

Accepted Manuscript

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PII: S0883-2927(16)30486-3

DOI: [10.1016/j.apgeochem.2017.04.012](https://doi.org/10.1016/j.apgeochem.2017.04.012)

Reference: AG 3869

To appear in: *Applied Geochemistry*

Received Date: 15 November 2016

Revised Date: 28 April 2017

Accepted Date: 28 April 2017

Please cite this article as: Cristancho, D., Akkutlu, I.Y., Wang, Y., Criscenti, L.J., Shale gas storage in kerogen nanopores with surface heterogeneities, *Applied Geochemistry* (2017), doi: 10.1016/j.apgeochem.2017.04.012.

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Shale Gas Storage in Kerogen Nanopores with Surface Heterogeneities

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Keywords: Shale Gas, Kerogen, Surface Area, Organic Nanopores, Molecular Simulation

Abstract

Recent advances in drilling and well stimulation technologies have led to rapid development of shale formations as an important natural gas resource. However a comprehensive understanding of the source rock geochemistry is currently needed in order to identify key factors in resource shale hydrocarbon assessment and production forecasting. Previous works indicated that significant amount of methane is stored in kerogen in adsorbed state. Adsorption is controlled by surface area and surface properties of the kerogen nanopore walls. In this paper using molecular simulations we investigate the influence of surface chemistry and heterogeneity on methane storage in model kerogen pores. The results show excess amount of methane due to nanopore confinement effect, which is found to be most pronounced under the subsurface conditions when the reservoir pore pressure is in the range of 1,000-5,000 psi. Among the investigated surface heterogeneities, nitrogen-doped graphene surfaces are the most influential on methane storage. Doping affects strongly the Langmuir parameters related to the adsorption capacity. These results indicate that kerogen maturation and the associated changes in its composition has the potential to impact gas storage in resource shale formations. The work gives new insights into the potential impact of the surface chemistry on natural gas storage in kerogen and emphasizes the significance of source rock geochemistry.

1. Introduction

By far, kerogen is the most abundant organic material on earth (Durand 1980) and it may be transformed through three major geological phases: diagenesis, catagenesis and metagenesis. As the consequence of these phases, kerogen pore network is formed and further developed to its present form.

During diagenesis, kerogen loses large amounts of its oxygen due to oxidation reactions taking place and the associated CO₂ and H₂O liberation. In this stage the transformation of kerogen is also driven by cracking reactions whose kinetics are temperature- and pore pressure-dependent. Liquid hydrocarbons are generated mainly during catagenesis, when the hydrogen and carbon atoms are lost from the kerogen structure. Consequently, according to Van Krevelen diagram, H/C atomic ratio of the residual kerogen decreases. During the catagenesis stage further loss of carbon oxides continues and, hence, O/C atomic ratio also continues to decrease. It is the late catagenesis and metagenesis phases when methane is liberated from the kerogen structure together with some non-hydrocarbon gases such as CO₂, N₂ and H₂S. Lorant and Behar (Lorant and Behar 2002) showed these gases are ejected from the solid kerogen surfaces using open and close system pyrolysis. More specifically, they form due to de-methylation of methyl-aromatics and due to cleavage of aromatic-ether bridges (Behar et al. 2000). In addition, cleavage of carbon and hydrogen bonds and auto-hydrogenation yield methane and carbon oxides.

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