Proceedings World Geothermal Congress 2005
Antalya, Turkey, 24-29 April 2005

Pechenga Super Deep-Level Hydrothermal System as an Analogue of Present-Day Hydrothermal-Magmatic Systems

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Keywords: system, hydrothermal, magmatic, structure, ore-forming, conceptual model

ABSTRACT

The article formulates a concept of deep-level (super deep-level) hydrothermal system of platform areas analogous or similar to high-temperature hydrothermal-magmatic systems of modern volcanism areas by its geologic structure and thermodynamic parameters. The Pechenga super deep-level hydrothermal system is studied: geological and hydrogeological structure, geothermal conditions, formation of geochemical ore barriers within its interior. The authors show possibilities to use such systems for investigation of geotechnical conditions of formation of ore deposits and isolation of various industrial waste when disposed of and buried directly within natural objects.

1. INTRODUCTION

The present work is continuation of studies on issue adsorption of alkaline and heavy ore elements in modern high-temperature hydrothermal-magmatic systems belonging to quaternary volcanism areas (Belousov, Rychagov et al., 1998, 1999). These systems are accessible to studies of properties of surface and deep-level natural waters and adsorption properties of both hydrothermal fluids and water-bearing and water-resisting rock masses. Thermal anomalies within hydrothermal systems have been studied by geologic-geophysical, geochemical and other methods for decades. They were drilled with numerous boreholes. Usually, a hole depth of 1.5-2.0 km is enough to study the whole range of hydrothermal-metasomatic processes ongoing in the interior of high-temperature hydrothermal systems. Results of super deep drilling in the Kola peninsula, in Tatarstan, in Azerbadzhan, in Ukraine and in other regions of former USSR allowed to identify zones of active circulation of thermal waters ($\geq$ 150-200^{o} C) within a depths interval of few kilometers to 9-12 km. This permits to formulate a concept of ***deep-level (super deep-level) hydrothermal system*** of platform areas analogous or similar to high-temperature hydrothermal-magmatic systems of modern volcanism areas by its geologic structure and thermohydrodynamic parameters. Therefore, we suppose that study of properties of hydrothermal fluids and water-rock interaction processes in the ways of migration of hydrotherms and in shield rock masses of modern hydrothermal systems will contribute to implementation of projects on investigation of possibilities to localize ore matter and bury various industrial waste at super deep-levels of platform areas.

2. HYDROTHERMAL (HYDROTHERMAL-MAGMATIC) SYSTEMS AND GEOTHERMAL DEPOSITS OF TRANSITIONAL ZONE FROM OCEANIC TO CONTINENTAL EARTH'S CRUST

The studies of hydrothermal activity in areas of Quarternary volcanism identified a geological-hydrogeological structure referred to as "a hydrothermal system". Generally, hydrothermal systems are thermal upper crust anomalies confined to certain geological structures and showing hydrothermal surface activity. A heat flux within the anomalies exceeds mean values for the Earth up to 50-100 times. This allows assuming that heat transfer takes place by means of convection. Within near surface conditions water and steam act as heat carriers. Hydrothermal systems are considered to be high-pressure hydrogeological structures similar to artesian basin or artesian decline occurring in the crust when a deep-level heat carrier – an overcritical water fluid or magmatic melt - penetrates water-bearing layers (Belousov, 1978).

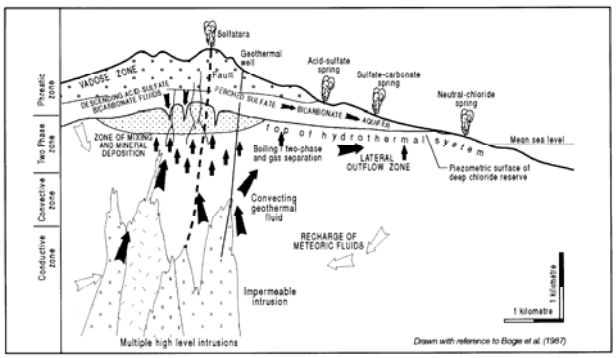


Figure 1: A conceptual model of hydrothermal-magmatic convective system (Corbett and Leach, 1998).

The development of conceptual models for formation conditions of epithermal ore and geothermal deposits, deep drilling of present-day hydrothermal systems and data on the composition of a restored endogenic fluid allowed to make the definition of volcanic-magmatic-hydrothermal systems over last 10 - 15 years (Giggenbach et al., 1990). There has been established a genetic correlation of volcanism and hydrothermal activity conditioned by a single upper-mantle anomalous heat flux, the stable development of which governs the long evolution of such systems. Usually, in near surface levels of the earth's crust the relation between intrusive or subvolcanic bodies of middle to base composition and circulation of hydrothermal solutions is analyzed **(Fig. 1)**. J. Hedenquist with coauthors (Hedenquist et al., 1996) developed a concept for

development of volcanic-hydrothermal, geothermal and ore-forming systems within a single geological structure: from subvolcanic high-temperature and high-sulfur to surface low-temperature and low-sulfur conditions of mineral and ore formation. Based on a large joint team research work at volcanoes and hydrothermal systems of present-day and ancient island arcs over 1980 – 90's, we located **new geological objects**: long-existing ore-forming hydrothermal-magmatic convective systems at the ocean - continent transition zone. It is these through-crust drain systems that govern the transfer of heat energy, melts, gases, hydrothermal solutions and chemical elements from the upper mantle level to near-surface levels of the crust. As the systems evolve, their agents (intertelluric flows, melts, magmatic gases, hydrothermal fluids) interact with host rocks, marine, underground, meteoric waters and actively influence the geological restructuring processes thus promoting isolation of the anomalous heat flux. The isolation process is self-controlled by deposition of silica and other secondary minerals around the hydrothermal-magmatic column. The latter circumstance causes the formation of epithermal ore, geothermal, porphyry and other type deposits in the upper parts of the earth's crust.

