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Amankeldy Toleukhanov*, Aidarkhan Kaltayev*,
Mikhail Panfilov**

STORAGE OF HYDROGENOUS GAS IN GEOLOGICAL FORMATIONS: SELF-ORGANISATION METHANE GAS

1. INTRODUCTION

The uniform distribution of natural gas over various regions distanced by thousands kilometres from gas producing sites represents a serious economical and technological problem for Kazakhstan. Taking into account that the gas transportation constitutes 70% of the overall price of natural gas, the investments into gas transport from the Caspian region to eastern and southern regions appears to be extremely expensive. Another, a priori more rentable solution might consist of using hydrogenous gas instead of methane, that can be produced from coal by degasification technique. This process may be performed directly at the central and eastern coal basins and consequently does need to transport hydrogenous gas over long distances. Such a gas, called the town or syntheses gas, represents a mixture of hydrogen (60%), methane (12%), and CO_2 and CO (20%) has a high energetic potential and is frequently used as a high-performance carrier of energy capable of replacing up to 60% of the natural gas used in non-industrial activity in several countries (Davison *et al*. 2009).

In the case of producing large amounts of hydrogenous gas, currently there are no problems related to basic techniques of hydrogen production and distribution, but the main technological problem will consists of storing it in order to regulate the difference between permanent or increasing gas production and seasonally modulated gas consumption. The most efficient and most inexpensive method of storing large amounts of hydrogen is to inject them in geological formations like aquifers, depleted gas reservoirs, or salt caverns

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* Al-Farabi Kazakh National University, Almaty
** Univeristé de Lorraine, Laboratoire d'Energétique et de Mécanique Théorique et Appliquée – CNRS, Nancy France

(Zittel and Wurster 1996). The cost is of order \$ 3.5 per 1 GJ (Taylor *et al.* 1986). Several underground storages of hydrogen (USH) or town gas exist in the word, as for instance, Teeside in the UK, in Texas, in Russia, Kiel in Germany, Lobodice in Czechoslovakia, Beynes – an ex-storage in France.

During several tens of years the storage of hydrogen was considered as something déja-vu, to be similar to that of natural gas, which is amplified by the chemical inactivity of hydrogen and its very low solvability in groundwater [Bulatov 1979; Carden and Paterson 1979; Lindblom 1985; Paterson 1983]. Nevertheless, quite unusual behaviour of UHS was discovered by in situ monitoring of the gas composition extracted during the cycle "production" which followed the cycle "injection". These observations (Smigai *et al.* 1990; Buzek *et al.* 1994) revealed high variations of gas composition in time and space. In particular, a significant reduction in the H2 and CO_2 contents and a simultaneous increase in CH_4 contents were observed in the Lobodice town gas storage facility (Smigai *et al.* 1990). Similar phenomena were recorded in Beynes. After several months of injection and storage, at the beginning of the cycle "production" the twofold increase of the methane contents in the reservoir gas and the twofold reduction of the CO_2 + CO contents was observed. The contents of hydrogen decreased by 1.4. The explanation to these observations has been done in (Buzek *et al.* 1994) in terms of the in situ methane generators by methanogenic bacteria which catalyse the reaction between hydrogen and CO_2/CO, by producing methane and water. Further observations have revealed even more unusual effects within the storage facility, such as creating a spatial alternation of the areas saturated preferably by hydrogen or methane. This proved an in situ natural separation of chemical components in space.

The aforementioned effects elicit four essential scientific and applied problems:

1. It is possible to lose significant amount of hydrogen in the reservoir due to bacteria activity.
2. A UHS possesses a mechanism of natural in situ CO_2 decomposition which is considered a positive effect in the light of modern political tendencies towards reducing the contents of acid gases in the atmosphere.
3. A UHS also possesses a mechanism of in situ natural methane production. This effect can be called the natural hydrogen enrichment, taking into account that the potential heat of a unit mass of methane is higher than that of hydrogen.
4. A UHS possesses an additional mechanism which causes natural separation of hydrogen and methane in space, which may play both a positive role (an opportunity to extract pure components from different wells) and a negative role (reduction of the total degree of hydrogen enrichment by methane).

Thus, we are dealing with a natural reactor which partially destroys CO_2 and H_2 and doubles the mass of methane. It is clear that the problem is important for industry as it concerns both the energy sector and ecology. The resulting economical efficiency of such a process can be estimated only after the physical and mathematical modeling of all possible scenarios of the reservoir behaviour. The development of such a model represents the main objective of this paper.

2. METHOD AND THEORY

The first attempt to analyse the transport in USH coupled with bacteria population dynamics was performed in [Panfilov *et al*, 2006], [Panfilov, 2010], and [Panfilov and Rasoulzadeh, 2010] in terms of the single-phase multicomponent model. In the present paper we propose the advanced conceptual model of the process which takes into account the presence of two phases (gas and water), several chemical components (H_2, CH_4, CO_2, CO, H_2O, N_2), chemical reactions caused by methanogenic bacteria metabolism, capability of bacteria to move towards the interfaces between gas and water and to create *neuston*: the biofilm just at the interface between gas and water, and the dynamics of the bacteria population caused by its growth, natural death, chaotic motion in space and oriented motion due to chemotaxis. The example of neuston is shown in Figure 1.

