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RESEARCH ARTICLE

APPLICATION OF NEW SORPTIVE POLYMERS FOR LEACHING OF METAL CONTAINING ORE

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ABSTRACT

The article deals with the problem of wastes disposal, which are produced from gold and uranium ore production. The article proposes novel method for disposal and burial of the wastes by means of leaching inside of opencast pits. In the scientific work complex approach is used, including analytic methods for the theoretical justification of temperature variations inside of the leached volume, analysis methods for calculated and experimental data processing, laboratory experiments for the determination of mechanical and thermal parameters of leached ores. The article contributes into the world scientific experience in the context of processing and disposal of wastes created by gold and uranium ore production. The authors for the first time proved that during the combined disposal of gold and uranium ores by means of heap leaching inside of the opencast mine the environmental pollution by dust and leaching solutions will be minimal if the heaps are formed at the bottom of the open pit mine. Temperature variations are in direct proportion with the total power of radioactive radiation sources and specific heat emission from ore leaching processes. For the proper estimation of the combined gold and uranium ores leaching methods the ecologic-economic factor $0 \leq \eta < 1$ should be used, which takes into consideration the burials of wastes inside of the exploited opencast mine.

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INTRODUCTION

Kazakhstan is one of the leading gold and uranium production countries. Currently there are more than 20 thousand hectares of fertile land in Kazakhstan are occupied by 10 billion tons of mining industry wastes. Whereas value of some waste products make up 25-50 % of the total value of produced mineral raw materials. Currently these wastes are considered to be as a source of additional valuable components. Big amount of consideration is also given to the problem of decreasing the influence these wastes impact on the environment. Heap leaching methods are widely used for uranium production and processing as well as for the recycling of unpayable and rebellious ores of precious and non-ferrous metals (Certificate of authorship №1351238, 1998). Heap leaching of gold, copper, zinc, lead, uranium and etc., involves application of toxic substances like cyanides and various acid solutions which negatively affect the environment (Begalinov *et al.*, 2001). Leakage of acid

solutions, release of dust and gases severely pollutes soil and ground waters as well as water basins of the region (Kolosov, 1987). Often used open-cut mining method causes formation of open mined-out spaces, refuse heaps and hillocks, which are also sources of contamination for the surrounding environment (Rzhevsky, 1985), and should be recycled, otherwise it is impossible to recover the land area (Kalibekov, 2006). It leads to loss of valuable components and decreases efficiency of production and processing of mineral resources by using heap leaching methods. Analysis of the previously done researches revealed that certain improvement should be done in the way of restricting the pollution produced by the ores heap leaching processes in the areas of open cut mines. That is why development of combined methods for recycling and disposal of wastes created by production and processing of gold and uranium ores, aiming to decrease environmental pollution, by using previously exhausted mines is an urgent scientific task. Aim of this work is to develop methods of recycling and disposal of wastes created by gold and uranium ores production and processing in open cut mines. Literature review shows that at the present time methods of combined leaching of gold and uranium ores do not exist. Analytical calculations of heap

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leaching process showed that release of dust emission, including radioactive dust, into the atmosphere behind the pit border decreases with the increase of wastes disposal depth. Besides that leakages of production solutions during the leaching process also decrease as the depth of heaps location increases inside of the pit. In order to decrease release of dust emission out of the pit border, taking into the account existing natural air ventilation schemes inside of the pits, the heaps should be located in the very deep part of the pit, where the intensity of air ventilation is not high. Thus release of dust emission would be minimal if the heaps are formed on the bottom of the pit. Analysis of water balance equation revealed that formation of heaps on the bottom of the pit would minimize leakages of the production solutions, thus possibility of ground water contamination by leaching solutions would be also eliminated. Formation of the heaps at the bottom of the pit also meets the conditions of the pit backfill technology during its reclamation. Thus the method of combined recycling of gold and uranium ores wastes would minimally influence the environment if the heaps are formed at the bottom of the pit. The mathematical model (1)-(3) is proposed for the analytical investigation of bulk temperature variations during the combined utilization of gold and uranium ores by means of heap leaching inside of the pit space. Variations of bulk temperature of combined leached gold and uranium ores is expressed by the inhomogeneous thermal conductivity equation:

$$C_p \rho \frac{\partial T}{\partial \tau} = \frac{\partial}{\partial x} \left(a \frac{\partial T}{\partial x} \right) + \beta_1 + \beta_2, \dots\dots\dots (1)$$