Studies, exploration and exploitation of geothermal deposits have been long conducted in many countries: Italy, New Zealand, Island, Indonesia, Philippines, USA, Nicaragua, Japan etc. A contribution of geothermal electricity generation in these countries constitutes to 22 %; geothermal heat supply is well developed too. Mineralized waters are used for balneology, for food and other industries; valuable components are derived from brines. In recent years the issue of extraction of thermal and electric energy from fractured deep-level rocks has been dealt with in some European countries – Germany, Switzerland, France etc. In large, this trend of fundamental science and technology – the study and exploitation of geothermal deposits – has good social and economic prospects worldwide. Russian scientists gained a significant experience in study and exploitation of geothermal deposits: near-surface low temperature therms (heat supply, balneology, food industry) and vapor hydrotherms deposits (Pauzhetka and Verkhne-Mutnovskaya geothermal plants; Mutnovskaya geothermal field exploration, etc.) were located in Caucases, in Kamchatka, in the Kuril islands. Nevertheless, exploration and exploitation of deposits in Russia are still at the level of pilot production. Beside general economic reasons, it is due to instability of many parameters of near-surface deposits and the complicated structure of hydrothermal systems in the deep levels of the crust.

Near-surface geothermal deposits are characterized by low P-T parameters and limited resources with a heat carrier having aggressive properties. These factors induce fundamental science and geothermal energy operations to study the deep levels of hydrothermal systems, factual data on interconnection of hydrothermal and magmatic deep-level processes were derived (Muraoka et al., 1995). Moreover, super deep drilling in USSR and Russian Federation (Kola, Saatlinsk, Minnibaevsk, Novo-Elkhovsk, Ural, etc.) helped to prove the existence of anomalous heat fluxes and overflow zones of gas-liquid fluids in the interior of ancient platforms and in the activated regions. A case study of Kola megablock of the East-European platform and Kola super deep level borehole SG-3 along with a study of data on the deep-level crust structure of Ural, Tatar and other regions showed a possibility of existence of present-day super deep-level ore-forming hydrothermal systems (Belousov, Rychagov et al., 2001), which are similar or close to high-temperature hydrothermal-magmatic convective systems of ocean-to-continent transition zone by their structural and thermodynamic parameters. This result is of the principal importance for the study of composition, dynamics and water flow formation mechanisms in the middle and lower sections of the crust and for comprehension of adsorption ore-forming processes at high P-T parameters.

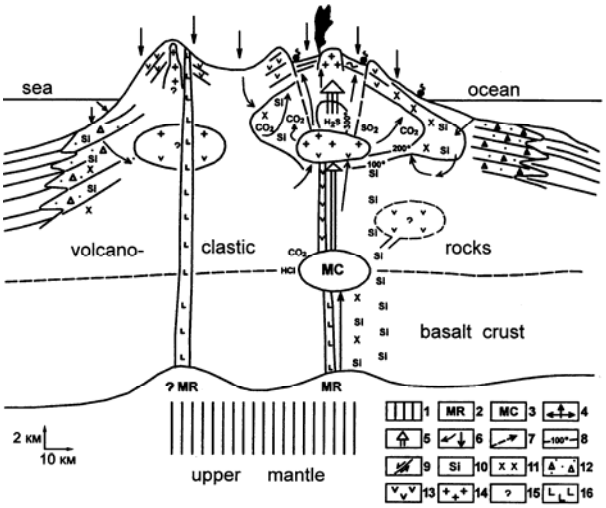


Figure 2: A model for structure of a hydrothermal-magmatic system of the present-day volcanic island arc. 1 – Generation zone of primitive basaltic magmas in the upper mantle; 2 – upper-mantle magmatic reservoirs – accumulation place for primitive melts; 3 – magmatic crust centers– a stop place for primitive melts in the crust; 4 – injection of primitive melts; 5 – injections of differentiated melts; 6 – infiltration of sea (oceanic) water; 7 – migration of underwater hydrotherms; 8 – isotherms of hydrothermal systems; 9 – discontinuous tectonic faults; 10 – hydrothermal alterations (silicification); 11 – same (propylitization: chloritization, epidotization and other); 12 – pyroclastic deposits; 13 – lavas of andesites; 14 – riolites; 15 – intrusive bodies; 16 – dykes of basalts.

3. PROPERTIES OF HIGH-TEMPERATURE HYDROTHERMAL SYSTEMS ENABLING THEIR HIGH ADSORPTION CAPABILITIES

Hydrothermal ore deposits related to volcanogenic structures have been existing since the Archaean Period up to now. Locations of the deposits are not hazardous as far as non-controlled distribution of harmful substances and their impact on living entities are concerned. In this respect we may conclude that the interior of volcanogenic structures hosts ongoing processes providing an ecologically safe accumulation and control of ore, alkaline, rare and other elements. Notwithstanding that ore-forming processes have been studied for many centuries, the issue of identification of sources of metals, mechanisms of their transfer, redistribution and accumulation still remains one of the most relevant issues in geology. Modern volcanogenic structures, namely, such as hydrothermal-magmatic systems of island arcs **(Fig. 2)** are most appropriate for study of the whole range of hydrothermal ore-forming processes. The whole geological section is disclosed in a relatively small territory: from Miocene-Pliocene or more ancient

volcanogenic-sedimentary basement to modern lava flows, intrusions and pyroclastic deposits.

Both zone of ascending flux of ore-bearing fluid and areas of lateral diffluence of hydrotherms are accessible to study. Temperatures of processes within the system's interior range from ≥ 1000°C to 0°C; chemical composition of surface and underground waters is diversified, concentrations vary from ultra saltless to brines and pH of solutions vary from alkaline to ultra acid. Volcanic and hydrothermal gases, also available for study, play an important role in ore formation. Therefore the given objects can be used for conducting pilot field experiments within a wide range of pressure and temperature parameters.