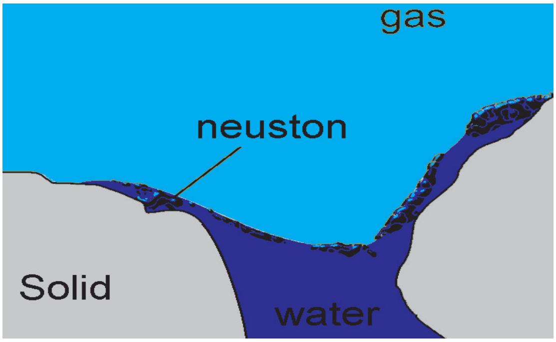

Fig. 1. Two-phase fluid and the neuston: the biofilm at the interface between gas and water at the pore scale

The mathematical model of the process consists of several equations describing the two-phase transport of muticomponent mixture with chemical reactions coupled with the following equation of the population dynamics:

$$\begin{aligned}\frac{\partial n}{\partial t} = {} & \frac{\eta_{ns} c_g^{(2)}(1-\theta)n}{t_{e,ns}(1+a_{ns}c_g^{(2)})} + \frac{\eta_w c_w^{(1)} c_w^{(2)} \theta^2 S n^2}{t_{e,w}\left(S^2+\dfrac{\theta^2 n^2}{n_{wm}^2}\right)(1+a_{w1}c_w^{(1)})(1+a_{w2}c_w^{(2)})} - \frac{n}{t_d} + \\ & + div\left(D_b(1-S)grad\frac{(1-\theta)n}{1-S}\right) + \\ & + div\left(D_b S grad\frac{\theta n}{S}\right) - div\left(D_{ch}(C^{(1)})\theta n grad C^{(1)}\right)\end{aligned} \tag{1}$$

$k=1,2$:

$$\phi\frac{\partial}{\partial t}\left(\rho_g c_g^{(k)}(1-S)+\rho_w c_w^{(k)}S\right)+div\left(\rho_g c_g^{(k)}V_g^{(k)}+\rho_w c_w^{(k)}V_w^{(k)}\right)=$$

$$=\frac{1}{\Omega}G^{inj}c^{(k),inj}-\frac{\phi\gamma^{(k)}(1-\theta)c_g^{(2)}n}{t_{e,ns}\left(1+a_{ns}c_g^{(2)}\right)}-\frac{\phi\gamma^{(k)}c_w^{(1)}c_w^{(2)}\theta^2 Sn^2}{t_{e,w}\left(S^2+\dfrac{\theta^2 n^2}{n_{wm}^2}\right)\left(1+a_{w1}c_w^{(1)}\right)\left(1+a_{w2}c_w^{(2)}\right)} \qquad (2)$$

where n is the number of bacteria in 1 m^3 of the medium, $c_w^{(1)}$, $c_w^{(2)}$ are the concentrations of H_2 and CO_2 dissolved in water, $c_g^{(1)}$, $c_g^{(2)}$ their concentrations in gas phase, $C^{(1)}$ is the total mole fraction of H_2 in both phases, D_b is the bacteria diffusion coefficient, S is the water saturation, ρ is the molar density, ϕ is the porosity, G^{inj} is the molar rate of gas injection, Ω is the total volume of the reservoir, $V_i^{(k)}$ is the transport velocity of component k in phase i, other coefficients are empirical and characterise the kinetics of population growth and death, the intensity of chemotaxis, the attenuation of growth for an abundance in resources, and so on.

3. RESULTS AND EXAMPLES

Equations (1) and (2) represent the coupled system which describes the bio-reactive transport in an underground storage of hydrogen and dynamics of bacterial population. The numerical code has been developed based on the suggested model. The results of numerical simulations and the bifurcation analysis have revealed the appearance of the auto-oscillations which are either the Turing spatial auto-waves or the Volterra spatial and time auto-oscillations, depending on the saturation of water and ratios between several parameters (Fig. 2).

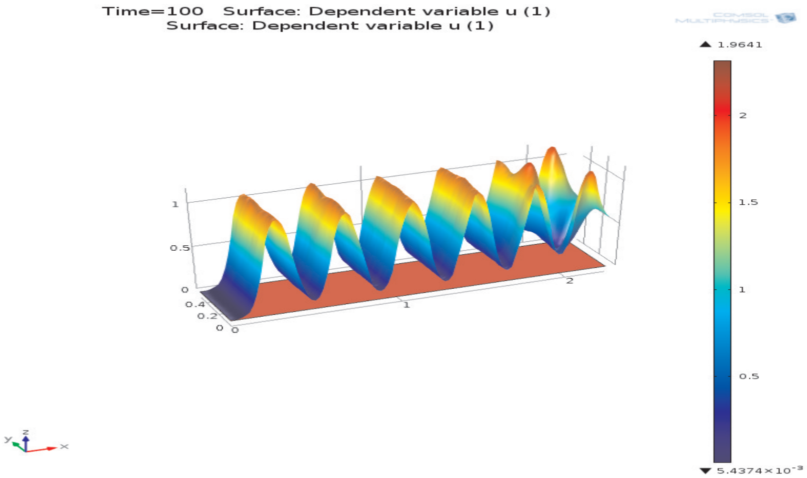

Fig. 2. Auto-oscillations of bacteria population and species concentration in space

The Turing auto-waves are stabilizing in space, while the Volterra waves are almost-periodic both in time and in space. Several scenarios of transition between these two limit regimes are possible. These results explain and confirm the possible natural separation of components in UHS, and the formation of spatial zone oversaturated with an individual component, hydrogen or methane.

4. CONCLUSION

The global behaviour of the UHS is thus very complicated and sensitive to kinetic parameters. The next objective of the research consists of experimental investigation of the kinetic parameters for bacteria population and their implementation into the developed mathematical model. The conceptual physical and numerical model of the process has been developed which can be used in order to control the storage behaviour and the feasibility of the technology.

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