the initial and boundary conditions are the following:

$$T|_{t=0} = T_0, \quad 0 \leq x \leq h_k,$$

$$T|_{x=0} = T_1; \quad T|_{x=h_k} = T_2, \quad (\tau > 0) \dots\dots\dots (2)$$

Analytic solution of the equation may be presented in the following way:

$$T(x, t) = \frac{2(\beta_1 + \beta_2)}{C_p \rho} \sum_{n=1}^{\infty} \frac{\mu_n \sin \mu_n + B(1 - \cos \mu_n)_i}{[\mu_n^2 + B_i(B_i + 2)] \cdot \left[a \left(\frac{\mu_n}{h_k} \right)^2 - \sigma_H \right]} \cdot \left[\exp - \sigma_H \tau - \exp - a \left(\frac{\mu_n}{h_k} \right)^2 \tau \right] \left(\cos \frac{\mu_n}{h_k} x + \frac{B_i}{\mu_n} \sin \frac{\mu_n}{h_k} x \right), \dots\dots\dots (3)$$

where β_1 - heat source created by the oxidation of the leached ores;

β_2 - heat source created by radioactive emission of uranium ore wastes.

Analysis of the obtained solution shows that bulk temperature variations inside of the leached volume is in direct proportion to the total power of heat sources created by the oxidation of minerals and by the radiation of uranium ores. Delphi software was used to solve problems (1) and (2), the software allows the determination of the bulk temperature variations, throughout space and time. For the combined leaching of gold and uranium ores new methods were developed (Akhmedzhanov *et al.*, 2010). The proposed method of combined leaching implies existence of the following indicators (Akhmedzhanov *et al.*, 2010):

- Height and porosity of the heap, density and minimal granular size of the ore;
- Coefficient which takes into the consideration dynamics of the process and content of 0-1 mm granular size fraction in the volume of the heap;
- Maximal molecular moisture capacity of the ore;
- Width of the colmatage layer and content of 0-1 mm granular size fraction;
- Surface tension of the leaching solution and wetting contact angle.

With the aim to increase efficiency of ore leaching, disposal and recycling of the wastes, the heaps are formed in the depleted pits, by means of mixing of ore volume (V_{or}) with volume of radioactive wastes (V_{rw}) in the following ratio $\frac{V_{or}}{V_{rw}}$

=3. After the heap is formed it is covered from all sides by radioactive wastes of nuclear industry. Mixing of leached ore with radioactive wastes allows to increase temperature and heat exchange between the solution and the leached ore, what leads to the intensive oxidation of the ore, and consequently to the increase of valuable components release into the solution. Formation of the heaps inside of the depleted pits and their consequent disposal during the reclamation works excludes appearance of new polluted areas. The proposed leaching method was tested on models in laboratory conditions. Results of the grain-size analysis of the tested gold ore are presented in the Table 1. As the table shows the sample is mainly represented by the fractional size 0-0,2m. This fraction was subjected to more detail grain-size analysis results of which are presented by the Table 2.

Physical model of the leached ore volume was represented as a medium with heterogeneous porosity. Imagine that the real leached ore volume is composed of N fractions. $N = N - N_1$ is a big size fractions which consists the main ore skeleton, N_1 is a filling fraction.

Considering the volumes of the filling fraction, skeleton and real object the relationship between the porosities of the filling fraction and the real object was obtained:

$$m_{N_1} = \frac{m_p}{\varphi_{N_1}} - \sum_{i=1}^{N_1} \varphi_i', \dots\dots\dots (4)$$

where m_{N_1} - porosity of the filling fraction;

m_p - porosity of the real object;

$\sum_{i=1}^{N_1} \varphi_i'$ - sum of the relations between the volume-parts of each fraction of the filling agent, which is determined by the following formula:

$$\sum_{i=1}^{N_1} \varphi_i' = \frac{\sum_{i=1}^{N_1} \varphi_i}{\varphi_{N_1}}, \dots\dots\dots(5)$$

here φ_i - volume-part of each fraction of the filling agent;

φ_{N_1} - volume-part of the whole volume of the filling agent;

Model porosity of the filling agent is calculated by using formula (4) and Tables 3 and 4.

