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Determination of Knudsen diffusion of gas flow in artificial porous media

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Abstract -The possibility of calculating the diffusivity coefficient for some materials was studied and what are the main factors that affect the diffusivity calculation and how to deal with it. Some materials were selected to calculate the diffusivity, such as sandstone. Dolomite. Limestone. Where the calculation was made based on the physical properties of porosity, permeability and other properties that directly affect the diffusion process in the presence and absence of samples. The diffusivity was calculated for all samples and the results were as follows: ($4.82225E-08 \text{ M}^2 / \text{S}$) This is the product of the total ethanol diffusivity that happen in air with the sandstone sample in Arnold cell ($6.97705E-08 \text{ M}^2 / \text{S}$) this is the product of the effective Knudsen diffusivity that happen only in the sandstone sample. ($4.38122E-08 \text{ M}^2 / \text{S}$) This is the product of the total ethanol diffusivity that happen in air with the limestone sample in Arnold cell. ($6.09006E-08 \text{ M}^2 / \text{S}$) this is the product of the effective Knudsen diffusivity that happen only in the limestone sample.

Key words: diffusivity coefficient, Knudsen diffusion, sandstone. Dolomite. Limestone

I. Introduction

A key concept in many scientific and engineering domains is gas diffusion in porous media. In fact, gas transport through porous materials is crucial for a variety of applications, such as modeling membranes for gas separation and natural gas evolution in reservoir formation [1]. Numerous experimental, numerical, and theoretical studies on the diffusion parameters of gas transport across porous surfaces have been conducted recently in a variety of sectors, including petroleum, chemical, and environmental engineering, among others. Diffusion models are frequently presented using empirically fitted constants, and these models seldom ever include extensive structural information for porous media. al. used a three-dimensional, off-lattice, diffusion-limited cluster to cluster aggregation approach to assess the diffusivity of a gas via fractal media. However, computational calculations of the effective diffusion coefficients for three-dimensional Gas diffusion in porous media is a process caused by the random thermal motion of gas molecules [2]. Knudsen diffusion and normal diffusion are typically the two types of gas diffusion seen in pore spaces. The sort of diffusion that is most likely to predominate is determined by the mean free route of the gas molecule and the relative length

of the pore diameter. The primary mechanism will be the collision of the gas molecules with the pore walls, or Knudsen diffusion, if the mean free path of the gas molecule is significantly bigger than the diameter of the pore through which it flows. The diffusion process will be controlled by gas molecule collisions (normal diffusion) if the mean free path is significantly lower than the pore diameter, but Knudsen diffusion will be minimal. Nevertheless, these two diffusion methods can both contribute to the gas in many actual circumstances. Whereas there is barely any Knudsen diffusion. These two diffusion processes, however, can often work together to contribute to the gas diffusion in pore spaces in real-world circumstances. [3]

When the scale length of a system is equal to or less than the mean free path of the particles involved, Knudsen diffusion, a kind of diffusion, takes place. This may be seen, for instance, in a lengthy pore with a small diameter (2–50 nm), where molecules repeatedly run into the pore wall .

Think about how gas molecules diffuse through tiny capillary holes. Gas molecules collide with the pore walls more frequently than with one another if the pore width is less than the mean free route of the diffusing gas molecules and the gas density is low. The Knudsen flow or Knudsen diffusion is the name of this phenomenon.

The Knudsen number is a good measure of the relative importance of Knudsen diffusion. A Knudsen number much greater than one indicates Knudsen diffusion is important. In practice, Knudsen diffusion applies only to gases because the mean free path for molecules in the liquid state is very small, typically near the diameter of the molecule itself.

The diffusivity for Knudsen diffusion is obtained from the self-diffusion coefficient derived from the kinetic theory of gases:[2]

