**MUDANÇA DE FASE EM SISTEMAS HIDROGEOQUÍMICOS DE EQUILÍBRIO LOCAL
COMO MECANISMO DE OCORRÊNCIA DE PRECURSORES FLUÍDICOS DE
TERREMOTOS**

**PHASE REARRANGEMENT IN LOCAL BALANCED HYDROGEOCHEMICAL SYSTEMS
AS A OF FLUID EARTHQUAKE PRECURSORS MECHANISM OCCURRENCE**

**ФАЗОВАЯ ПЕРЕСТРОЙКА В ЛОКАЛЬНО-РАВНОВЕСНЫХ ГИДРОГЕОХИМИЧЕСКИХ
СИСТЕМАХ КАК МЕХАНИЗМ ВОЗНИКНОВЕНИЯ ФЛЮИДНЫХ ПРЕДВЕСТНИКОВ
ЗЕМЛЕТРЯСЕНИЙ**

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RESUMO

Cientistas de todo o mundo estão fazendo grandes esforços para resolver o problema da previsão de terremotos. Várias dezenas de diversos fenômenos e sinais foram descobertos que poderiam servir como precursoros de terremotos. Mas esses efeitos descobertos se mostraram muito instáveis e tinham um caráter de mosaico no tempo e na área de seu desenvolvimento. Eles nem sempre apareceram e nem antes de cada forte terremoto. Existem muitos casos em que terremotos ocorreram sem o aparecimento de precursoros. Tais "fenômenos" instáveis não nos permitem formular modelos adequados para a preparação de fortes terremotos e métodos para prevê-los. Entre os precursoros bem estudados e encorajadores de terremotos, os mais promissores são as anomalias hidrogeodinâmicas e hidrogeoquímicas, que têm antecedentes científicos razoavelmente sólidos. Nesta classe de precursoros, o mecanismo de sua formação durante a preparação de terremotos permanece discutível. Há perguntas sobre a fonte de concentrações anormais e seu transporte durante um curto período de tempo nos pontos de observação. No presente estudo se afirma que o principal mecanismo para a ocorrência de anomalias exóticas de fluidos transientes é a reestruturação interfásica no sistema hidrogeoquímico mais equilibrado localmente nos objetos de observação como reação aos efeitos da preparação de fortes terremotos. A reestruturação da fase interna no próprio sistema hidrogeoquímico é realizada através do relaxamento estrutural como resultado da ocorrência de reações físico-químicas de troca rápida.

Palavras-chave: *geodinâmica moderna, transições de fase, estado físico das águas subterrâneas, estresse tectônico, previsão de terremotos.*

ABSTRACT

Scientists all over the world are making tremendous efforts to solve the problem of earthquake prediction. Dozens of different phenomena and signs that could serve as precursors of earthquakes are found. But these effects were very unstable and had a mosaic character in the propagation of development and time. They appeared not always and not before every major earthquake. There are many cases where earthquakes occurred without the manifestation of any precursors. Such non-stationary phenomena do not allow formulating adequate strong earthquakes preparing models and their methods of predicting. Among well-studied and encouraging earthquake precursors, hydrogeodynamic and hydrogeochemical anomalies are considered to be the most promising, which has a sufficiently substantiated scientific background. In this class of precursors, the mechanism of their formation during the preparation of earthquakes remains debatable. There are several problems due to abnormal concentration sources and their transport during a little time period in the observation points. In the proposed work, it is proved that the main mechanism of the occurrence of exotic transient fluid anomalies is the interphase rearrangement in the local balanced hydrogeochemical system as a reaction to strong earthquakes factors preparing influence. The internal phase reorganization in the hydrogeochemical system is realized through structural relaxation as a result of the occurrence of rapid physicochemical reactions exchange.