If the porosity of the filling agent is less than that of the real object then one or part of one, or several big size fractions are transferred from the chosen fraction of the filling agent N_1 to the skeleton of the real object and the calculations are continued until the porosity of the filling agent will be equal to the porosity of the real object ($m_{N_1} = m_p$). Amount of the fractions N_1 at which $m_{N_1} = m_p$ is chosen as the real object model. In some cases porosity of the filling agent N_1 may be bigger than that of the real object. In this case one or part of one, or several small size fractions are taken from the skeleton of real object and are added to the preselected filling agent N_1 and the calculations are continued until the porosity of the filling agent will be equal to the porosity of the real object ($m_{N_1} = m_p$). Amount of the fractions N_1 at which $m_{N_1} = m_p$ is chosen as the model for laboratory investigations.

The same principle was used to tailor the porosity of radioactive wastes with which leached ore was mixed. Calculated values of the porosity of the leached ore and radioactive wastes were checked experimentally. Calculated and experimentally measured porosities of the leached volume with the radioactive wastes matched with relative error of 15 %. Thus the selected model is representative to the real object. Results of the calculations are summarized in the Table 3. As it is seen from the table 3, for the ore number 10 the porosities of the model and the real object match. For the model first ten fractions which are summarized in the table 2 are taken, i.e. the following fractions 0-1,0; 1,0-2,5; 2,5-5,0; 5,0-7,5; 7,5-10,0; 10,0-12,5; 12,5-15,0; 15,0-17,5; 17,5-20,0; 20,0-30,0 (intervals in mm). For the ores number 9 the ninth fraction from the table 2 is taken for the laboratory experiments with the porosity value (table 3) 0,3. For the ores under the numbers 1, 2, 6, 7 and 13 for the laboratory experiments first eight fractions are taken, the porosities of these fraction (Table 3) are the following - 0,26, 0,25, 0,27, 0,24 and 0,24. For the ores under

the numbers 3, 4, 5, 8, 11 and 12 first seven fractions are taken for the laboratory experiments, with the following porosities (Table 3) - 0,25, 0,26, 0,26, 0,24, 0,25 and 0,26. Thus the model is formed on a concrete slab with a hole for the leakage of the leaching solution. The concrete slab has concave form what allows the solution to flow out through the hole. The model pile was formed in such a way that relation between the volumes of the ore V_o and the radioactive wastes V_{rw} is equal 3, and also surface of the pile was covered by the radioactive wastes. The model pile was sprinkled with 2 % thiosulfate solution. New sprinkling method was proposed to increase the efficiency of sprinkling during the heap leaching process and to increase the covering efficiency of the leaching solution (7), the main point of which consists in the following. According to this method heap leaching of the unpayable and rebellious ores consists in the covering of the heap by a layer of low porosity material, this material might be, for example, wastes of uranium ore. This will increase the efficiency of ore mass treatment. The thickness of the covering layer is calculated by the following formula:

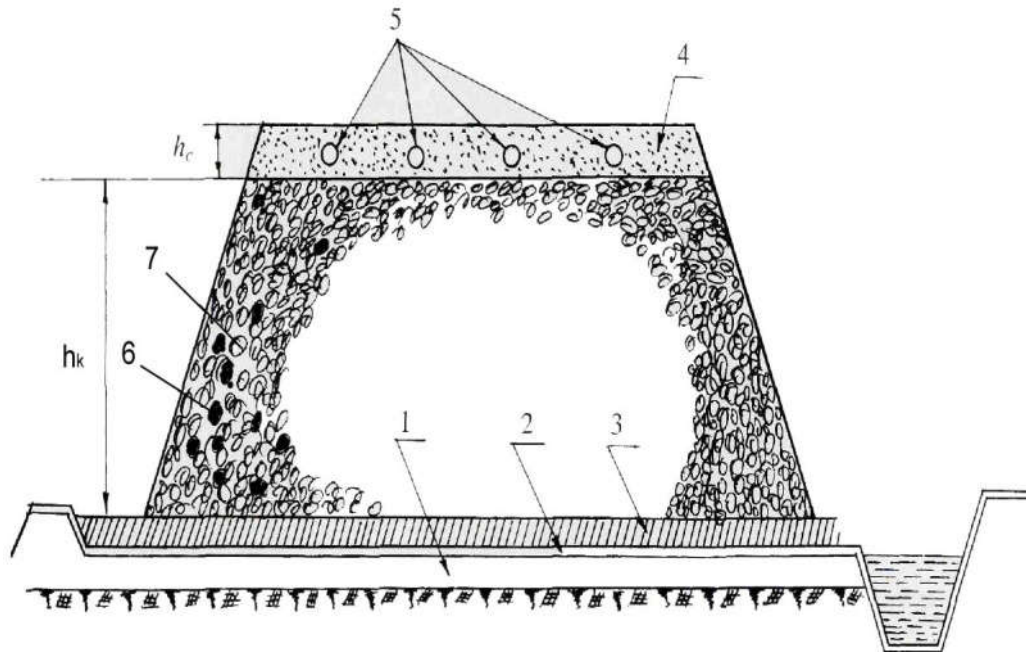
$$h_c = h_k \left[\frac{Q_0 \cdot t \cdot \rho_p}{q_k \cdot M} (1 - m_k) - 1 \right], \dots\dots\dots(6)$$