1.1. Diffusivity

The calculation of effective diffusion coefficients for porous media, given the geometrical characteristics, has received renewed attention because of the recent progress in both computer hardware and porous medium imaging techniques (such as X-ray tomography, NMR imaging). It is not unrealistic to try to compute directly transport properties given the description of a real porous system. Still, three-dimensional computations are very demanding in terms of

computer resources, therefore calculations are restricted to unit cells involving a few particles. These unit cells must be representative, in some sense, of the real structure to give effective diffusion coefficients close to the actual measurements. This question was extensively based on experiments and numerical solutions of the closure problem associated with the determination of the effective diffusion coefficients. However, computations were carried out on two-dimensional structures while the experimental data were obtained from truly three-dimensional porous systems (packing of spheres, disks, and mica particles). The two-dimensional using parallelepipeds in planar configurations. The purpose of this work is to present more realistic results based on the systems To begin with, three types of cubic packing of spheres were used to study the process of diffusion in isotropic systems. To model the anisotropic systems studied by Kim et al., three dimensional unit cells containing randomly oriented disks were studied. The closure problem associated with the calculation of the effective diffusion coefficient is solved numerically using a finite difference scheme. Several calculations are performed for different unit cells representing various types of unconsolidated isotropic and anisotropic porous media.[2]

1.2. Closure Problem and the Diffusion Coefficient

Diffusion process taking place in a porous system made up of two-phases as it is illustrated in Figure 2.1, the α -phase is the solid phase, and the β -phase is the fluid phase filling the pores. The characteristic pore length-scales associated with the two-phases are denoted l_1 and l_2 , while the porous system itself has a characteristic length-scale, L , such that $L \gg l_1, l_2$.

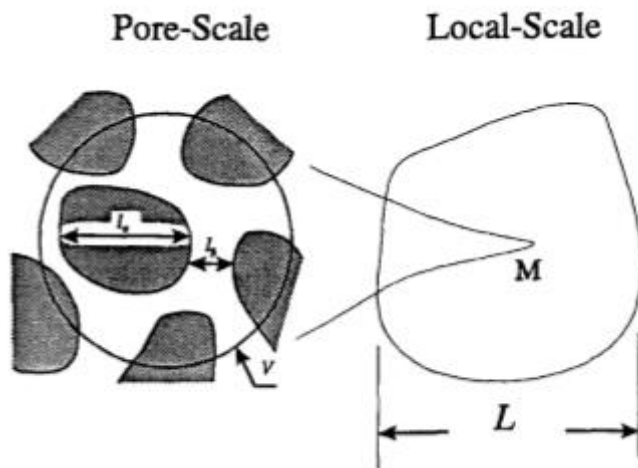


Figure 1. Pore-scale and local-scale of a porous medium

For the problem under consideration here, this method requires a large number of voxels per unit volume to be accurate enough. Since the resulting linear system is large and the associated matrix is not sparse, calculations proved to be very cumbersome in the case of three-dimensional noncontiguous.[2]

1.3. Gas Diffusion Coefficient in Highly Permeable Porous Media.

Gas diffusion is a fundamental transport process in porous media. However, to date, there is no method for experimentally estimating the gas diffusion coefficient of coarse porous media. A modified method is proposed here and was validated against sand media. The method was tested using particle sizes ranging between 1 and 4 cm: $\sim 10^{-3} \text{ cm}^2 < \text{intrinsic permeability} < \sim 10^{-2} \text{ cm}^2$. Laboratory experiments were conducted in large horizontal columns filled with different homogeneous spherical particles inside a climate-controlled laboratory under isothermal ($\pm < 0.2^\circ\text{C}$) and windless conditions. Diffusion coefficients were similar for particles sizes of 1 and 2 cm, which indicates that pure diffusion governs gas transport. Above 2 cm, diffusion coefficients were higher, suggesting an enhanced gas transport, most likely initiated by small, local advective components. Because the enhanced transport can be modeled by the diffusion equation using a larger diffusion coefficient, it is associated with the enhanced diffusion concept.

Accounting for gas fluxes within a porous media (PM), such as the upper vadose zone, requires partitioning the flux contribution from two transport mechanisms—diffusion and advection. For purely Fickian diffusion, the key parameter comprising PM properties is the diffusion coefficient. Published models that are used to predict the gas diffusion coefficient (DP) were developed from studies performed in PM of low and moderate intrinsic permeability, k , hereafter called permeability (sand-sized particles and below, $\sim 10^{-5} \text{ cm}^2$). The lack of experimental tests performed in coarse PM leaves open the question if pure diffusion can be assumed to dominate transport in natural and artificial systems of high k even when ambient conditions that would drive advection are stable, within sensor resolution (i.e., no wind, thermal gradient, or pressure gradient conditions).[7]

