Recycling of gold tailings of silicate-carbonate ores

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Abstract. The implementation of the method for extracting gold, including melting of silicate-carbonate technogenic material, showed that gold passes into a free state, and the size of its particles increases significantly. However, when the material melts, a significant amount of slag is formed, which must be disposed. The chemical composition of the slag was compared with the norms of approximate and maximum permissible concentration. The content of Cu, As, Sb, S, and Mn exceeds the established standards. When the melt is blown with air and natural gas, sulfur, arsenic, and antimony will pass into the gas phase. In this regard, the installation must be equipped with a dust and gas cleaning system. If 12 wt.% CaO is added to the initial mixture before melting, the slag will meet the requirements when it is used as a correcting additive in cement.

1. Introduction
A large proportion of the world's gold reserves is concentrated in technogenic waste. In Russia, the resource potential of technogenic gold-bearing objects is estimated as 55-60% of the volume of gold mined in the country [1]. In such materials, gold is often represented by microdisperse particles, which complicates its enrichment. The implementation of the developed method for extracting microdisperse gold in a patented device [2], which includes melting the material, grinding it after cooling, and enrichment by gravity methods using the refractory carbonate-silicate materials as an example, showed that gold went into a free state, whereas the size of Au particles noticeably increased. This makes it possible to significantly increase its extraction by gravity methods. When the material is cyanidized after heat treatment, almost all gold is recovered [3]. However, when a technogenic material is melted, a significant amount of slag is obtained, which should also be disposed of. Previously, harmful impurities (S, As, Sb, etc.) should be removed from the slag and its basicity (CaO / SiO₂) should be increased. The purpose of this work is to develop a scheme for an integrated and environmentally safe processing of gold-containing silicate-carbonate tailings.

2. Methods
The initial material is represented by silicate-carbonate tailings of the gold deposit. It consists of (wt%): calcite (45.8), quartz (18.5), dolomite (12.5), plagioclase (9.2), muscovite (5.9), goethite and hematite (4.5), pyrite and arsenopyrite (3.6). Gold is represented by microdispersed particles, less than 10 microns in size. It is associated with pyrite and arsenopyrite, to a lesser extent with quartz, iron oxides and hydroxides, contains impurities of Pd (1.9-23.7%), Hg (2.5-3.8%), and Pt (up to 41%).

The mineral composition was studied on a JSM 639LV scanning electron microscope equipped with an EDS x-max 80 energy dispersive analyser. The chemical composition of the slag was determined on an ELAN 9000 inductively coupled plasma quadrupole mass spectrometer.
3. Results

The slag after melting is a homogeneous glassy mass of dark brown colour. The composition of the slag is dominated by SiO$_2$, CaO, and Al$_2$O$_3$. CaO / SiO$_2$ = 0.6 (Table 1), increased contents of Cu, Zn, Pd, and As are noted. Slag is 90-95% glass (tempered). The chemical effect of rainwater on the slag leads to removal of heavy metals and pollution of soils and nearby water bodies.

Besides glass, there are two minerals in the slag, the total amount of which does not exceed 5-10% of the total volume: iron oxide and an alloy of Cu and Zn. Fe oxide forms grains of isometric shape, small crystals, and intergrowths of crystals in the form of dendrites, the size of which rarely exceeds 30 μm. The mineral contains impurities Mn (1 wt.%). The Cu / Zn alloy consisting of Cu (62.2 wt.%) and Zn (35.0 wt.%) exists in the form of hook-shaped grains in a porous matrix with dimensions of 15-20 microns and contains Ni impurities (0.8 wt%).

Slag application options should primarily be based on an analysis of the possible impact on the environment and, in particular, on the soil. For this purpose, the chemical composition of the slag (Table 1) was compared with the norms of approximate permissible concentration (APC) and maximum permissible concentration (MPC) for neutral soils in terms of pH [4, 5]. The content of Cu, As, Sb, S, and Mn exceeds the established standards. When the melt is blown with air to float and increase the dispersed gold droplets, sulfur, arsenic, and antimony will pass into the gas phase. It is shown by thermodynamic analysis carried out using the HSC Chemistry 9.0 software package (Fig. 1). From the point of view of a more complete removal of arsenic, the melt should also be purged with natural gas [6]. The calculation showed that the required gas volume with an As content in the slag equal to 460 g/t was 22 m$^3$ per ton of melt (Fig. 1). The amount of arsenic in the slag will decrease up to 3.6 g/t. This complies with the APC norms. The forms of copper occurrence were analysed under the same conditions. The main part of copper (99%) will be in the Cu compounds, such as CuFeO$_2$, CuO, and CuO • Fe$_2$O$_3$. Metallic copper will be recovered along with gold during gravity separation. The total amount of non-recoverable metal in the slag (at a methane consumption of 22 m$^3$ per ton of melt) will be about 150 g/ton. This value of 18 g/ton is higher than the prescribed one for neutral soils. With an increase in methane consumption to 30 m$^3$ per ton of melt, the amount of copper in the oxide form will be within the limits of the APC norms. To capture gases, the installation [2] should be equipped with a dust cleaning and CO$_2$ removal system, several stages of removal of SO$_2$, SO$_3$, and gases containing arsenic and antimony. However, it is impossible to remove Mn using economically viable methods.