Geochemical barriers are main obstructions in the way of migration of chemical elements and their compounds. Adsorption-desorption properties of minerals, composing enclosing rocks, constitute the basic characteristic of barriers. Volcanogenic minerals, especially the hydrothermal-metasomatic ones, are good metal absorbers. In the past few years, Institute of Volcanology, FED RAS, has been developing a new trend of research in adsorption capabilities of high-temperature hydrothermal-magmatic systems located in volcanic islands and in the ocean-continent transition zone (Belousov, Rychagov et al., 1998; Rychagov, Belousov et al., 2002). Such systems are surrounded by seawater areas and influenced by sea waters penetrating many kilometers to enclosing rocks and to a depth of 2.5 km or more. The interiors of systems ceaselessly reproduce minus charged substances capable of adsorbing metals (positively charged cations), practically, in unlimited quantities for many tens and hundreds of thousands years. One of the most common adsorbents is silicic acid (silica gel), which has the following properties. Usually gels coagulate after gaining positively charged particles and precipitate. After sorption, silica gel moves on together with a hydrothermal fluid towards its discharge point and precipitates as fluid temperature falls. Sea water, bearing magnesium and calcium, is an impassable barrier for silica gel and it causes an almost complete silica deposition from hydrotherms ([Janecky, Seyfried, 1984). In such a way, granted that metals are transported by silica acid gel, ingress of heavy metals from high-temperature hydrothermal systems to sea areas is practically excluded. Silica gel performs two, so it should seem, mutually exclusive functions: 1) precipitates chemical elements and their compounds at geochemical barriers and facilitates the formation of ore deposits; 2) reduces concentration of metals in solutions because of its wide propagation across the whole geological area where the hydrothermal system functions. The latter is promoted by high fracture and porous permeability of enclosing volcanic and volcanogenic-sedimentary rocks and other properties of the system.

4. THE PECHENGA SUPER DEEP-LEVEL HYDROTHERMAL SYSTEM

The study of structure and geothermic examination of strata composing the base of the East European Platform, drilled open with deep and super deep boreholes, indicate that the interior of the platform accommodates hydrothermal systems (solution temperatures ≥ 150°C). The latter belong to convective cells the lower parts of which according to indirect data are sunk not only into the base of the earth's crust but also into the upper mantle. We believe that such a system is the Pechenga one, drilled open with the Kola super deep borehole SG-3 (Kolskaya…, 1984; Khristoforova et al., 1999). Formation, migration and discharge of hydrothermal solutions are predetermined by geological history of this region.

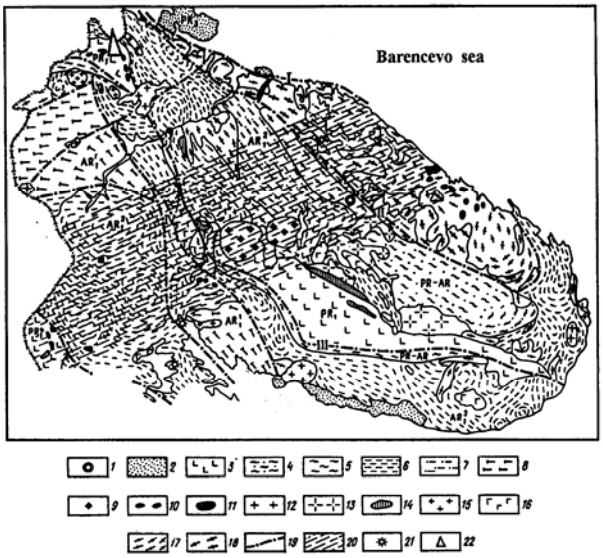


Figure 3: Geological setting of the Kola peninsula (Kolskaya…, 1984). 1 – Sedimentary-volcanogenic complex of Contozerian structure, upper Devonian and lower Carbonian; 2 – suits of Hyperborean series, of the Rybachiy peninsula – conglomerates, sandstones, clay shales; of the Sredniy peninsula, the Kildina and Terskaya islands - quartzites, sandstones, parti-colored clay shales, dolomites; upper Proterozoic; 3 – Pechenga,Imandravarzuga, Kuolajarvi complexes – base, middle, ultra-base lavas, tuffogenic formations, dominantly metamorphosed to green-shale facies; lower Proterozoic; 4 – Keivy series– aluminous, carbonic, micaceous and other shales, quartzites and sandstones, dominantly metamorphosed to epidote-amphibolitic facies; lower Proterozoic– upper Archean; 5 – Tundra series– amphibolitic, micaceous, aluminous shales and gneisses, amphibolites, ferric quartzites, dominantly metamorphosed to epidote-amphibolitic facies; lower Proterozoic – upper Archean; 6 – biotite-plagioclase, biotite-amphibolitic-plagioclase, two-mica gneisses etc., a dominantly amphibolitic facies of metamorphism; lower Archean , Kola series; 7 – biotite-plagioclase gneisses, amphibolites, pyroxene-amphibolitic-plagioclase gneisses and crystalline shales, ferric quartzites, dominantly metamorphosed to amphibolitic facies; lower Archean , Kola series; 8 – granulitic complex – cordierite-garnet-biotite-plagioclase gneisses, pyroxene-amphibolitic-plagioclase gneisses and crystalline shales, amphibolites and migmatites related to them; lower Archean, Kola series; 9 – complex of alkaline nepheline syenites, upper Devonian; 10 – complex of base, ultrabase and alkaline rocks, upper Devonian; 11- gabbro, diabases, granophyres, upper Proterozoic; 12 – granites, granodiorites, lower Proterozoic; 13 – alkaline granites, syenites, lower Proterozoic; 14 – gabbro, gabbro-norites, pyroxenites, peridotites, olivinites, lower Proterozoic; 15 –

granites, grandiorites, upper Archean; 16 – gabbro-anorthozites, metagabbro norites, metagabbro, upper Archean; 17 – granites, granite-migmatites, lower Archean; 18 – granodoirites, gneiss-granites, plagiogranites, lower Archean; 19 – major fractures (I – Karpinsky, II – Keivy-Uraguba, III – Pechenga-Varguza, IV – Lapland-Kolvitsky); 20 – zone of maximal Caledonian activation; 21 – diatremes; 22 – the Kola super deep borehole.