II. Materials and Methods

Experiments using packed PM in large horizontal columns were conducted inside a climate-controlled laboratory (CCL) under isothermal and no-wind conditions. Each column was filled with identically sized spherical particles, testing a total of four particle sizes (1–4 cm). An additional column was used for validating the method, using packed sand and comparing the results to sand diffusion coefficients. For a tracer, CO_2 -enriched air (0.2%).[3]

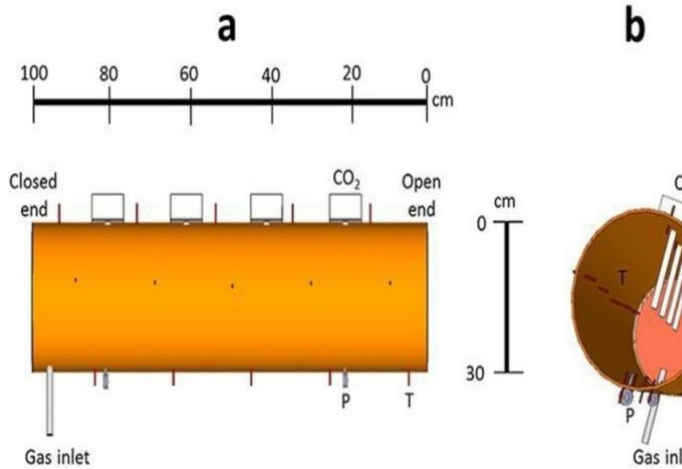


Figure 2. Schematic of the column showing location of gas inlet and sensors: (a) two dimensional and (b) three dimensional, showing differential pressure transducer (P) and temperature (T) and CO₂ sensors.

For each of the two differential pressure sensors, the first pin was connected to a small-diameter tube that was inserted ~2 cm into the porous medium and the second pin was open to the ambient atmosphere of the laboratory.

2.1. Knudsen diffusion.

Knudsen diffusion occurs either at very low gas pressures or at normal pressures in very narrow pores. The criterion for Knudsen diffusion is that the mean free path for the void gas phase molecule_ molecule scattering

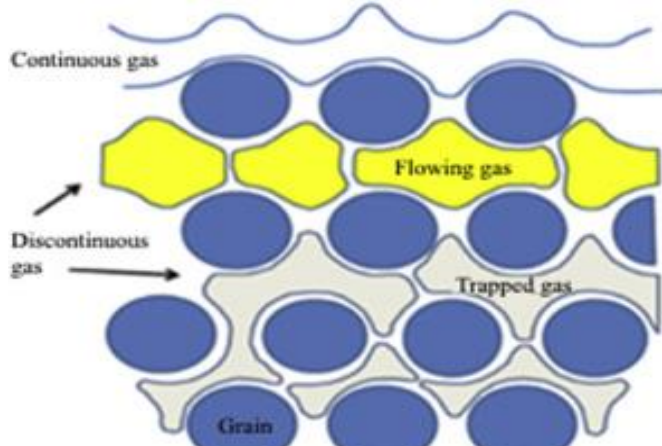


Figure 3. Show diffusivity in porous. [3]

Let us now turn our attention to the opposite condition, namely $\lambda \geq \lambda_p$. In terms of the so-called Knudsen number, $K_n \gg 1$, this condition is $Kn \gg 1$. In this case the probability of a molecule-molecule collision is negligible compared to that of a molecule-wall collision. Those molecules rebounded or re-emitted by the wall do not collide with other molecules and the particle can thus only influence those molecules that collide with it—the molecules that have not collided with the

wall cannot "learn" about its existence from the rebounded molecules. This kind of molecule-wall interaction will be called a *Knudsen collision* and the corresponding phenomenon will be called the *Knudsen regime*. [4,5]

If in a multicomponent system with Knudsen flux there is a concentration gradient for a given component, there will be a net flux of the molecules of that component. As in a system devoid of walls, the species segregate, but collide with only one other species—the particles. A given species cannot "learn" about the existence of the other species since there are no molecule-molecule collisions. Then, because pressure affects only the concentration of the molecular species, the Knudsen flux of a given component will be directly proportional to the pressure of the system. [5]