Keywords: *modern geodynamics, phase transitions, the physical state of groundwater, tectonic stress, earthquake prediction.*

АННОТАЦИЯ

Ученые во всем мире прилагают громадные усилия для решения проблемы прогнозирования землетрясений. Обнаружены несколько десятков различных явлений и признаков, которые могли бы служить предвестниками землетрясений. Но эти обнаруженные эффекты оказались весьма неустойчивыми и имели мозаичный характер как во времени, так и на площади их развития. Они проявлялись не всегда и не перед каждым сильными землетрясениями. Имеются немало случаев, когда землетрясения происходили без проявления каких-либо предвестников. Такие нестационарные «феномены» не позволяют сформулировать адекватные модели подготовки сильных землетрясений и методы их прогноза. Среди хорошо изученных и обнадеживающих предвестников землетрясений наиболее перспективными считаются гидрогеодинамические и гидрогеохимические аномалии, которые имеют достаточно обоснованные научные предпосылки. В этом классе предвестников дискуссионным остается механизм их формирования в процессе подготовки землетрясений. Существуют вопросы источника аномальных концентраций и их транспорт за небольшой отрезок времени в пунктах наблюдений. В предлагаемой работе доказывается, что основным механизмом возникновения экзотических быстротечных флюидных аномалий является внутрифазовая перестройка в самой локально-равновесной гидрогеохимической системе в объектах наблюдения как реакция её на воздействие факторов подготовки сильных землетрясений. Внутренняя фазовая перестройка в самой гидрогеохимической системе реализуется путем структурной релаксации в результате протекания быстрых обменных физико-химических реакций.

Ключевые слова: *современная геодинамика, фазовые переходы, физическое состояние подземных вод, тектонический стресс, прогноз землетрясения.*

1. INTRODUCTION

Underground water-gas systems as the most mobile component of the earth's crust in various geostructures are sensitive locally-equilibrium hydrogeochemical systems with a very thin structural-phase ratio in the rock-water-gas triad. Such systems are sensitive to external influences and adequately respond to them (Abdullaev, 2002). The formation of the physicochemical composition of underground fluids is directly dependent on the composition of the host rocks and the hydrogeological conditions for their localization (Posokhov, 1975; Driver, 1985; Kraynov and Shvets, 1992). An important area of research to identify the main mechanisms of the occurrence of hydrogeochemical precursors of earthquakes is the dynamics of rapid changes in the ion-salt composition in the hydrogeochemical systems themselves (Talabi, 2018; Warsaw, 1985). It is necessary to proceed from the postulate that in calm seismic situations in conditions of "rest" of the geological environment, the functioning of water-gas systems in time is characterized by local equilibrium in individual hydrogeological structures (Hörbrand *et al.*, 2018).

Studies of the relationship and interdependence of the chemical composition of

groundwater and external factors (changes in atmospheric pressure, temperature, tidal forces of the Moon and the Sun) and endogenous factors (the occurrence of earthquakes) were carried out on the analysis of long-term continuous observations at the Almaty Seismic Active Prognostic Testing Ground (APF) in Southeast Kazakhstan (Figure 1), where since 1980. continuous monitoring of variations in a wide range of parameters of the gas chemical composition of groundwater is carried out in order to search for earthquake precursors. To understand the causes and mechanisms of the occurrence of exotic spatiotemporal hydrogeochemical anomalies that occur on the eve and in the periods and implementation of strong earthquakes, we consider the main key properties of these phenomena. According to (Abdullaev, 2002; Nikolaev, 1984), such anomalies are intermittent alternating changes of the so-called "indicative" parameters, which have 4 main morphogenetic groups: "bays", "humps", "jump-pulsation" and spontaneous "bursts" that are in amplitude altered surpass background levels by 20-50% to 100%, and sometimes times (Movchan and Yakovleva, 2017). They in the square have the property of "flickering" in time (Aitaliev *et al.*, 2005; O'Neill *et al.*, 2018).

In genetic terms, they are "returnable" i.e.,

after the event, and the “marker” parameter returns to its original level. The last property of the anomalies is long-range action, which is found in tens and hundredths of a km. from the source of earthquakes, i.e., they arise not only in the focal zone, but also far beyond its borders. A long search for adequate mechanisms reflecting all these phenomenal properties leads us to the conclusion that they must be “in themselves” and appear due to tectonic factors of earthquake preparation. In this aspect, we proposed a phenomenological model of the formation of exotic seismohydrogeochemical anomalies in the field of tectonic forces and the interaction of reversible chemical processes occurring between substances (Figure 2, right block) and a locally equilibrium open hydrogeochemical system (central block). In this model, all processes are reversible and can develop everywhere and not only in the focal zone covered by the influence of abnormal stresses but also beyond. The most important properties of exotic hydrochemical anomalies are explained quite convincingly here. In this interconnection, all processes universally occur at the vapor-water contact: (hydrolysis, dissolution, precipitation, ion exchange, complex formation, fluid migration, sorption-absorption, wringing of pore solutions, and many others, which are shown in the left block of Figure 2.