4.1. Geological setting of the localization area of the Pechenga system

The Pechenga area is confined to the north-west flank of the Central Kola sedimentary-volcanogenic belt (Kolskaya…, 1984), **Fig. 3.** Borehole SG-3 reached the northern part of this structure. This area is one of best studied in the Kola region due to a good nakedness and availability of the Pechenga ore field. The geological section starts with the Loustarinsky series – a sedimentary-volcanogenic rock mass which accordantly or insignificantly discordantly along the contact overlays metamorphic complexes in the northern setting of the structure (Zagorodniy et al., 1964). The basement of the Loustarinsky series is composed of mufic lavas of andesite-basalt composition. Petrogeochemical characteristics of rocks and their composition allow to state that lavas are formed as a result of a volcanic activity of the island arc type or the continental margin type. Upper strata contain high-grade titanic mafic volcanites and more acidic volcanites up to dacite composition. The volcanogenic strata are underlain and overlaid by terrigenic sediments and carbonate rocks. A higher alkalinity of basaltoids, their geochemical characteristics, as well as sedimentation conditions suggest that the layer was formed in conditions of transition from the rift zone to the active continental margin. Rocks of Loustarinsky series are overlaid by tholeiitic pillow lavas of basalt composition of Nickelsk series. Years of field work, as well as drilling results from SG-3 support a tectonic origin of the contact between pillow lavas of Nickelsk series and arkosic gravelites of Loustarinsk series (Kolskaya…, 1984). The section of basalt pillow lavas contains randomly distributed lenticular bodies of picrites and shows a band of acidic vitroclastic tuffs showing signs of underwater landslip deformations. A silica acid content is widely varied in tuffs, reaching 75-80%, what is connected with silicification of rocks. A close association of picrites with tholeiits of oceanic type suggests that they are derivatives of oceanic islands. Underwater or inland volcanoes may be sources of acid pyroclastic flows.

Sedimentary rocks form a thick rock mass in the middle part of Nickelsk series. This productive rock mass is prevailingly composed of volcanoclastic sediments and mafic pyroclastics, enriched with coaly substance and syngenetic sulphides. Structural-textural features of rocks of these strata allow us to place them among analogues of modern submarine turbidite flows. The productive strata host numerous bodies of ore-bearing gabbro-verlites to which the copper-nickel mineralization of the Pechenga ore field is related (Kazanskiy et al., 1980). Disposition of gabbro-verlites and picrites in the section of ocean-type rocks, geochemical features of rocks and signs of intrusive introduction of gabbro-verlites into sediments suggest that the formation of the picrite-gabbro-verlite association corresponds to the conditions of modern oceanic islands volcanism. Gabbro-verlites are considered to be subvolcanic formations. Gabbro-diorites as heat and ore feeders are the analogues of these rocks in modern hydrothermal-magmatic systems of island arcs.

4.2. Hydrogeological description of the Pechenga high-temperature hydrothermal system

Studies of hydrothermal activity within Quarternary volcanism areas helped to single out a geologic-hydrogeologic structure named «hydrothermal system». Hydrothermal systems generally imply the upper crust thermal anomalies confined to certain geological structures and bound to a surface hydrothermal discharge. A heat flux within these thermal anomalies exceeds the Earth mean parameters by 50-100 times. This makes it possible to assume that heat transfer is conducted by means of convection here. Water and water vapor are heat carriers in the nearsurface conditions. Hydrothermal systems are considered to be high-pressure hydrogeological structures occurring in the earth's crust when water-bearing layers are penetrated by a deep-level heat carrier – a supercritical water fluid or a magmatic melt (Belousov, 1978). The given concept changed into «geothermal system» in consequence of drilling programs implementation on dry hot rocks and lately a new scientific school has been formed: a research in hydrothermal-magmatic convective systems (Giggenbach, 1990; Belousov, Rychagov et al., 1998; Rychagov, Belousov et al., 2002). There has been established a genetic interconnection between volcanism and hydrothermal activity, conditioned by the single upper-mantle source of an anomalous heat flow, development of which determines a long evolution of hydrothermal-magmatic convective systems. The Pechenga super deep-level hydrothermal system does not fit into the given definition only by one parameter – a heat flux value, which exceeds the Earth mean level only by two times at the bottom of hole SG-3 (Moiseenko, Smyslov, 1986). However, firstly, the lower parts of this system are much deeper, where a rate of heat flow is higher; secondly, present-day data attest the existence of hydrothermal-magmatic convective systems at low rates of heat flows.

Only hydrogeology of the upper levels of the Pechenga system has been studied in a detail. The nearsurface part of the structure accommodates the homonymous super-artesian basin, disposed in the zone of exogenous jointing (Kolskaya…, 1984). Cold ultrasaltless and saltless waters of hydrocarbonate-calcium composition with a mineralization of up to 0.5 g/l are circulating here. Sulphide zones in rocks contain sulphate-calcium solutions (a content of sals ≥ 2 g/l), that occupy a part of the section and sometimes propagate throughout the zone of exogenous jointing. Their formation may be associated with sulphide oxidation. A composition of dissolved gases is evidential of the atmospheric origin of waters. Hydrothermal solutions at depths of more than 2-3 km have higher alkalinity (pH = 8,5-9,4) and a chloride-hydrocarbonate sodium-calcium composition with a mineralization of 3.7 to 51 g/l. There were established affluxes of chloride-sodium (possibly, hydrocarbonate-chloride) solutions at a depth interval of 5345 to 7000 m. Below 7050 m they become high-pressure highly mineralized chloride-calcium and chloride-calcium-sodium solutions. High pH values of waters drilled open by SG-3 are evidential of an active ongoing «hydrogen metamorphism» of rocks in the given geological section at present. A complex of hydrothermal minerals is formed as result of a hydrogen metamorphism: calcium zeolites, potassium-containing micas, adularia, quartz, montmorillonite and other (Naboko, 1962). The hydrothermal solution upon interacting with allumosilicate rocks develops a higher amount of dissolved SiO_2, a part of