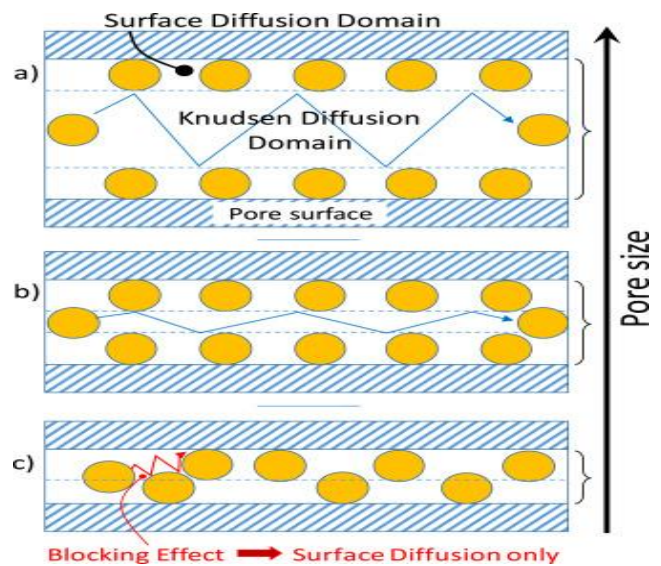


Figure 4. method Knudsen flux between the grain. [6]

When the concentration of a component varies from place to place within a fluid mixture, the random motion of molecules tends to transport that component from areas of high to low concentration, so that over time the system becomes homogenized with respect to the concentration of each mobile component. The transport of matter in this way is known as molecular diffusion. Pure diffusion in the absence of advection and chemical interaction, ie, the transport of an ideal tracer. [4, 5]

III. Experimental work:

Experiment Steps:

1. Prepared and cleaned the sample of impurities with gasoline.
2. Dry the samples before weighing them.
3. Weigh the samples dry.
4. Place the samples in chamber 2.
5. Fill chamber 3 with water.
6. Open the valve 1.
7. Close the valve 2.
8. Turn on the vacuum pump.
9. Leave the valve 2 closed for a period of time until air is drawn from the samples.

10. After the air is drawn, open the valve 2.
11. Water will be drawn from chamber 3 to chamber 2.
12. After the samples are covered with water, close the valve2.
13. Allow the device to work until samples are saturated with water.
14. Weigh the samples as saturated.
15. We repeated this experiment 3 times and took the average for it.

Porosity and Density Calculations.

In this experiment, we calculated the porosity and density using the saturation method of the four samples, which will be mentioned later.[7.8].

We calculated porosity using the saturation method using a law:

$$\varphi = \frac{Vp}{VB} \quad VP = \frac{Wwet - Wdry}{\rho Water}$$

Calculated the density using a law:

$$\rho sample = \frac{Wdry}{VB}$$

The samples are calculated from the following types:

1. Sandston.
 2. Dolomite.
 3. Limestone.
 4. Mix dolomite and limestone.
- Four natural core samples are studied which are including Sandston, dolomite, limestone mix dolomite and limestone as shown in figure 3.1 below. These samples were taken after permission from Dr.Amer Alshareef.

Table. 1 Geological information and well description

GENERAL DATA	Lithology
1. A.WELL NAME:H34-NC115 1. B.OPERATOR:REPSOL OIL OPERATIONS 1. C.TYPE OF WELL: OIL PRODUCER 1. D.LOCATION	<p>SHALE: (grey, light grey, brownish to light brownish, poor to moderately indurations, blocky to subblocky, fissile to subfissile, silty, pyretic, micaceous.</p> <p>SANDSTONE: white, off white, light grey, light brown, transparent to translucent, medium hard to hard, fine to medium grained, subangular to subrounded, moderately sorted, siliceous cement, kaolinitic and pyretic in par. Core Depth:4738ft</p>
1. A.WELL NAME: E62H-103E 1. B.OPERATOR:REPSOL OIL OPERATIONS	<p>CLAYSTONE: reddish to reddish brown, brown, soft to poor indurations, sticky, subblocky, sandy, slightly dolomitic.</p>

1. C.TYPE OF WELL: OIL PRODUCER 1. D.LOCATION	<p>DOLOMITE: grey to light grey, light brown, medium hard, cryptocrystalline, microcrystalline in parts, slightly to moderately argillaceous, Core Depth: 6030ft</p> <p>LINESTONE: grey, dark grey, beige, off white, soft to medium hard, cryptocrystalline to microcrystalline, moderately to highly argillaceous. Core Depth: 6300ft</p>
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Samples.

