed by the activity of organic matter and bacteria (Marakushev, 1997). The scale and mechanism of these processes have not been investigated in detail. When studying gold in dumps of more than 30 anthropogenic placers in the landscape and climatic conditions of central Siberia, we did not observe gold coarsening caused by processes other than amalgamation.

There is much evidence for the influence of mercury on the transformation of gold morphology in dump deposits and tailings of gold-concentrating plants. Mercury was used in sluice equipment for capture of small- and fine-grained gold and accumulated in dumps both in free form and as amalgams. Gold amalgams of anthropogenic placers are characterized by wide variations in the gold and mercury proportions. They differ in color, aggregation state, and composition of mineral impurities and chemical compounds. The above placers also contain liquid metallic mercury in the form of spherical and hemispherical particles with 0.5 to 3.0% Au. Anthropogenic mercury favors intergrowth of natural gold particles and changes in their microrelief, with the formation of cavities and openwork growths (Fig. 6a, b). Native gold in dumps and tailings of gold-concentrating plants is often cemented with iron hydroxides (Fig. 6c). Probe microanalysis showed that the rims of gold particles from the crushed gold ore of placers mined with the use of mercury are several times richer in it as compared with the cores. Note that these particles have a porous or spongy surface (Makarov and Samorodskii, 2018).

Tailings of gold-concentrating plants usually contain mercury-covered native gold (Fig. 6f). There are also metal objects overgrown with gold: copper wire, nails, lead shot, etc. (Fig. 6d, e). Fragments of nonferrous scrap are in most cases covered with gold-containing mercury amalgam.

Thus, the results of study of the typomorphic features of gold from anthropogenic placers lead to the following conclusions: In crushed gold ore, accumulated gold is finergrained as compared with that of original sands and pebble dumps, the portion of lamellar gold is higher, and the weight of gold particles of certain size classes is lower. The changes in the composition and morphology of gold grains in anthropogenic placers and tailings of gold-concentrating plants are mostly due to the influence of mercury used for the extraction of small- and fine-grained gold. Newly formed "anthropogenic" gold is present as ultrafine crystals deposited on various metal particles (wire, shot, etc.) and as amalgamated gold in the form of pseudomorphs intergrown with iron oxides and other minerals.



Fig. 6. Newly formed anthropogenic gold and gold–mercury amalgam in placer dumps and tailings of gold-concentrating plants. a, Aggregates of amalgamated gold particles; b, gold grains of different compositions cemented by amalgam; c, aggregate of gold particles cemented by iron hydroxides; d, gold grown over a fish hook; e, pseudomorph of amalgamated gold after a copper wire; f, mercury-coated native gold from dredging tailings. Materials from Parii and Amosov (1998) (a, c, d, e) and Naumov (2010) (b).

NATIVE GOLD IN THE EOLIAN CONDITIONS

In the eolian conditions, under wind and sand impact, gold particles undergo a mechanical transformation and acquire different forms depending on their initial shape: flaky (toroidal and, then, hollow spherical), lamellar (elevation along the periphery), tabular (disc-shaped), and dendritic (lumpy massive, with rounded protrusions), which was proved experimentally (Fig. 7) (Filippov and Nikiforova, 1988). In contrast to gold from alluvial deposits, the surface of perfectly rounded eolian gold has a dense and smooth microrelief (visible at any magnification). At \times 300 magnification, this gold has a specific film-fibrous surface (Fig. 7*c*). At higher magnification (\times 2000–5000), the surface of eolian gold remains the same: even, dense, and smooth, with scarce pores. The film-fibrous surface forms by the following mechanism: Under eolian conditions, gold is polished by sand grains and stretches out as ultrathin films, which superimpose on each other to form a dense smooth film-fibrous surface (Fig. 7*o*). The same surface was obtained experimentally.

Mechanical and chemical processes affect strongly the mechanogenic transformation of native gold and thus cause changes not only in its grain shape but also in its chemical composition and internal structure (Table 1). A clear regularity has been revealed: an increase in the gold fineness from 900 to 980‰ and more, a decrease in the content of impurity elements, and reduction in microhardness during the transformation of flaky gold into toroidal and, then, hollow spherical ones.