which changes into colloid silica acid (silica gel) that actively participates in cation sorption (Belousov, Rychagov et al., 1998). High pH values are typical for porous solutions of present-day hydrothermal systems, having a certain chemical composition similar to waters of the Pechenga system. We assume, that this analogy may indicate a long circulation of solutions within the Pechenga structure interior. In all, the available hydrogeological and hydrochemical data allow to draw only a general analogy between a super deep-level hydrothermal system and hydrothermal-magmatic systems of modern volcanism areas. A detailed analysis of altered rocks and minerals may contribute to study of hydrotherms.

4.3. Geothermic investigation

A temperature growth rate within crystalline basement rocks of the Baltic Shield is 2 - 2,1^{o}C per 100 m, what is 2 times higher than the predicted geothermic gradient (Kolskaya…, 1984; Kolskaya…, 1998). A temperature at a depth of 10 km is ≥ 150-180^{o}C, at a depth of 12 km – 216^{0}C (Borevsky et al., 1995). Similar data were obtained from other super deep boreholes – Minnibaevsk, Novo-Elkhovsk and Saatlin (Moiseenko, Smyslov, 1986; Muslimov et al., 1980; Neprimerov et al., 1983). Experience of geothermic investigation at numerous boreholes of the Kuril-Kamchatka region and at geothermal deposits worldwide allows to assume the following. Since the whole rock mass drilled by SG-3 was water-bearing, measurements of temperatures were taken in water, not at the contact. Forced convection, inevitably arising in the borehole even after long standing, also causes some problems. Besides, the available hydrothermic information makes it possible to assume both the presence of an ascending heat flux, conditioned by temperature difference between upper and lower levels of the earth's crust, and the existence of lateral movement of hydrotherms in laminated rock mass, what significantly complicates taking measurements of temperatures of separate blocks of rocks in the geological structure under study and does not allow to unambiguously interpret the origin of internal variations of heat flux. Thus, special investigations performed by N.N. Khristoforova with co-authors showed that the temperature gradient in crystalline basement rocks amounted to 3^{o}C/100 m. The temperature curves clearly show a regular interstratification of rocks with a high and low thermogradient. Low- and high-gradient layers are considered to be zones of convective heat and mass transfer (Khristoforova et al., 1999). Considering above, we suppose that real temperature regime within the structure of the Pechenga hydrothermal system may substantially differ from that accepted. This problem can be resolved by means of a set of other techniques (thermobarometry, petrological, isotope-geochemical etc.,) and by comparison with other better studied in this respect modern hydrothermal-magmatic systems. A reason for this conclusion is suggested by important genetic inferences deduced by investigators of the Pechenga structure. They assume that the structure underwent the development stages of underwater oceanic volcanic ridge and island-arc uplift. The analogues of these structures are middle oceanic ridges that host submarine high-temperature hydrothermal systems depositing massive and embedded sulphides in paragenesis with silica minerals (Bogdanov, 1997). The Pechenga present-day hydrothermal system inherits the geothermic regime that took shape at the early stages of the infancy of the crust in the Baltic shield area. Its basic distinction from hydrothermal systems of present-day volcanism is in a deeper localization of a convective hydrothermal cell. This imposes on evolution of super deep-level hydrothermal systems some peculiarities: hydrodynamic isolation of water-bearing collectors is possible and hydrotherms may be positioned in a substantially different area of P-T diagram for aqueous solutions as compared to the solutions of the modern volcanism regions. If thermodynamic conditions of "volcanic hydrotherms" are close to the boiling curve **(Fig. 4)**, then the solutions of "platform hydrotherms" adjoin the line of an average geothermic gradient. In this respect, a more detailed study of such systems is needed to undestand the realization mechanisms of geochemical barriers.

5. ADSORPTION PROPERTIES OF SILICIC ACID AND SILICA IN PROCESS OF FORMATION OF ORE-FORMING HYDROTHERMAL-MAGMATIC SYSTEMS

The issue of concentration of ore elements in the hydrothermal process has been open to discussion up to now. It is quite a common presumption that highly-diluted solutions infiltrate through fractured rocks in large amount thus forming ore deposits over a long time period. Such solutions contain chlorides, fluorides, sulfates, bicarbonates, sulfides of alkaline and heavy metals; silicates of alkaline, alkaline earth and heavy metals; dissolved gases – CO_2, H_2S, SO_2 and

others. Many researchers consider diluted solutions to be exceptionally true (ion-disperse), whereas others assume, that hydrothermal ore-forming solutions are mainly colloid (Chukhrov, 1955; Boydell, 1925). There are notions about moving of ore elements both in alkaline and acid environments. Without diminishing the meaning of different types of solutions in transferring and deposition of chemical elements and their compounds, we would like to address the behavior of siliceous acid and silica within the interior of the present-day hydrothermal-magmatic system in view of the wide distribution of silica minerals within the structure of the systems.and heavy metals; dissolved gases – CO_2,

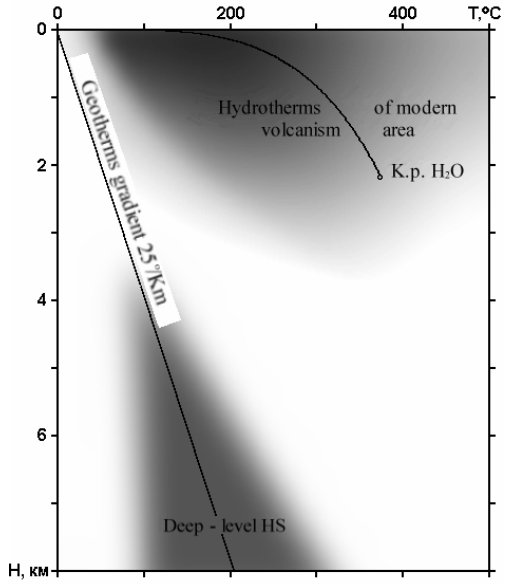


Figure 4: P-T diagram of modern volcanism hydrothermal systems and deep-level hydrothermal systems of ancient platforms.