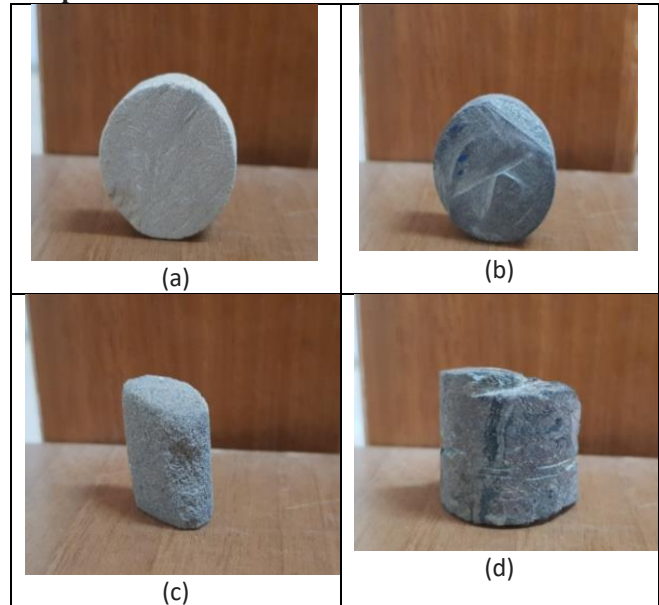


Figure 5: (a) Sandstone, (b) Dolomite (c) Limestone(d) Mix dolomite and limestone

Saturation device components.

The vacuum saturation method as shown in figure 3.2, is in fact one of the very basic methods of obtaining the pore volume of a rock sample.

One of the advantages is the fact that pore volumes of multiple samples can be determined in one step. The method uses a large enough vacuum flask or a beaker, filled with liquid, normally water, in which dry rock samples are placed. Subsequently, as soon as the evacuation of the vacuum flask is initiated, air bubbles are seen in the saturating liquid as it replaces air from the pore spaces of the rock samples. The disappearance of

The air bubbles gives an indication the saturation is complete and at this point the evacuation is terminated, and porosity is calculated.

1. vacuum pump .
2. Standard chamber 1.
3. Chamber 2 for samples.
4. Chamber 3 contains water.
5. A valve 1 and a valve 2

$$CBm = \frac{CB2 - CB1}{LN \cdot \left(\frac{CB2}{CB1}\right)}$$

Mass transfer and diffusion device components.

1. Electric control panel.
2. Transparent tank.
3. Micrometer.
4. Arnold cell.
5. Air pump.

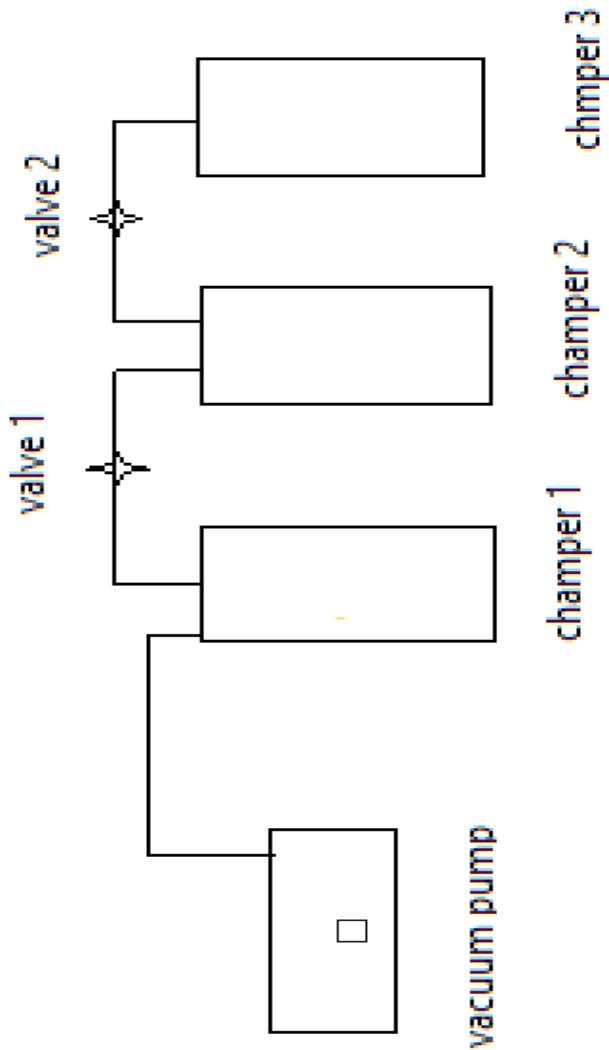


Figure 7 . mass and transfer diffusion experiment.