The AAS data show an increase in the gold fineness from 810 to 970‰ (Table 1) during the transformation of a flaky gold particle into a hollow sphere. Flaky gold contains a



Fig. 7. Morphology of gold from eolian deposits (×300): row 1, successive transformation of native flaky gold particles in the eolian conditions (a-c): *a*, Flake with a thin elevation along the periphery, *b*, toroidal particle, *c*, hollow spherical particle with a film-fibrous surface; row 2, internal structure of the native hollow spherical gold particle in cross section (d-f); row 3, successive transformation of the flaky gold during the experiment (g-i): *g*, flake with an elevation along the periphery, *h*, toroidal particle, *i*, hollow spherical particle; row 4, sections of hollow spherical gold particles of different shapes during the experiment (j-l); row 5, colian gold: *m*, lamellar particle with an elevation along the periphery (Witwatersrand), *n*, lumpy particle with signs of eolian processes (Tuva), *o*, experimental film-fibrous surface.

number of impurity elements, such as Fe, Pb, Ni, Cu, Mn, etc., whereas spherical gold contains only Fe, Cu, and Mn.

Local probe microanalysis (Fig. 8; Table 2) showed a fineness of 747 to 780‰ in the core of medium-grained gold flake S-9a and of 950 to 988‰ in the shell. Gold flake P-138

shows a fineness of 814–860‰ in the core and 900–970‰ in the partly transformed rim. The maximum fineness of completely transformed flake 8642 is 990–1000‰. The fineness of hollow spherical gold is also high. For example, the fineness of grain P-60a (17 analyses) varies within 992–

Shape of gold particle	Average mass of gold particles over fractions (mm), mg 0.1 0.25		Average size, mm	Average thick- ness, mm	Average settling velocity, m/s	Surface mor- phology	Fineness, ‰ (range/aver- age)	Impurity elements, wt.%	Gold forging stage	Major internal structures
Flaky	0.009	0.05	0.25–0.5	0.02	5–6	Pitted-tubercular to polished	810–970/890	$\begin{array}{l} Pb = 0.003\\ Cu = 0.05\\ Fe = 0.1\\ Mn = 0.01\\ Pd = traces\\ Ni = traces\\ Hg = traces \end{array}$	Initial	Luder bands in grains; high-fineness shell; partial regrowth
Toroidal	0.012	0.05	0.15-0.2	0.05	7–10	Finely shagreen, polished	920–970/940	Fe — 0.1 Cu — 0.02 Mn — 0.03 Ni — traces Hg — +	Middle	Fine- and medium- grained high-fineness shells, intergranular vein- lets, partial and complete regrowth
Hollow spherical	0.013	0.05	0.1–0.16	0.1	12–17	Finely shagreen, polished	960–990/970	Fe — 0.1 Cu — 0.017 Mn — 0.001	Final	Fine- and ultrafine- grained gold, complete regrowth (decompaction)

Table 1. Typomorphic features of strongly deformed (eolian) gold from the Lena-Vilyui interfluve

1000‰, and that of grain P-60b (13 analyses) is 1000‰. The shells of the hollow spherical particles steadily show a high fineness. There are only few grains that have preserved primary gold with a fineness of 860 to 919‰ in the septum, e.g., grain B-19b. In general, the study of the internal struc-

ture of gold particles and their chemical composition showed a predominance of ultrafine and fine grains, mostly an extremely high fineness, and the minimum content of impurity elements. It is known that the deformation of gold involves its recrystallization with leaching of silver and its following



Fig. 8. Sections of gold particles and fineness measurement points.

