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Review

# Storage of hydrogenous gas mixture in geological formations: Self-organisation in presence of chemotaxis

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ABSTRACT

Unlike natural gas, gaseous mixtures of hydrogen stored in underground reservoirs undergo active chemical transformations under the influence of methanogenic microorganisms inhabiting in geological strata. These reactions lead to the reduction of concentrations of hydrogen and carbon dioxide and increase the methane concentration. This chemical activity coupled with bacterial dynamics and gas/water transport through porous medium causes the phenomena of self-organization, such as the occurrence of autowaves, whose dynamics is characterized by a multiplicity of scenarios and the bifurcation between them. The foundations of this theory were published in Ref. [9]. In the present paper, we extend the qualitative theory of self-organization in underground hydrogen storage for more complicated cases, which take into account the two-phase flow in porous medium and include chemotaxis, which is one of the main types of bacterial movement. The analysis of scenarios is based on the model of two-phase compositional flow coupled to the population dynamics. We show analytically and numerically that the chemotaxis retains the effects of self-organization, but leads to the appearance of non-periodicity in the structure of spatial oscillations.



## Contents



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## Introduction

Among all renewable energy sources hydrogen ($H_2$) and fuel cells are considered as the energy of the future [1,8,12], due to the low greenhouse-gas emissions related to it and due to the progressively increasing efficiency of fuel cells application to vehicles. The global energy cycle related to $H_2$ includes [8]: the renewable energy generation from windmills and solar cells, the conversion of the excess power to hydrogen through electrolysis, the hydrogen storage to regulate the difference between intermittent production and permanent gas consumption, the hydrogen extraction and the use in fuel cells to produce electricity. In such a scheme, storing $H_2$ comes down to storing electricity. Along with this, $H_2$ may also be produced from fossil fuels, for instance by coal gasification, and used further as fuel. The use of the clean energy in the form of $H_2$ is frequently more attractive economically than the classical use of coal, and not only ecologically.

In the case of producing large amounts of hydrogen, the most efficient and the most inexpensive method of storing is to inject it in geological formations like aquifers, depleted gas reservoirs, or salt caverns [8,14,17]. Several underground storages of hydrogen or town gas (the mixture of $H_2$, CO and $CH_4$) exist in the world, for instance, Teeside in the UK, Kiel in Germany, Lobodice in Czech Republic, Beynes in France and storages in Texas and Russia. The more detailed review is given in Ref. [8]. Frequently the produced hydrogeneous gas is the mixture of hydrogen with other chemical components, as $CO_2$ and other.

In the first papers devoted to hydrogen underground storage [2,4,6], the process was considered as to be similar to the classical storage of methane. However later it was shown in Refs. [3,8,9,13] that the behaviour of hydrogen in natural rocks is very different from that of the natural gas, as $H_2$ is chemically active in presence of anaerobic bacteria, which initiate the following chemical reaction between the injected $H_2$ and $CO_2$:

$$4H_2 + CO_2 = CH_4 + 2H_2O\,, \quad \text{or} \quad 3H_2 + CO = CH_4 + H_2O, \tag{1}$$

The in-situ and laboratory observations really have revealed the increase of $CH_4$ in the stored gas contents and the decrease of $H_2$ and $CO_2$. Along with this, other effects have been observed, as the creation of spatial alternation of the areas saturated preferably by hydrogen or methane [13]. The explanation to these effects have been done in Ref. [9] where it was shown that the coupling between $H_2$ and $CO_2$ transport in the reservoir and bio-chemical reactions leads to the appearance of auto-waves equivalent to the observed spatial alternations.

In contrast to previous papers [9,10] where the fluid was considered as a single-phase gas with residual immobile water, and only the diffusion was retained as the mechanism of bacterial motion, we introduce the following new elements in the present paper:

- The flow is two-phase; water is mobile;
- The additional form of bacterial colony: the neuston, which is a thin film living at the interface between water and gas; a stable neuston exists essentially in stagnant zones, where it cannot be destroyed.
- The chemotaxis as the second important form of bacterial motion. The chemotaxis is the mechanism of bacterial motion to the direction of nutrients.

The first version of the developed mathematical model was presented in Ref. [15]. Therein we published the differential equations of multicomponent two-phase transport with bio-chemical reactions coupled with the equations of bacterial dynamics. However the analysis of the impact of the chemotaxis was not performed. This analysis is the main objective of the present paper.

## Generalized model of the process

### *Model of population dynamics*

Let us assume that a mixture of $H_2$ and $CO_2$ with domination of hydrogen is injected in an aquifer, which contains water, gas and an initial population of methanogenic bacteria. Bacteria can live only in water, then all the chemical reactions occur only in water or at the interface between water and gas.

We introduce the chemotaxis, which is the mechanism of bacterial motion to the location of nutrients (to the