Where h_c —thickness of the layer, m; h_k —height of the heap, m; Q_0 —maximal flow rate of the pump, which pumps the leaching solution on the surface of the layer, m³/sec; t —time, during which the whole volume of the heap is treated, sec; ρ_p —packed density of the ore inside of the heap, kg/m³; q_k —specific flow rate of the leaching solution needed for the ore treatment, m³/m³; M —mass of the formed heap, kg; m_k —average porosity of the heap, m³/m³.

Porosity of the covering layer is calculated by the following formula:

$$m_c = \frac{\rho_c m_k [\varphi_0 - (W_k + 2\varphi_k + \varphi_k)]}{\rho_p (1 - m_k) [\varphi_{oc} - (W_c + 2\varphi_{oc} + \varphi_c)] + \rho_c m_k [\varphi_0 - (W_k + 2\varphi_k + \varphi_k)]} \dots\dots\dots(7)$$

To prevent ore losses as a result of colmatage and to decrease environmental pollution by the solutions new sprinkling method was developed and approbated. The method provides steady-state filtration mode of the leaching-out liquid inside of the muck pile. The main point of the method consists in the following. Before the sprinkling of the leaching solution a layer of low porosity material is formed on the surface of the heap. Let h_k be a height of the formed heap and h_c is a thickness of the layer. The layer composed of low porosity uranium ore is intended for steady-state and regulated injection of the solution covering the whole surface of the heap. Besides that, the layer prevents intensive evaporation of the leaching solution because the feed point is isolated from the atmosphere what is very important in the case of cyanide leaching of gold. The method also increases the efficiency of the leaching process by the following factors: preventing colmatage phenomenon; providing evenness of the treatment; extension contact time between the solution and the ore.



1- mud; 2 - polyethylene film; 3 - sand; 4 –low porosity piled up layer; 5 –tubes for fluid supply; 6 – mining industry wastes; 7 – nuclear industry wastes.

Figure 1. Scheme of closed irrigation during the heap leaching process

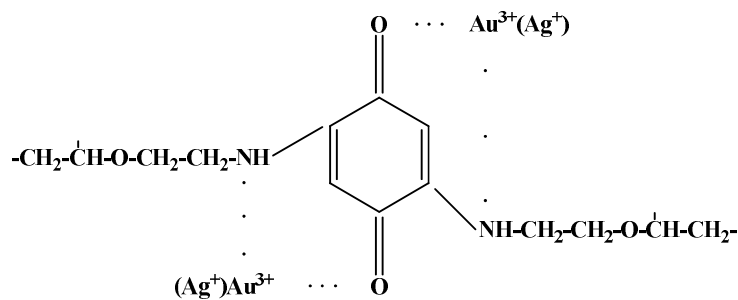


Figure 2. Complexing of redox (co)polymers with noble metals on the basis of quinoid derivatives of VEMA

Table 1. Grain-size analysis of the leached ore

№	V, m ³ volume	Grain-size composition of the ore, %, m				
		0-0,20 m	0,21-0,40 m	0,41-0,60 m	0,61-0,80 m	0,81 m
1	7,7	50,2	21,5	13,4	12,6	2,2
2	8,1	53,1	22,2	12,7	10,4	1,5
3	9,0	55,5	21,5	12,5	8,7	1,7
4	7,0	56,6	21,7	11,7	7,6	2,3
5	10,2	57,4	20,6	13,4	7,3	1,2
6	9,5	47,4	19,4	15,9	15,0	2,2
7	8,5	52,5	21,0	12,3	11,7	2,4