Table 2. Local fir	eness of gold	particles, 9	6
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Fineness measurement point	Gold morphology											
	Flaky			Hollow spherical								
				Recrystallized		Regrown						
	S-9a	P-138	8642	B-19a	V-19b	P-141a	P-141b	P-123	P-60a	P-60b		
1	759	973	998	903	860	990	997	1000	996	1000		
2	747	941	996	912	867	N.d.	992	955	996	1000		
3	759	933	998	907	869	N.d.	983	997	999	1000		
4	756	N.d.	984	904	919	990	991	999	1000	1000		
5	757	908	994	912		988	985	996	999	1000		
6	755	920	997	902		996	996	995	997	1000		
7	758	860	996	901	_	980	987	1000	992	1000		
8	769	892	997			999	984	938	992	1000		
9	766	978	1000			980	970	930	999	1000		
10	953	814		_	_	989	995	993	994	1000		
11	783	896				994	985	997	1000	1000		
12	979	806	_			993	995	995	1000	1000		
13	986	956	_	_	_	990	987	N.d.	1000	1000		
14	970	937	_	_	_	920	995	N.d.	1000			
15	974	973	_			932	1000	903	1000			
16	988	974		_	_	994	988	912	1000	_		
17	970	900	_	_	_	998	987	910	1000			
18			_			987	1000	1000	1000			
19	_	_		_	_	_	998	999	_	_		
20	_		_	_	_	_	998	N.d.	_			
21	_	_		_	_	_	991	N.d.	_	_		
22	_		_	_	_	_	1000	N.d.	_			
23	_	_	_	_	_		983	N.d.		_		
24	_	_		_	_	_	996	1000	_	_		
25	_	_	_	_	_		998	1000		_		
26							999	1000		_		
27		_	_	_	_	_		1000	_			
28				_	_	_		1000	_	_		

Note. Location of the analytical points is shown in Fig. 8.

regrowth contributing to the removal of impurity elements. Intense forging of gold during its complex deformation results in ultrathin gold films, which superimpose on each other to form a shell on the globules. This increases the gold surface for active chemical interaction with the medium components. Therefore, the long transformation of gold led to the maximum removal of silver and impurity elements from it. This explains the decrease in the number and contents of impurity elements in gold, the increase in its fineness in the transition from flaky to spherical forms, and the ultrahigh fineness of the gold shell. The process of mechanochemical refining of gold was experimentally proved by Letchman (1979). During his experiments, a refined layer of pure gold formed as a result of repeated microforging and treatment of a gold–copper alloy (≤12% Au) with a weak ammonia solution.

Thus, a mechanogenic impact of sandy material on flaky gold in the eolian conditions leads to the transformation of its morphology as a result of its complex deformation. Each deformation stage manifests itself as a regular change in the internal structure of gold, an increase in its fineness, and a decrease in the content of impurity elements. The flaky gold is fine- to medium-grained and has Luders bands and welldeveloped high-fineness shells. The toroidal and hollow spherical gold particles are mostly recrystallized ultrafineand fine-grained, with distinct high-fineness shells.

The change in the internal structure of gold led to the removal of silver and impurity elements, which resulted both in the refinement of gold and in the reduction of its microhardness, 21 kg/mm² (Fig. 9; Table 3).

The microhardness of hollow spherical gold particles was measured using a PMT-3 microhardness meter. A total of 55



Fig. 9. Points of measurement of the microhardness of hollow spherical gold particles. I-XII, gold samples (see Table 3).

measurements were made in 12 sections of the particles (Fig. 9; Table 3). In four particles (1–4), the microhardness increases from the center of the septum to the edge. Moreover, the microhardness is twice different between gold particles 1 and 2. Different sites of hollow spherical particles 6, 7, and 9 demonstrate the same microhardness. In particle 12, the microhardness grows from the outer part of the edge to the center of the septum. The other four gold particles show no regular distribution of microhardness.