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2. MATERIALS AND METHODS

To elucidate the mechanism of the ongoing restructuring in hydrogeochemical (GHC) systems on the effect of seismic processes, the salt composition of groundwater was calculated at the GHG stations Tau-Turgen, Gorelnik, and Chushkaly for 1990. and 1995 (Figure 1) due to the occurrence of two strong earthquakes in this area (Baysorunskoye, $K = 15.3$ and Kegenskoye, $K = 13.7$). Recalculation of chemical analyzes of groundwater from the ionic form to salt was carried out in accordance with the solubility of salts. In the first place, sparingly soluble, and then increasingly soluble compounds are combined. Thus,

according to daily observations at the stations, by converting the results of chemical analyzes from the ionic form to the salt form, a possible hypothetical salt composition was calculated. Using the equivalent form for expressing the chemical composition of groundwater, it is easy to calculate combined ions in salts from the ion-equivalent form. (Posokhov, 1975; Driver, 1985; Kraynov and Shvets 1992; Laptev, 1995).

The ionic form of the chemical composition of groundwater is fully characteristic only of low salinity waters (Rouhi and Kalantari, 2019). With an increase in the concentration of dissolved salts between the ions, the interaction intensifies, and the process of association develops. Associated neutral pairs of CaSO_4^0 , MgSO_4^0 , CaCO_3^0 or charge-carrying molecules $\text{Mg}(\text{HCO}_3)^{2+}$, $\text{Ca}(\text{HCO}_3)^{2+}$ are formed in the solution. The ability of atoms and ions to attach or hold electrons is determined by their electronegativity, which in turn is a characteristic of the generalized acid-base properties of elements, i.e. a measure of how typical for a given element the role of a donor (cationic acid) or an acceptor (base anion) of an electron in compounds (Mattos *et al.*, 2018).

3. RESULTS AND DISCUSSION:

The general scheme of the binding of ions to salts according to the degree of solubility is as follows: Ca^{2+} ions first bind sequentially with HCO_3^- , SO_4^{2-} , Cl^- depending on its content, and then Mg^{2+} and Na^+ bind in the same sequence. On the example of several strong earthquakes that occurred at the Almaty landfill in 1990-1995, we see that at the stations of Chushkaly, Turgen and Gorelnik the possible calculated salts of sodium, calcium, and potassium are Na_2SO_4 , NaHCO_3 , NaCl , $\text{Ca}(\text{HCO}_3)_2$, CaCO_3 , K_2CO_3 the ratio of the concentration of these salts in the hydrogeochemical system itself on the eve of strong earthquakes in time has changed: some increase and others decrease, and the change in salt concentration is in antiphase due to a decrease in some and an increase in other salts. This process is clearly visible on the example of the temporary course of salts at the GKH station of Chushkaly during the period of the strong May and Baysorun earthquakes in 1990. in the center of the Almaty seismic-prognostic test site (Figures 3, 4), where a strong event ($K = 15$) was preceded by three impulse disturbances.

Figure 4 shows the course of the CaHCO_2 salt change at Tau-Turgen station in connection with the indicated earthquakes, which experiences an abrupt change in concentration before the May

earthquake, increasing before the Baysorun event. Figure 5 clearly shows the course of the change in Mg salts over time using another strong Kegen earthquake in 1995 as an example. ($K = 13.7$). Theoretically calculated mineralization of thermal waters on the eve of strong earthquakes shows that a change in the course of mineralization itself can also indicate a change in the geodynamic state around a given well (Figure. 6). Similar calculations at the Tau-Turgen station show that salt formation or decomposition of salts in the rock-water system occurs in the same way as in the first case: the concentrations of some salts increase, while others decrease in antiphase. As a result, a certain stable variation corridor is preserved, which can be seen in salt calculations $Mg(HCO_3)_2$ and $MgSO_4$ (Figure 3). At the same time, salt concentrations change sharply in the antiphase before and during events.