Table 2. Grain-size composition of the ore sample with grain size 0-0,20 m

№	Size range, mm	1	2	3	4	5	6	7
		φ1, %	φ1, %	φ1, %	φ1, %	φ1, %	φ1, %	φ1, %
1	0-1,0	4,5	4,3	5,2	5,0	3,7	3,5	4,1
2	1,0-2,5	6,0	5,8	6,5	6,1	7,0	6,9	6,0
3	2,5-5,0	7,3	7,5	6,9	8,0	7,2	7,5	7,2
4	5,0-7,5	6,4	6,6	7,9	8,5	7,5	8,5	6,9
5	7,5-10,0	7,7	7,3	9,2	8,7	9,3	9,4	7,3
6	10,0-12,5	9,4	9,0	9,6	9,3	9,6	10,4	9,0
7	12,5-15,0	8,9	9,2	10,7	9,5	10,2	9,1	9,2
8	15,0-17,5	9,9	10,0	7,2	8,5	11,0	7,7	9,4
9	17,5-20,0	7,3	6,7	6,5	4,8	4,1	4,5	9,8
10	20,0-30,0	6,5	7,5	7,0	8,5	6,8	6,9	6,7
11	30,0-40,0	5,9	5,5	5,1	4,5	5,3	6,6	7,5
12	40,0-50,0	6,0	5,8	4,9	4,4	5,9	6,8	5,5
13	50,0-100	5,4	5,6	5,1	4,0	4,5	4,0	5,8
14	100-150	4,8	5,0	4,5	5,3	4,7	5,2	3,6
15	150-200	4,0	4,2	3,8	4,5	3,2	3,0	2,0

Table 3. Parameters of the real object and the model

N n/n	m_p , m^3/m^3	$\sum_{i=1}^{10} \varphi'_{10}$	$\sum_{i=1}^9 \varphi'_i$	$\sum_{i=1}^8 \varphi'_i$	φ_{10}	φ_9	φ_8	m_{10}	m_9	m_8	m_7
1	0,26	-	-	0,60	-	-	0,30	-	-	0,26	-
2	0,27	-	-	0,60	-	-	0,32	-	-	0,25	-
3	0,25	-	-	0,63	0,560	-	0,35	0,31	-	0,08	0,25
4	0,25	-	-	0,64	0,551	-	0,36	0,31	-	0,06	0,26
5	0,25	-	-	0,66	0,545	-	0,38	0,31	-	0,02	0,26
6	0,27	-	-	0,63	-	-	0,30	-	-	0,27	-
7	0,26	-	-	0,60	-	-	0,31	-	-	0,24	-
8	0,24	-	-	0,59	0,513	-	0,36	0,32	-	0,08	0,24
9	0,29	-	0,697	0,62	-	0,30	0,26	-	0,30	0,49	-
10	0,25	0,72	-	0,59	0,26	-	0,22	0,24	-	0,54	-
11	0,24	-	-	0,62	0,519	-	0,37	0,31	-	0,03	0,25
12	0,24	-	-	0,61	0,512	-	0,37	0,31	-	0,04	0,26
13	0,26	-	-	0,60	-	-	0,31	-	-	0,24	-

Table 4. Sorptive properties of new copolymers towards the ions of the noble metals

Homo- and copolymers	CE _{Ag}		Extraction degree Ag,%	CE _{Au}		Extraction degree Au,%
	mg/g	mg-ekvg		mg/g	mg-ekvg	
-VEMEA-Q-	463,20	4,29	43,89	95,32	0,48	93,58
-VEMEA-Q-VEMEA-	381,60	3,53	45,33	98,18	0,49	96,12
VEMEA-Q:AA	274,80	2,55	30,22	96,96	0,49	91,66
VEMEA-Q:ST	420,00	3,89	41,09	95,16	0,48	92,77
VEMEA-Q:2-VP	450,10	4,02	43,57	98,78	0,50	93,07
VEMEA-Q: 4-VP	452,40	4,19	45,58	97,28	0,49	94,22
VEMEA-Q:N-PVP	268,80	2,49	46,64	97,58	0,50	93,63
VEMEA-Q-VEMEA: AA	369,60	3,43	37,67	97,58	0,50	97,35
VEMEA-Q-VEMEA:ST	320,40	2,97	32,07	97,88	0,50	97,47
VEMEA-Q-VEMEA:4-VP	243,60	2,26	29,68	96,66	0,49	97,75
VEMEA-Q-VEMEA:N-VP	262,80	2,44	28,11	83,64	0,42	97,47