The study of the microhardness of eolian gold revealed its dependence on the changes in the internal structure and chemical composition of gold under exogenous conditions. It was found that seriously deformed eolian gold with a recrystallized structure, high fineness (up to 1000‰), and lack of impurity elements has low microhardness. We have first established the lower limit of the microhardness of seriously deformed hollow spherical eolian gold, 21 kg/mm². In general, the studied eolian gold has low microhardness as compared with alluvial-placer gold. The microhardness of native gold was earlier studied by many researchers (Lebedeva, 1963; Badalova et al., 1968; Petrovskaya, 1973; Popenko, 1982). They established that the microhardness varies, on average, from 40 to 100 kg/mm² and depends on the chemical composition of gold (contents of silver and impurity elements in it) and on its internal structure. As known, Ag and Cu impurities significantly increase the microhardness of gold; Pt, Sn, and Al impurities also cause its drastic increase. The highest microhardness, 100 kg/mm², is specific to lowfineness (550-650%) gold (Popenko, 1982), and the lowest one is 40 kg/mm² (Lebedeva, 1963). It is not ruled out that the low microhardness of hollow spherical gold is due to the decompaction of some sites of its shell and edge. Petrovskaya (1973) found that regrowth structures lead to gold decompaction, stress relief, and removal of silver and impurity elements. The studied hollow spherical gold has decompaction structures and a negligible content of impurity elements, which led to an increase in its fineness to 1000‰ and a decrease in its microhardness.

A vast dispersion halo of toroidal and hollow spherical gold was first discovered in deposits of different ages within the Siberian Platform. This gold occurs both in the head and in the tail of placers. Toroidal and hollow spherical gold particles can serve as a search criterion for a discovery of eolian gold placers. Note that eolian gold does not preserve its morphology under hydrodynamic conditions but becomes flattened (flakes and lamellae).

Native gold transformed in the eolian conditions occurs not only as toroidal and hollow spherical particles but also as more massive grains: lamellar, tabular, thickened along the periphery, trough-, canoe-, and boomerang-shaped, dumbbell-like, and massive-lumpy. All these grains have a film-fibrous surface specific to gold transformed in the eolian conditions. They are 0.25–5.0 mm (and more) in size and 0.1–0.8 mm in average thickness, with an average mass of 0.5–50 mg and an average fineness of 750–900‰. The grain size distribution and chemical composition of this gold depend on the type of the primary source.

Lumpy gold is massive, with rounded and oval protrusions with a film-fibrous surface specific to eolian transformation. The gold grains are larger than 0.25 mm and differ in the average fineness and impurity elements, which reflect the initial composition of native gold in the primary sources. This gold has a coarse- and medium-grained internal structure or is an aggregate grain, with thin shells of high-fineness gold. In general, it is chemically less transformed, with larger grains, and therefore is almost immobile. It is found

Table 3. Microhardness of section elements in gold globules, kg/mm²

Sam-	Section elements										
ple	Edge 1	Septum	Edge 2	Base of	Shell						
				edge 1	edge 2	-					
Ι	$\frac{65.4}{4}$	$\frac{41.2}{2.3}$	$\frac{55}{1}$	_	_	_					
Π	$\frac{25.7}{3.4}$	$\frac{21}{2}$	$\frac{28.6}{1}$	_	_	_					
III	$\frac{41.2}{6}$	$\frac{25.7}{5.4}$	$\frac{32.1}{1}$	$\frac{55}{7}$	$\frac{41.2}{2}$ $\frac{25.7}{3}$	$\frac{23.2}{8}$					
IV	$\frac{46.2}{1}$	$\frac{42.0}{2.3}$	-	$\frac{64.2}{6}$	_	$\frac{57.2}{5}$ $\frac{46.7}{4}$					
V	$\frac{47.3}{4}$	$\frac{32.1}{3}$ $\frac{25.7}{2}$	$\frac{25.7}{1}$	$\frac{55}{5} \frac{23.2}{6}$	_	_					
VI	$\frac{43.0}{1}$	$\frac{41.2}{2}$	-	_	_	_					
VII	$\frac{64.5}{4}$	$\frac{64.5}{5}$	$\frac{64.5}{2}$	$\frac{64.5}{3}$	$\frac{41.2}{1}$	_					
VIII	$\frac{47.3}{1}$	$\frac{47.3}{4}$	$\frac{32.1}{5}$	$\frac{32.1}{2.3}$	_	_					
IX	$\frac{28.6}{1}$	_	-	$\frac{28.6}{2}$	_	$\frac{28.6}{3}$					
Х	$\frac{47.3}{1}$	$\frac{55.0}{2}$	_	_	_	_					
XI	$\frac{36.2}{1}$	$\frac{47.3}{2}$ $\frac{36.2}{3}$	$\frac{36.2}{4}$	$\frac{41.2}{5}$	_	_					
XII	$\frac{47.3}{3}$	$\frac{55}{2}$	$\frac{41.2}{1}$	$\frac{36.2}{4}$	$\frac{32.1}{5}$	-					