This process is universal and can occur at various depths. The aforesaid can be imported by the example of GHC anomalies, manifested on the eve of the Baysorun earthquake on 11/11/1990. $K = 15.3$ at stations in the Issyk-Kul depression, where we see at different depths (well 6-163 m. Well 20-520 m) the same process is out of phase changes in carbonate equilibrium at station Jets-Oguz at a distance of 65 km from the epicenter. At Dolinka station, at a depth of 1183 m, the antiphase ion change is clearly visible SO_4^{2-} and Cl (Figure 7) during the course in time before the strong Baysorun earthquake at a distance of 45 km from the epicenter. Observations show that a few days before the seismic events, the concentrations of some salts in both cases ($NaHCO_3$, $NaCl$, $Mg(HCO_3)_2$) sharply increased, and some decreased (Na_2SO_4 , $CaCO_3$, K_2CO) in the form of a deep bay, and then returned to original values. Clear instabilities are observed in the balance of salts, i.e., a sharp increase in mineralization before the push and a sharp decrease during the event due to an increase in the proportion of reduced salts (Figure 6). Interestingly, after the seismic event, the level of mineralization recovered.

All these facts indicate that during the preparation and implementation of strong seismic events, the additional substance does not enter the equilibrium GHC system from afar, and anomalies appear as a result of structural relaxation. at the local site of the "steam-water-gas" system. At the same time, theoretical calculations showed that, on the eve of the earthquake, a rather intense association process might be taking place. In connection with the action of abnormal stresses in the locally-

equilibrium system, an internal phase rearrangement occurs in full accordance with the action of the well-known law in the physical chemistry of Le Chatelier (Abdullaev, 1991).

An analysis of the ionic-structural relationships of thermal waters and dynamic parameters (flow rate, level, annular pressure) on the eve of earthquakes shows an extremely unstable geodynamic situation that develops spasmodically in time until the stress release (Baimakhan *et al.*, 2009a, Baimakhan *et al.*, 2009b). To describe the dynamic instability in underground water-gas systems in physical chemistry, the concept of "entropy" (s) is usually used as a thermodynamic function of the state of the system, showing the level of chaos or ordering of the system (Abdullaev, 2002; Abdullaev and Tinibaeva, 2007; Abdullaev, 1991; Kraynov and Shvets, 1992; Nicholas and Prigogine, 1979). In statistical thermodynamics, entropy is the concept of the degree of disorder in the location and movement of a large number of homogeneous structural elements (molecules, ions, atoms, complexons) in some isolated systems. It can be defined by the well-known Boltzmann-Planck Equation (Equation 1).

Where P – the number of different (equally probable) microstates of some isolated system, $k=1,3810^{-16}$ erg / deg (Boltzmann constant). It seems to us that the time course of the development of internal structural relaxation of a local equilibrium GHC system during the formation of concentration anomalies is better characterized in physical and chemical terms through a thermodynamic state – entropy (s).

The vibrational nature of the entropy factor of the basic chemical reactions of carbonate equilibrium in hydrogeochemical systems indicates the formation or decay in time of hydrogeochemical dissipative structures of local partial equilibria. In the thermodynamic aspect, this means an extremely rapid change in the energy state of the system through fluctuations and energy release. This transition is reflected in the course of entropy (S) as the cumulative state of the controlled system, in the form of an alternating jump from "+" to "-", which is illustrated in the fundamental deterministic model in the coordinates: time (x) and intensity of fluctuations (y) in on the eve of the tectonic earthquake (Figure 8). The aforementioned fleeting changes in the basic phase rearrangements in the local equilibrium system in the rock-water-gas triad are fully explained in the framework of the theory of self-organization of geodynamic systems (Nicholas and Prigozhin, 1979) and self-organized

criticality, quite convincingly recently disclosed by P. Buck (2014).

The leading elementary partial equilibrium, of which the local physicochemical equilibrium of the hydrogeochemical system as a whole is taken, is taken as homogeneous basic state elements (its main cells). A theoretical analysis of the information content of such systems shows that main partial equilibria Equations can be distinguished in them, namely (Derr *et al.*, 1974):

- carbonate (Equation 2);
- chlorine sulfate (Equation 3);
- silicate (Equation 4);
- equivalent cation exchange (Equation 5);
- gas exchange (N_2 , O_2 , CO_2 , CH_4).