Table 5. Results of physical simulation of gold ore leaching in laboratory conditions

№	Type of the ore	Degree of gold extraction	
		Without radioactive wastes	With radioactive wastes
1	Sulfide ore from mine field Akbakay with gold content 1,6 g/ton	75-78	80-85
2	Ore with gold content 2,0 g/t	87-96	93-97

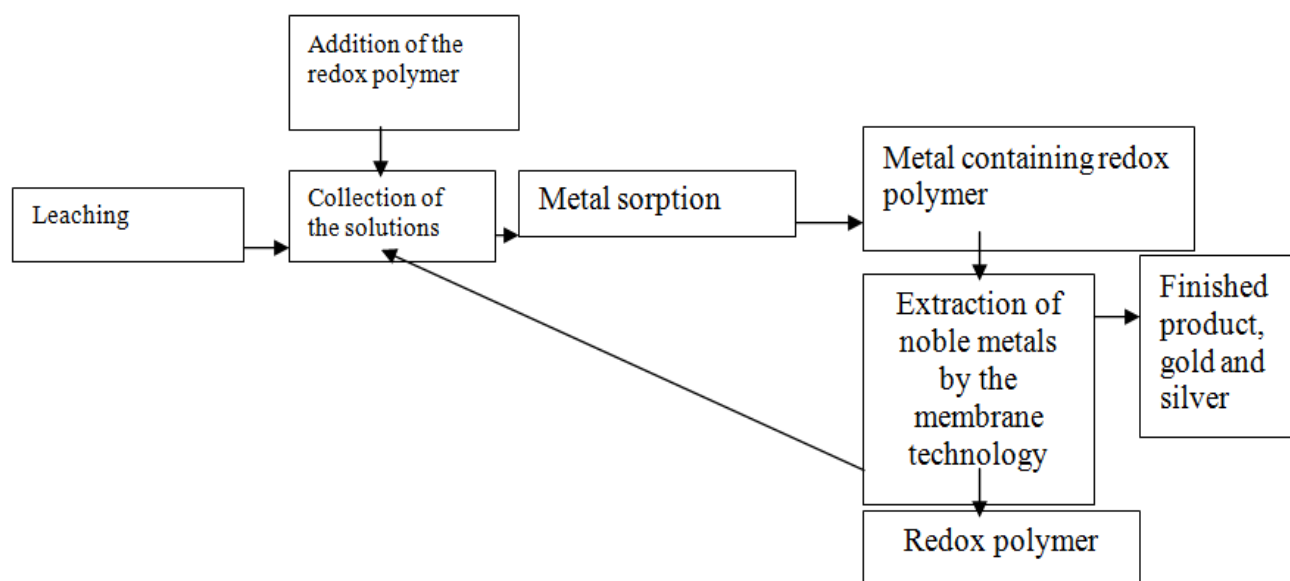


Figure 3. New technological scheme of leaching solutions collection for the extraction of noble metals

These factors not only increase extraction of valuable components but also decrease cost of the leaching process due to the fact that to obtain needed concentration of the valuable components in the solution multiply circular and tautological retreatment of the ore is not required any more, millponds and other irrigation constructions are not required as well. The above mentioned factors should decrease the environmental pollution. The proposed method of ore treatment by leaching solutions is implemented on the basis of calculated parameters of the piled up layer. As a result of proper chose of the piled up layer properties it is possible to increase the efficiency of the leaching process due to the improvement of covering coefficient and extraction of the valuable components. Creation of the piled up layer excludes the need of repeated construction of irrigation system facilities, and due to the evenness of the irrigation, steady-state liquid filtration regime and wetness of the surface layer decreases evaporation and leakages of the solutions and dust release from the horizontal surface of the heap. Pilot test of the proposed method at the mine field "Karerno" showed it to be highly effective. Thus, the method allows to increase leaching efficiency and to decrease environmental pollution as well, due to the simultaneous disposal of mining and nuclear industry wastes in the depleted pits. After ore leaching process sorption technologies are usually used for gold extraction from the productive solutions. Costly, imported substances like ion-exchange resin, charcoal from fine wood are used as sorbent agents. To decrease cost of gold extraction from the leaching solutions. In this regard ability of new polymers to sorb gold ions was studied. Unique regeneration abilities of these compounds provide possibility of multiple usage and recovery of expenses, what makes this class of high molecular compounds perspective. For the purpose of gold extraction the productive solution after the leaching process was treated by redox-ionits instead of ion-exchange resins. The authors have developed one-step method of obtaining unsaturated nitrogen-containing redox monomers on the basis of cheap and available materials - vinyl ether monoethanolamine (VEMA, Temirtau city, JSC "Alash"). The method also allows to obtain quinones of various chemical structures and synthesize new redox-ionits on their basis. Study of physicochemical, redox and complexing properties of these substances and investigation of the most perspective ways of their practical application were done (Ergozhin *et al.*, 2002).