Table 1. The chemical composition of the slag obtained after melting of silicate-carbonate material at 1300°C (in wt.%). Content in g/ton.

| Oxide     | Macrocomponents | Microcomponents |  
|-----------|-----------------|-----------------|
| SiO$_2$   | 38.83           | Ni              |
| Al$_2$O$_3$| 18.3            | Cu              |
| TiO$_2$   | 0.46            | Zn              |
| Fe$_2$O$_3$| 7.6             | As              |
| FeO       | 3.86            | Mo              |
| MgO       | 4.5             | Sb              |
| CaO       | 23.4            | Pb              |
| MnO       | 0.43            | Au*             |
| K$_2$O    | 1.85            | Ag*             |
| Na$_2$O   | 0.84            | Pd*             |
| P$_2$O$_5$| 0.098           | Pt*             |
| SO$_3$    | 0.098           | Cd*             |
| Σ         | 100.17          |                 |
Figure 1. (a) Equilibrium composition of antimony compounds under heating the silicate-carbonate material to 1300°C (oxygen content of 1 litre per 100 kg of material). (b) Dependence of the change in the equilibrium composition of arsenic compounds on the amount of methane in the silicate-carbonate material at 1300°C (methane content from 0 to 2.2 m³ per 100 kg of material).

The use of the investigated slag for laying open pits and other disturbed lands of the same type is prohibited in accordance with GOST 15.5.1.03-86 [7], as well as hygienic standards 2.1.7.2511-09 and 2.1.7.2041-06 [4, 5] due to exceeding the norms of APC and MPC by the Mn component and the predominant glass content in the slag. For the same reasons, slag cannot be used in road construction and agriculture. But it can be used when laying mine workings as a filler for filling mixtures together with overburden and waste rocks of deposits. Usually, steelmaking and converter slags are disposed of in such a way [8].

After increasing concentration of gold by gravitational methods, the slag is in the form of a dispersed powder with a fraction of 0.071 mm. The wind will easily disperse it from the slag dumps, polluting the air and destroying the fertile soil layer. One of the disposal options is its use in the production of cement and backfill mixtures for mine workings.

Table 2. Quality criteria of the investigated slag.

| Criterion | Formula for calculation | Estimated indicators | Indicators required by GOST | Calculated indicators after adding CaO |
|-----------|-------------------------|----------------------|-----------------------------|--------------------------------------|
| Basicity module | M₀ = (CaO + Mg) / (SiO₂ + Al₂O₃) | 0.49 | Not standardized | 0.70 |
| Activity modulus | Mₐ = Al₂O₃ / SiO₂ | 0.47 | Not standardized | 0.47 |
| Coefficient according to GOST 31108-2003 [10] | K = (CaO + MgO) / SiO₂ | 0.72 | No less than 1.00 | 1.03 |
| Quality factor according to GOST 3476-74 | K = (CaO + Al₂O₃ + MgO) / (SiO₂ + TiO₂) | 1.18 | No less than 1.20 | 1.48 |

In the cement industry, metallurgical slag is used as a corrective additive. Granular blast-furnace slag regulated by GOST 3476-74 [9] is mainly used. GOSTs are standards for the chemical composition of the slag and criteria for its quality. Four criteria are used, two of which are reflected in GOSTs: basicity modulus, activity modulus, basicity coefficient according to GOST 31108-2003, and quality coefficient according to GOST 3476-74. The basicity modulus indicates the resistance of the
slag to lime decomposition. The activity modulus is responsible for the rate of solidification of the slag in the crushed state when interacting with water: the higher the modulus, the faster the slag solidifies. To assess this activity, a quality criterion was introduced according to GOST 3476-74. The coefficient according to GOST 31108-2003 determines the basicity of the slag; for any type of slag it must be greater than 1. The calculated indicators for the slag under study are given in Table 2.

From Table 2, it is seen that the slag does not meet two criteria: the quality coefficient and the coefficient according to GOST 3476-74 and GOST 31108-2003, accordingly. In terms of chemical composition (including the content of impurities), the slag meets the requirements. To ensure that the slag meets all the criteria, 12 wt% CaO should be added to the initial charge. This will also reduce the viscosity of the melt, which will increase the rate of coagulation of the gold particles.

4. Conclusion
A scheme of environmentally friendly complex processing of gold-containing silicate-carbonate tailings by heat treatment has been proposed. It allows extracting micro-disperse gold and using the resulting slag when laying mine workings as a filler for filling mixtures and in the cement industry in the form additives. The composition of the slag is adjusted during heating and melting. The proposed approaches can be used for the disposal of technogenic waste with various options for enrichment of silicate-carbonate gold-bearing ores.

Acknowledgements
The research was supported by the Ministry of Science and Higher Education in accordance with the state assignment for Ural State Mining University No. 0833-2020-0008 “Development and environmental and economic substantiation of the technology for reclamation of land disturbed by the mining and metallurgical complex based on reclamation materials and fertilizers of a new type”. We obtain the scientific results using the equipment of the Centre for the collective use of scientific equipment of the Federal Scientific Centre of biological systems and agricultural technologies of RAS as well (No. Ross RU.0001.21 PF59, the Unified Russian Register of Centres for Collective Use -http://www.ckp-rf.ru/ckp/77384).

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