Hydrothermal solutions in high temperature hydrothermal-magmatic systems of the modern volcanism areas are saturated and often over-saturated due to alteration of P-T conditions during overflow of hydrotherms at different hypsometric levels and from some hydrogeological structures into others. It has been established that hydrothermal solutions, containing significant amounts of siliceous acid, have a high adsorption capacity with regard to cations of various metals, in particular, the heavy ones (Ayler, 1982). Properties of hydrosols of siliceous acid substantially differ from those of ordinary sols (Eytel' , 1962). After joining cations of metals they do not coagulate but form a gel instead as temperature of hydrotherms decreases during dispersion of heat in enclosing rocks when steam forms and solutions mix with cold waters. Effective precipitation of silica in the form of cryptocrystalline quartz and chalcedony occurs when hydrotherms, saturated with siliceous acid, mix with seawater containing Mg. It is known, that the coagulating effect of cations depends on atomic mass of elements (Cs has a stronger effect, Li – a weaker one) and valence: under influence of chlorides of alkaline metals, hydrosol of siliceous acid coagulates slowly while $Ba(OH)_2$ or solution $AlCl_3$ precipitates this sol immediately. It is experimentally established that an especially strong coagulation effect of Al cations, released during the interaction of hydrotherms with volcanic allumosilicate host rocks, is manifested with respect to hydrosols of siliceous acid, amorphous colloid particles and coarser weighed particles of quartz (Belousov et al., 1998). Liophillic sols, whose properties are shared by hydrosols of siliceous acid, are capable of stabilizing liophobic sols (similar to suspensoids of metals and sulfides) having a high sensitivity to the coagulating effect of cations. Liophillic sols envelop the particles of a liophobic sol thus forming a shell which significantly strengthens resistance of the latter to coagulation (for instance, sulfides of metals occurring in underwater-marine high temperature hydrothermal systems). That is why liophillic suspensoids (siliceous acid in hydrotherms) of this type act as protecting colloids.

Seabed «black smokers» is an illustrative example of interaction of siliceous acid colloids and sulfides in hydrothermal solutions. Underwater high temperature hydrotherms, localized in the zone of mid-ocean ridges, are formed by means of seawater infiltrating into the innermost depths of oceanic lithosphere to a depth of 10 -11 km and more towards the influence zone of magmatic hotbeds. Upon mixing with previously formed hydrotherms containing sols of siliceous acid, they lose Mg. Marine hydrothermal solutions thus metamorphosed continue to get saturated with sols of siliceous acid and gases (CO_2, H_2S, SO_2 and other.) while migrating within enclosing rocks. Carbon dioxide retains ions of Ca and their concentration increases what causes interaction with siliceous acid sols and formation of Ca-silicagel, which differs from the common form of H-silicagel by a higher ($\geq$ 50 times) adsorption capacity with respect to cations of metals. Simultaneously, cations of metals Fe, Cu, Zn, and others react with hydrogen sulfide, which results in the formation of a colloid solution of sulfides, protected from coagulation by siliceous acid sols. Besides, minus charged sols of sulfides adsorb cations of metals, insignificantly present in hydrotherms. We can summarize that amorphous silica and anhydrite have high adsorption capacities with respect to a large number of cations.

5.1. Solubility of silica in hydro- and lithosphere

Silica is dissolved and precipitates at a larger part of the earth surface. As was described earlier, soluble silica is mostly produced as a result of destruction of silicates under atmospheric conditions. This process is the leading one in the active volcanism regions, where the interaction of meteoric, underground and seawaters with magmatic rocks is constantly occurring. Concentrations of silica in river waters range within 5-35 mg/kg^{-1}, sometimes reaching 75 mg/kg^{-1}. A concentration of silica in seawater is 2 to 14 mg/kg^{-1}. A concentration of silica in near surface waters of the Pacific ocean may be as low as 0.3 mg/kg^{-1}. Inasmuch as seawater contains 2-14 mg/kg-1 of silica, it is assumed that these values represent an equilibrium solubility of colloid allumosilicate in suspension. It is experimentally proved that pure amorphous silica, dissolved in water, yields 11-100 mg/kg^{-1} concentrations of monosiliceous acid (Eytel' , 1962). In the presence of multi- charged cations of metals (Fe, Al, etc.), colloid silicates with a significantly lower concentration of monosiliceous acid are formed. Addition of Al cation reduces solubility of amorphous silica approximately from 110 to 10 mg/kg^{-1}. Mn and Mg may be reaction-capable and may precipitate soluble silica thus reducing its concentration in water down to 0.3 mg/kg^{-1}. The role of Mg in precipitation of silica from hydrotherms in high temperature hydrothermal systems, where a concentration of silica amount to 600 mg/kg^{-1} and more, is decisive in the emergence of volcanogenic-siliceous formations, constituting the peripheral part of hydrothermal-magmatic systems at volcanic underwater ridges, oceanic islands and island arcs. Volcanogenic-siliceous formations are widely spread in the ancient geosynclinal areas. A similar process takes place in the underwater hydrothermal systems (Von Damm et al., 1985).