Application of cheap domestic materials found in Kazakhstan – VEMA will allow simplification of the synthesis and decrease the cost of redox-polymers. Possibility of their multiply regeneration and decrease of the transportation expenditures will compensate production costs. Besides that, existence of amino group in the structure of VEMA excludes additional amination stage, gives hydrophilic properties to the polymer and improves kinetic properties. Due to the complexing and redox properties of the polymers they can be used as oxidants, deoxidants and dehydrating agents in chemical industry and hydrometallurgy for the separation and concentration of heavy and noble metals ions, waste water treatment, catalysis and to solve variety of ecological problems. Atomic absorption analysis is one of the most precise methods used to study sorptive power of polymers. Sorption of metals from HCl solution with normal concentration equal to

0,5% and with pH=1 was conducted in the laboratory. As the results showed, under the imposed conditions all studied redox polymers exhibit selectivity towards the ions of the noble metals (Table 4). As it is seen from the above table 4, the highest sorption degree (97%) is demonstrated by polymers on the basis of disubstitutedquinoid derivatives of VEMA, what evidently is caused by high concentration of amino groups. Lower degree of silver ions extraction allows selective sorption of gold ions from the mixture of these metals. Complexing of redox (co)polymers on the basis of quinoid derivatives of VEMA takes place because of oxygen atoms, ether linkage and nitrogen from amino group of vinyl ether monoethanol amine (MEA), and also because of carbonyl group of quinones and metal ions (Fig.2). How it is seen from the Figure 2, redox(co)polymer will extract gold along with silver from ore leaching solutions. Besides that in the structure of copolymers based on the quinoid derivatives of VEMA nitrogen atoms of 4-vinylpyridine and 2-vinylpyridine heterocycles are involved in the process. As opposed to the donor nitrogen atoms linked with metal ions donor oxygen atoms from coordinated ligands usually easily protonated and cause the distraction of the chelate rings.

Leaching of gold containing ore mixed and covered with radioactive wastes in the ration $\frac{V_{go}}{V_{tw}} = 3$ allowed to increase gold

extraction on 20 % in comparison with the same ore leaching without addition of the radioactive wastes. Results of physical simulation of gold ore leaching in laboratory conditions are presented by the Table 5. Thus, the proposed ore leaching method allows to increase the efficiency of the leaching process and to decrease environment damage due to the in situ disposal of mining and nuclear industry wastes at the depleted pits.

Realization of the project will allow to solve variety of nature protection tasks and to decrease anthropogenic impact on the environment in mining areas of Kazakhstan. On the basis of the conducted research the following innovative technological scheme of gold extraction was proposed (Fig. 3). Promising outlook of the proposed scientific work consists in the development of domestic technologies of gold, uranium and other metals extraction from wastes of mining industry, what allows to solve variety of ecological problems in the country.

Conclusion

1. In Kazakhstan there are a lot of depleted pits and slag-heaps of unpayable gold and uranium ore which are located in close proximity form each other and should be disposed;
2. New methods based on the usage of the pit space and allowing effective disposal of unpayable gold and uranium ore by means of heap leaching were developed;
3. Mathematical model of temperature variations inside of the leached ore volume was developed. The model allows forecasting the efficiency of the combined heap leaching of gold and uranium ores depending on their physicochemical and thermal properties;
4. New redox(co)polymer synthesized from wastes of carbide industry is recommended to be used for gold and silver extraction from the leaching solutions.

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