Note. The numerator marks the microhardess value, and the denominator, the measurement number.

only in the basal eolian horizon not far from its source. This gold can be present in high contents and thus be of commercial significance. Lamellar and tabular gold particles elevation along the periphery as well as lumpy gold with rounded protrusions were found within the Vilyui syneclise (Izbekov, 1972), Anabar anteclise (Shpunt, 1974), and in the Urals (Ryzhov et al., 1977). Since the eolian features of deflationary gold are less evident, its dispersion halos are not so wide as the halos of toroidal and hollow spherical gold.

Analysis of the literature data on the distribution regularities of toroidal and hollow spherical gold showed that it is widespread on all platforms. It was found in Proterozoic to Cenozoic deposits, in particular, in the east of the Siberian and European platforms and in the North American, South American, African, and Australian platforms (Nikiforova et al., 2005).

Eolian gold occurs in both Proterozoic and Cenozoic deposits. It formed in areas with intense eolian processes in different epochs of the Earth's evolution. It is not only of mineralogical interest but is also valuable for its commercial reserves. For example, in the famous Witwatersrand deposit, eolian gold particles with extremely high Au contents are found in specific sediments (conglomerates) including windkanters and lacking clay material (Minter et al., 1993; Filippov et al., 1994; Safonov et al., 2000). High contents of eolian gold, up to 30 g/m³, were detected in the Devonian conglomerates of an ancient placer of the Timan Ridge (Nikiforova et al., 1991). According to the earlier studies, eolian gold is widespread in Quaternary deposits of all platforms. Gold with signs of eolian transformation was found within the North American Platform, in the Alberta and Abitibi placer deposits (Ontario and Oregon, Canada), and on Alaska, near Cape Nome (Giusti, 1986). Eolian gold was discovered within the South American Platform, in placer deposits of Bolivia, Columbia, Panama, and Equador, and within the southeastern African Platform, in Mozambique, Zimbabwe, and Tanzania (DiLabio, 1988). Gold of this kind was also revealed within the European Platform, in the North Urals (Ryzhov et al., 1977), Baltic Shield (Negrutsa, 1973), and Kola Peninsula (Surkov, 2000), and within the central Russian Platform (Kal'nichenko et al., 1995; Luk''yanenko and Kolpakov, 1995). Eolian gold was found in placers of Mongolia (Zaamar mine) and Tuva (Tanku-Tuva). Since eolian gold particles are not only of mineralogical interest but also occur in high contents, we have substantiated the probable presence of eolian gold placers of different ages in all platforms as well as in Tuva and Mongolia.

NATIVE GOLD IN ANCIENT GOLD-BEARING CONGLOMERATES AND METAMORPHIC STRATA

In gold-bearing conglomerates (ancient buried placers), gold does not preserve its morphology but becomes a pseudo-ore. Gold of this kind forms under the impact of the lithostatic pressure of overlying strata and of horizontal movements on the formed gold placer. As a result, minerals of the host deposits imprint into the gold, and it takes an ore habit, which was proved experimentally (Nikiforova and Filippov, 1990).

Pseudo-ore gold particles measure mostly 0.10-0.25 mm, sometimes have ragged edges or through holes, and are characterized by a high fineness and a transformed internal structure. Pseudo-ore gold often forms aggregates ("intergrowths") with quartz and other minerals, which are not typical of ore gold, because these gold particles are not tightly intergrown with the minerals. Pseudo-ore gold from ancient conglomerates has a coarse-pitted, tubercular, and fine-cellular surface with imprints of minerals from the host deposits (Fig. 10a, b). These imprints differ from the primary imprints of ore minerals in having rounded shapes without sharp protrusions along the pit edges. They also differ from the corrosive relief structures of the hypergenesis zones in having deeper pits, 0.01 to 0.05 µm. The surface of the examined gold particles has angular elongate scar-like dents, grooves, scratches, and sites with a polished mirrorlike surface (Fig. 10c).