5.2. Silica gel in high temperature hydrothermal systems

The formation of silica gel can be considered as a two-stage process. During the first stage molecules of $Si(OH)_4$ condense forming colloid particles. Further slow growth of size of particles is the only subsequent alteration in the diluted solution. When a concentration of silica in solution amounts up to 1% such primary particles are already able of compensating together forming an open and yet continuous all-pervading structure. In this manner, a certain degree of hardness is provided for the given environment. Condensation takes place with formation of siloxanic combination (Si-O-Si). At the second stage, due to impossibility for two particles to coincide on a common section of surface, a number of Si-O-Si combinations between particles will be less as compared to a number of combinations within the particle itself. These siloxanic combinations are adequate only for binding adjacent particles in the fixed position with respect to each other, which leads to formation of hard, high-porous, interwoven three-dimensional grid of forked chains. When studying behavior of silica gel in water solution it is necessary to account for the influence of pH, which is roughly shown on Fig. 5. ABC curve characterizes the behavior of silica when no salts are present in the system.

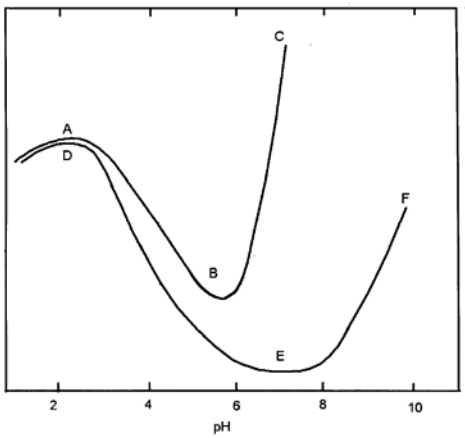


Figure 5: Impact of silica on the process of gel formation in absence of sodium salts (ABC curve) and in presence of sodium salts (DEF curve) (Eytel, 1962).

The longest stability of sols with the longest period of gel formation is at a pH = 1,5-3, and the shortest stability of sols with the fast process of gel formation is at a pH = 5-6. Above a pH = 7 gel does not form at all, because in this case particles of silica charge and grow. DEF curve reflects the behavior of silica when electrolytes are present in the system. The presence of salt causes reduction of a particle surface charge. To form the siloxanic combination capable of irreversible retention of sol particles, the catalytic effect of hydroxyl ions is needed. This can be proven by the fact that the speed of silica gel formation with a pH = 3-5 increases with an increase of pH and in proportion to a concentration of hydroxyl ions. Above a pH = 6 the lack of hydroxyl ions does not restrict the speed of gel formation any more. However, the aggregation speed decreases due to a less frequent number of particle collisions caused by an increased value of their surface charge; that is why it decreases with increase of pH. When salts are not present in a range of a pH = 8-10, sols are usually stable. Below pH = 1,5 the presence of traces of HF has a catalytic effect on aggregation and gel formation.

The given characteristics of aqueous silica solutions and silica gel formations relate to hydrotherms of the active volcanism regions. High temperature hydrotherms are aqueous silica solutions, which ceaselessly produce silica gels over the whole life span of active hydrothermal-magmatic systems. Silica gels interact both with cations of metals and with other colloidal particles having the positive electrostatic charge, what results in the formation of new compounds. Thus, combining of silica gel particles with hydroxide of Al causes the formation mixed-layer clayey minerals of the montmorillonite group (Eytel' , 1962). The similar interaction with colloidal formations of Fe promotes the formation of the minerals of the chlorite group, etc. It seems to us that many mineral complexes within hydrothermal systems of the active volcanism regions are formed on the basis of silica gels or with their indirect participation.

6. DISCUSSION OF RESULTS

The received now radioactive wastes should be presented in the different physical-chemical forms: in a firm form (contaminated equipment, stuffs, soils etc.), hardened form (bituminous, cement and glass-like units) and liquid form (radioactive solutions and pulps stored in special containers and open basins, and also solutions, pumped in deep horizons of underground waters). According to requests of IAEA the disposal of radioactive wastes (RAW), including liquid RAW, should be effected in special storehouses and underground depositories. For medium- and low-active wastes with short-lived nuclides the disposal in a liquid for (injection) in deep permeable formations and by the way of hardening pulps in low-permeable rocks is supposed. In connection with complication of a solution of a problem of reliable isolation of radionuclides and major financial costs on technology on processing, temporary storage and disposal of waste, the simple and economic method of liquid wastes disposal in deep seated aquifers with delayed water exchange was applied. Such method implemented by activity of the Siberian chemical plant (Tomsk), Rock-chemical plant (Krasnoyarsk), Research and development institute of nuclear reactors (Dimitrovgrad). In OкRidge National Lab (USA) the exhaust-and-forced method of injection in a strata of shales liquid medium-active wastes with their after hardening on depth practised. For this purpose through injection hole under excessive pressure injected a mixed fluid of waste with portland cement and clay. The injected mixture organized strata-like injection in a cavity of a fluid fracturing of a layer, which one at setting cement cured. The designed and applied technologies of processing of liquid RAW do not ensure their full rendering, and only allow to reduce volumes and to transfer in more stable form (i.e. to conclude them in glass, concrete, bitumen etc.). After processing the secondary waste or concentrates requiring of a storage and disposal will be derivated. The deep disposal of the liquid RAW directly after their formation takes out a numbers of complications with the circulation of “secondary” waste, demands essentially smaller costs wearing unproductive character. Therefore for some types of RAW the deep disposal was long time unique substantial technology and now prolongs to attract attention of the experts. There is a constant accumulation of the radioactive wastes. General radioactivity of waste, derivated at the Ministry of Atomic Energy of the Russian Federation, estimates in 3 - 4 billion curie. Their processing, reliable controlled storage in engineering facilities and final disposal becomes complicated in connection with increase of requests to observance of the existing domestic and international standards and rules on a guard of population and environmental protection. The further steps on development and introduction of new more reliable and economic methods disposal RAW are apparently indispensable.