In real hydrogeochemical systems, all elementary equilibria are closely interconnected and develop in conjunction, as indicated in our phenomenological model (Figure 6). As our experience and direct observations show, the most sensitive equilibrium, extremely fast and the first to respond to changes in the stress-strain state (VAT), is carbonate equilibrium (Abdullaev, 1997; Kraynov and Shvets, 1992; Driver, 1985). It is possible to isolate and calculate the thermodynamic parameters of the following three basic (model Equations) chemical reactions of carbonate equilibrium (Equations 6-8). Where K_p – equilibrium constant

Calculations show that Gibbs free energy (ΔG_e), for example, GGH station Gorelnik on these reactions has the following meaning: a=-6622 cal/mol, b=-2656 cal/mol, c=-1063 cal/mol. Obviously, when a GC system is exposed to the deformation of different signs, a chemical reaction primarily occurs with a smaller energy reserve. It starts earlier and is subject to a much larger oscillatory process than a chemical reaction with a large supply of energy. Based on the foregoing and comparing these reactions with each other in the energy aspect, we can distinguish as a basic chemical reaction of the form: b) as the most sensitive to external energy influences (Equation 9). All these reactions are, of course, the result of the development in groundwater of physico-chemical processes of dissolution, precipitation, complexing, hydrolysis, drainage of gas components from tectonic cracks. From a physical and chemical point of view, a locally-equilibrium hydrogeochemical system in seismically active regions can be represented as a conditional ball consisting of interconnected conjugate partial equilibria that can be deformed into an ellipse under the influence of anomalous

stresses (σ_{an}) during the preparation of a tectonic earthquake (Figure 9).

As a result of this, a total disturbance (ξ) is generated in the system as a result of the transformation of the “ball” into an “ellipsoid”. Such a model is a further development and confirmation of our ideas that the GCH system in seismically active areas is a very thin physicochemical system with an easily excitable and mobile medium that is sensitive to geodynamic changes. Therefore, it adequately reflects transient geodynamic processes, including the preparation of earthquakes.

4. CONCLUSIONS:

In the search for real mechanisms for the emergence of hydrogeochemical precursors of earthquakes, numerous researches, and experimental studies have been conducted. They mainly concentrated within the framework of the concept of transporting new portions of solutions of different composition or individual components to the epicentre zone. Even with the assumption of super-diffusion or convection, or rapid mixing of various fluids in a sound field, it is difficult to imagine the possibility of the occurrence of observed concentration increases of certain components in groundwater by 20-100% or more during the day or several days on the eve of major earthquakes and at distances of tens and hundreds of kilometres from their epicentre. Of course, there should be a completely different mechanism for the emergence of short-term exotic hydrogeochemical precursors, which arise at the observation sites, and it should be able to explain the diversity and phenomenal properties of these anomalies.

In this regard, proposed and justified by us in this work the phenomenological model of interconnections between fast-flowing (sometimes spontaneous) tectonic stresses and the preparation of strong earthquakes most convincingly explains that phase restructuring in open HGC systems is not only the main mechanism for generating anomalies but also the main reason for their occurrence in significant area in seismically active areas during periods of earthquake preparation. These achievements certainly advance the theory of seismic catastrophe prediction.

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$$S = k \cdot 1nP/ \quad (\text{Eq. 1})$$

Figure 1. Almaty seismic hydrogeochemical prognostic test site in South-East Kazakhstan

Figure 2. Phenomenological model of the formation of exotic temporary hydrogeochemical anomalies in the underground waters of seismically active regions during the periods of preparation of strong earthquakes

Figure 3. Time course of antiphase changes of calcium salts at Chushkaly station on the eve of the May (03.05.1990, $K = 12.2$) and Baysorun earthquakes (11/12/1990, $K = 15$): 1 – CaCO_3 ; 2 – $\text{Ca}(\text{HCO}_3)_2$

Figure 4. The time course of changes in $\text{Ca}(\text{HCO}_3)_2$ in time in 1990 at station Tau-Turgen before strong earthquakes

Figure 5. Distribution of Mg salts in antiphase due to the Kegen earthquake (30/10/1995, $K=13.7$) at the Tau-Turgen station: 1 – $\text{Mg}(\text{HCO}_3)_2$, 2 – MgSO_4

Figure 6. The course of the estimated mineralization of thermal mineral waters at the Tau-Turgen station on the eve of the Kegen earthquake (31/10/1995, $K=13.7$)

Figure 7. GHC manifestations of anomalies before the Baysarun earthquake (11/12/90) in the Issyk-Kul depression at various depths

Figure 8. A fundamental model for the development of the time course of entropy (S) of a local equilibrium GHC system (LHHHC) under the influence of tectonic stress

Figure 9. Phenomenological model of phase adjustment in locally equilibrium physicochemical systems with a generation of dissipative concentration anomalies (Abdullaev, 2002)