The effect of constant temperatures and pressures in ancient conglomerates caused regrowth of gold (Fig. 10*j*) accompanied by the removal of silver and impurity elements, which refined the gold and increased its fineness. Some



Fig. 10. Pseudo-ore gold from deposits of different ages from the eastern Siberian Platform and the East European Platform: row 1, flakes with imprints of minerals of the host deposits: *a*, With ragged edges, *b*, with rounded imprints; *c*, lamella with grooves and scar-like dents on the surface, $\times 25$; row 2, pseudointergrowths of gold particles with: *d*, quartz, *e*, ilmenorutile, *f*, garnet (Timan Ridge), $\times 20$; row 3: *g*–*i*, gold surface with imprints of fine sand particles, formed during the experiment, $\times 25$; row 4, internal structure of gold: *j*, regrowth ($\times 2500$), *k*, numerous deformation lines ($\times 2500$), *l*, granulation ($\times 1100$).

gold-bearing conglomerates contain single gold particles with granulation structures (Fig. 10*l*), which resulted from the replacement of medium-fineness gold (with the removal of silver) and the formation of low-fineness veinlets around high-fineness gold. It was established that horizontal movements cause numerous deformation lines in gold (Fig 10*k*), which were earlier explained by the impact of mechanical processes in the hydrodynamic environment (Petrovskaya, 1973; Nikolaeva et al., 2015).

Pseudo-ore gold was first discovered by us during the study of gold from a Devonian placer of the Timan Ridge (Nikiforova et al., 1991). This placer was buried beneath thick (up to 700 m) deposits as a result of tectonic processes. Therefore, the gold-bearing deposits of the placer were subjected to both vertical and horizontal movements. The lithostatic impact of overlying strata and their displacement led to vertical and horizontal micromovements of fine-clastic

material, which caused deformation of the placer gold particles. The vertical lithostatic pressure made the sedimentary strata compact, and the downwarping of the deposits induced horizontal movements. This led to the displacement of clastic material parallel to the bedding planes. The vertical pressure of overlying strata made minerals of the host deposits imprint into the gold particles, and the horizontal displacement of the deposits resulted in scratches, grooves, and slickensides on the gold particles, up to their break. Deformation of gold particles under lithostatic pressure (vertical and horizontal micromovements) was proved experimentally (Fig. 10g–i) (Nikiforova and Filippov, 1990).

Ore-like gold (Beligeskhai (Izbekov, 1972) and Olenek (Shpunt, 1974) types) was discovered earlier in the Lena– Vilyui and Anabar–Olenek interfluves in the eastern Siberian Platform. We consider it pseudo-ore gold. Such gold was found in all watercourses of the eastern Siberian Platform



Fig. 11. Morphology and internal structures of metamorphogenic gold: row 1, morphology of gold particles: *a*, Flake with ragged edges; *b*, *c*, lamellae; row 2, internal structures of metamorphogenic gold: *d*, *e*, numerous deformation lines, *f*, flake with a thick high-fineness shell.

(Nikiforova et al., 2013). The presence of pseudo-ore gold in alluvial placers indicates that they formed at the expense of gold supplied from ancient intermediate rather than primary sources. Thus, pseudo-ore gold is a marker of the sources of gold in gold-bearing placers.

In the Archean and Paleoproterozoic metamorphic strata, placer gold is present mainly as high-fineness flaky and thin-lamellar particles 0.10-0.25 mm in size with a minor set of impurity elements: Ag, Cu, and Fe (Fig. 11a, b, c). The gold is characterized by recrystallization and regrowth structures, numerous deformation lines, and thick high-fineness shells (Fig. 11d-f). Such typomorphic features of gold are specific to metamorphogenic gold, because metamorphism of sedimentary strata at constant temperature and pressure leads to the transformation of gold (Moiseenko, 1965). In this case, the change in the internal structure of primary gold involves the removal of silver and impurities, which leads to the formation of recrystallization and regrowth structures, high-fineness shells, and, under dynamometamorphism (horizontal movements), numerous plastic deformations. Indeed, during deep epigenesis (prolonged impact of a constant pressure), impurity elements transfer from the inner parts of minerals to their periphery, i.e., the minerals purify themselves from impurities (Kopeliovich, 1965). Note that, although metamorphogenic gold was studied by many well-known gold mineralogists (Petrovskaya, 1973; Popenko, 1982; Savva, 1990; Nikolaeva et al., 2015; etc.), this problem calls for a serious research.