Thermal energy and matter coming up to the surface of the Earth in a form of magmatic melts, volcanic gases and hydrothermal solutions form volcanogenic formations. As a result, long-existing hydrothermal-magmatic systems, the roots of which are sunk to the depth of the upper mantle, are formed. These systems control distribution (flows) of ore-forming fluids in the earth crust and are self-isolating. Self-isolation of hydrothermal-magmatic systems leads to formation of ore deposits and geothermal fields and serve as a basis for design of models for formation of artificial ore deposits. Minerals of silica, zeolites, chlorite-smektite and other hydrothermal clays, which are good sorbents for cations, especially cations of heavy metals are formed in the zone of active circulation of hydrothermal solutions and their interaction with rocks. The most important feature of such natural mineral sorbents is continuity of their reproduction within tens of thousands and hundreds of thousands years. There form thick strata of duplicate hydrothermally altered rocks retaining either in a dispersed form or in certain zones metals as mineral compounds being in sustainable thermo- and hydrodynamic balance with the environment.

In recent years, Institute of Volcanology of FED RAS have been developing a study of adsorbing properties of high temperature hydrothermal systems, located in volcanic islands. Interiors of such present-day geological structures constantly reproduce adsorbents with a negative charge having a capability to retain all metals within their structure. Gels of silicic acid (silica gel) are the most optimal sorbents. Unlike other minerals with adsorption properties, silica gel, following sorption of metals, continues to move together with hydrotherms towards their discharge point and precipitates as temperature of solution falls. Sea water, having a content of dissolved magnesium and calcium, is an impassable barrier for silica gel inasmuch as the said elements practically completely precipitate sols of silicic acid. Thus, ingress of mineral elements, including radioactive ones from hydrothermal systems into sea water area is either completely excluded or their content in sea water shall not be higher than that of clarke. Tests, carried out on volcanogenic rocks forming water-containing strata of the Pauzhetka (The South Kamchatka) and North-Paramushir (the Kuril islands) hydrothermal systems, have shown, that uranium is sorbed by them completely and concentrations of radioactive isotopes of strontium and caesium decrease by ≥ 70%. On this basis there arises a possibility to control processes of liquid radioactive waste disposal: to dissipate radioactive elements in volcanogenic formations till safe concentrations are reached or immobilize them in mineral aggregates at geochemical barriers and produce artificial ore deposits (patent RU 2069397 “Burial techniques of liquid radioactive waste”, registered in the Russian Federation State Register of Inventions of 20.11.1996; patent RU 2111564 “Burial techniques of radioactive and other chemically-hazardous waste”, registered in the Russian Federation State Register of Inventions of 20.05.1998). And local geothermal resources may cover energy consumption during technological operations.

The comparison of present-day island arcs with palaeo-platforms has shown that the interiors of platforms at a depth of more than 2-3 km develop high temperature hydrothermal systems, thermodynamic parameters and geological structure of which are analogous or similar to those of hydrothermal-magmatic systems of volcanic regions. For instance, using the Petchengsk deep-level hydrothermal system in the aggregate with other natural objects and their technogenic analogues as an example, there have been showed possibilities of its use for study of geotechnical conditions of isolation of nuclear materials when disposed of directly into natural objects. In terms of practical application, the assessments are primarily focused on volcanogenic formations of platform regions, including rocks of existing or potential nuclear sites of Kolsky peninsula, industrial amalgamation “Mayak”, Siberian chemical plant and Krasnoyarsky mining-chemical plant of the Ministry of Nuclear Energy, which is not exclusive of consideration of conditions and selection of analogous sites in the Far East. The Pechenga super deep-level hydrothermal system along with other natural objects and their technogenic analogues has been viewed under aspect of chemical technology of allumosilicate processing. We have demonstrated possibilities to use such systems for study of geotechnical conditions of isolation of various industrial waste when disposed of or buried directly within natural objects. Accenting the given issue, the authors proceed from proposition that water, circulating in the crust, presents hydrothermal solutions formed from interaction of underground hydrosphere with heat flux of the Earth. An intensive study of hydrothermal solutions (chemical composition, dynamics, water-rock interaction processes, conditions of transfer and deposition of chemical compounds) will allow to find qualitatively new technological approaches in the sphere of modern mineral- and ore-formation and creation of artificial ore deposits. Relevant regime observations and field experimental investigation were commenced at hydrothermal-magmatic systems of the Kuril-Kamchatka region. The stated approach expands the outlook for choice of sites and conditions of isolation of industrial waste at geological platforms.

7. CONCLUSION

Earlier on, when studying Petchengsk syneclise, no sufficient attention was paid to hydrothermal solutions, formed out of interaction of underground hydrosphere with Earth’s heat flow. Our researches have shown that it is impossible to resolve the problem of safe storage and disposal of radioactive materials in geological formations of palaeo-platforms without a detailed study of present-day hydrothermal solutions – their chemical compositions, dynamics, water-rock interaction processes, conditions of formation of hydrothermal minerals; leaching, transport and deposition of chemical compounds - at all levels of formation and circulation of solutions. A research of the said processes carried out solely on deep sections of the Kolsky peninsula is going to be very complicated and extremely expensive. Employment of proposed sites of the Kuril-Kamchatka region for these purposes will allow to suggest new technological decisions on safe storage and disposal of radioactive materials in the district of nuclear sites of the Kolsky peninsula, Siberian chemical plant, Krasnoyarsky mining-chemical plant of the Ministry of Nuclear Energy and other nuclear sites of palaeo-geological platforms.

The work has been done with a financial support of the Russian Fund of Fundamental Research (projects 03-05-64044a and 04-05-79051k).

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