

Characterization of the porous medium diffusivity by use of isotopic tracing. Application to gas darceo-diffusive transport in sedimentary basins

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The importance of diffusion phenomenon in losses of gases in sedimentary basins leads to study the quantification of the diffusive flow which requires the knowledge of the diffusivity value. This geometric parameter is defined as the ratio of a gas diffusion coefficient in the water filling the porous medium to the same gas diffusion coefficient in pure water; it is at first sight linked to the tortuosity of the porous medium. However, this parameter is generally not measured and its variations with the different characteristics of the medium are poorly known. As a consequence, some authors often disagreed on estimations of residence time of gas in a structure (Krooss *et al.*, 1992; Nelson *et al.*, 1992).

Therefore, the aim of this study is to improve the understanding of diffusivity and its variations by measuring directly its value for some porous media of various porosity, for different diffusing gas species and by determining isotopic fractionations. It has already been shown that isotopic ratios of hydrocarbons are affected in some ways by transport (like diffusion through water). A previous study carried out in this laboratory, pointed out that hydrocarbon diffusion in shale may entail some isotopic fractionation (Pernaton *et al.*, 1996). This study, hence, will allow us to examine how far isotopic fractionations, when they occur, depend on the medium and its geometry.

The results of our work will allow us to model the diffusivity as a function of the porous medium parameters.

To carry out the experiments, we used a diffusion cell (fig.1) made of two gas reservoirs separated by a water-saturated porous medium. The gas reservoirs were respectively filled with different gases (hydrocarbons, nitrogen) and were kept at the same pressure in order to avoid any darcean flow. The porous medium used was a water-saturated, reconstituted sandstone, of measured thickness and porosity. Gas samples were collected during the course of the experiment to monitor the progress of the diffusion

process. Those samples were analysed by a Toepler line, gas chromatography and mass spectrometry (GC-C-IRMS) in order to get chemical proportions and isotopic ratios for carbon ($\delta^{13}\text{C}$) and hydrogen (δD).

From gas analysis, we could plot two kinds of diagrams: migrated gas production curve which represents the diffused gas amount versus time and migrated gas isotopic fractionation curve which represents the difference between the isotopic ratios of migrated and initial gases versus time.

Then we used a numerical model to determine diffusivity values. This model simulates the transport of gas molecules in the cell (Schneider *et al.*, 1995) : each part of the cell is described by a physical law (Fick's law and a solubilisation law at the gas-water interfaces). Model results are adjusted on experimental ones. The model adjusting parameter is called the migration rate. In its expression each parameter is known except diffusivity. Then from the value of the migration rate given by the model, we can deduce the diffusivity value.

So far, three experiments have been carried out at different porosities (30 to 45%), in order to study compaction influence on diffusivity. The diffusing gaseous species studied for the two first experiments were methane and nitrogen as counter-gas. In the third experiment, as we wanted to study the influence of the diffusing gas species, we used a mixture of three hydrocarbons: methane, ethane and propane, the counter-gas used was still nitrogen. Experimental conditions were the same for all the experiments: gas pressure at 25 bars in both chambers of the cell, and ambient temperature. The porous medium used was also the same in the three experiments: a Fontainebleau sand of 100 μm compacted, and saturated with water to form a reconstituted sandstone. Comparisons of our results in sandstones with those of a previous work using reconstituted and natural shales (Pernaton *et al.*, 1996) allowed us to study the influence of the medium lithology.

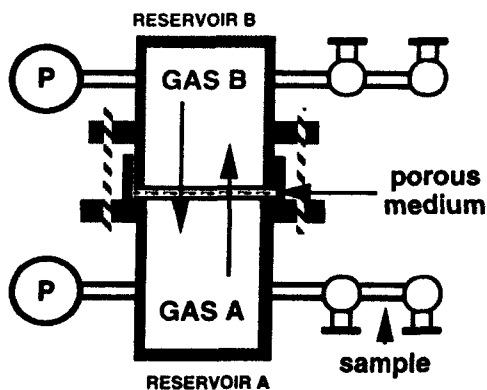


FIG. 1. The cell diffusion.

This study gives us the following conclusions: firstly, the diagram representing diffusivity versus porosity shows that diffusivity is independent of porosity in the studied range even though its overall trend is to decrease with compaction (fig. 2). On this diagram were added points, taken from the literature (Chen, 1973; Krooss, 1988), relating to more compacted natural media. Secondly, diffusivity depends on the medium lithology: in sandstone, it is higher than in shale (with a factor between four and five). Thirdly, diffusivity may also depend on the diffusing gas species; this result is new because diffusivity was only considered from now on as a geometric parameter of the porous medium. Fourthly, as in shale, we can observe an isotopic fractionation in sandstone, though in the latter this isotopic fractionation is lower than in the former. Then, we may infer from this result that isotopic fractionation could also depend on the nature of the porous

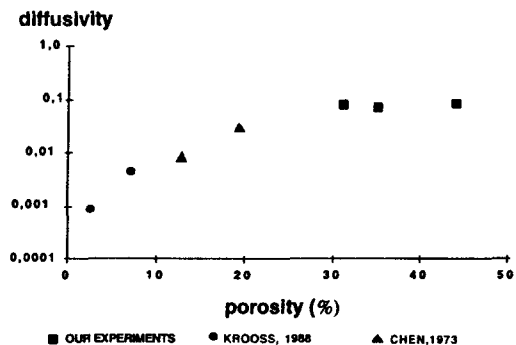


FIG. 2. Diffusivity-porosity diagram for water-saturated sandstones.

medium. At last, the fact that differences between diffusing molecules may influence diffusivity and that isotopic fractionation depends on the porous medium, leads us to think that diffusion is not the only phenomenon we observed: some surface phenomena such as adsorption may also occur.

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