CONCLUSIONS

Along with the well-known factors responsible for the transformation of native gold under exogenous conditions,

we have first established some aspects of the transformation of its morphology, chemical composition, and internal structure depending on the ambient medium and the processes that the gold underwent.

Hypergene native gold forms along with endogenous gold in gold-bearing laterite and kaolinite weathering crusts. It is found as newly formed crystals, drusoid aggregates, isometric grains, balls, and finest growth forms on the surface of older gold particles. The removal of dispersed Au from sulfides, tellurides, and other minerals leads to its coarsening and refinement.

In the hydrodynamic environment, placer gold, independently of its form (hemihedral, euhedral, interstitial, etc.), flattens and undergoes a simple deformation, but its chemical composition and internal structure change little. We have established that the chemical composition and internal structure of gold change during its long occurrence in the environment and under its repeated redeposition from ancient (Precambrian) into younger (Quaternary) deposits.

Gold of anthropogenic placers transforms in both morphology and chemical composition. It is present as finest crystals deposited on various metal particles (wire, shot, etc.) and in the amalgamated form, as pseudomorphs intergrown with iron oxides and other minerals. It has been established that gold transformation depends on the technology of placer mining and on the landscape and climatic conditions.

In the eolian environment, during mechanogenic transformation, placer gold of different shapes tends to become a globule with a film-fibrous surface. The change in the shape of gold is accompanied by an increase in its fineness, a decrease in the content of impurity elements, and, as a result, reduction in the gold microhardness. In ancient gold-bearing intermediate sources, placer gold has not preserved its morphologic features but has transformed into pseudo-ore gold. Under the lithostatic pressure of overlying strata, gold formed unstable pseudointergrowths with minerals of the host deposits and got scars, scratches, through holes, etc.

In metamorphic strata with constant temperatures and pressures, gold underwent recrystallization and regrowth accompanied by the removal of impurity elements and the gold refinement.

We have established that exogenous processes change the morphology, mineralogical and geochemical properties, and internal structure of native gold. The unique Witwatersrand deposit, formed for 2.7-3.1 Gyr, is a prominent example of the influence of exogenous processes on native gold. We think that the gold found in the Witwatersrand gold-bearing conglomerates bears evidence for the influence of the hypergenesis zone processes, hydrodynamic and eolian environments, lithostatic pressure, and P-T conditions. The identification of toroidal and hollow spherical pseudo-ore and metamorphogenic gold testifies to the sedimentary origin of the Witwatersrand deposits (Nikiforova and Filippov, 1990; Filippov et al., 1994). The genesis of these rocks is still debatable. The presence of placer gold with typical indicators of alluvial and eolian deposits in ancient gold-bearing conglomerates and metamorphic strata suggests the sedimentary origin of the Witwatersrand deposits (Oberthur and Saagger, 1986; Minter et al., 1993; Filippov, 1997; Safonov et al., 2000) and the following superposed mineralization.

Thus, we have first substantiated the specific spatial and temporal evolution of gold under exogenous conditions. Decomposition of sulfides, tellurides, and other unstable compounds of gold in weathering crusts gave rise to new gold nano- and microparticles. The results of experimental and mineralogical studies prove that eolian processes change not only the shape but also the chemical composition of native gold. Under the lithostatic pressure of overlying strata, placer gold transforms into pseudo-ore gold. In metamorphic strata with constant temperatures and pressures, gold becomes refined.

The identified indicators of placer gold of different exogenous environments make it possible to reconstruct the geologic and geomorphologic conditions of gold placer formation, namely, to determine the genetic type of placers (formed in weathering crusts, alluvial, eolian, etc.) and the gold sources (intermediate or primary). This helps to find a more correct technique for the search for gold placer and ore deposits.

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