Figure.6. the saturation device used in the experiment illustrates

Diffusivity Calculations

We calculated the diffusivity using the mass transfer and diffusion experiment in Arnold cell it once without a sample and twice in two samples and This was done at a temperature of 60 ° C and with a concentration ethanol of 75% Using the following laws.[7].

These laws are used to calculate total diffusivity;

$$DAB = \frac{\rho l}{2 \cdot Mwt. slope \cdot CA} * \left(\frac{CBm}{CT}\right)$$

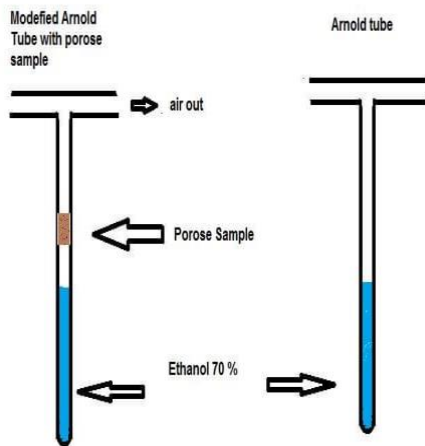


Figure.8.arnold cell.

Calculation the Diffusion without sample.

Calculated the diffusivity of ethanol in the air at the previously described specifications.

Steps of the experiment.

1. Fill the Plexiglas tank with pure water.
2. Fill the capillary glass tube (Arnold cell) with ethanol.
3. Turn on the heating resistance until the temperature reaches 60 C.
4. Place the capillary tube in a Plexiglas tank.
5. Turn on the air pump.
6. Take reading over time.

Calculation the Diffusion in sample.

In this case, the diffusivity of the sandstone sample and the lime-stone sample and this was calculated after placing the samples inside the capillary tube (Arnold cell) and with the same liquid used (ethanol) and at the same conditions.



Figure 9b..Sample (1) sandstone



Figure 9a.Sample (3) lime-stone



Figure 9c. Sample (3) after cutting



Figure 9d..sample (1) afterlime-stonecutting sand stone



Figure9e.The sample (3) is inside (Arnold cell)

1. Fill the Plexiglas tank with pure water.
2. Fill the capillary glass tube with test liquid.
3. We made a system to insert the sample into the capillary glass tube (Arnold cell).

4. Turn on the heating resistance until the temperature reaches 60 C.
5. Place the capillary tube in a Plexiglas tank.
6. Turn on the air pump.
7. Take reading over time.

IV. Results and Discussion

Table .2 Samples Measurements and Calculations.

Diffusion calculation without sample.

In this process, we calculated the diffusivity of ethanol in the

		sample1	sample2	sample3	sample4
	W dry(g)				
1		7.7615	10.4346	5.9469	14.2145
2		7.7454	10.419	5.9188	14.05
3		7.7409	10.4273	5.9099	14.149
	AVG Wdry	7.74927	10.427	5.9252	14.1378
	W wet(g)				
1		8.147	10.4451	6.028	14.3281
2		8.278	10.44	6.0529	14.2977
3		8.4681	10.559	6.1459	14.537
	AVG Wwet	8.2977	10.4814	6.0756	14.3876
	VP(cm ³)	0.54843	0.0544	0.1504	0.24977
	Vbulk(cm ³)	3.2	4	1.8	5.2
	porosity%	17.1385	1.36	8.35556	4.80321
	density(g/cm³)	2.42165	2.60674	3.29178	2.71881

air at the previously described specifications.

Table.3.This table contains the results diffusion without sample.

Time(min)	Time(ks)	ΔL(mm)	t/ΔL
28	1.68	0.562667	2.985782
84	5.04	1.253333	4.021277
168	10.08	1.703333	5.917808

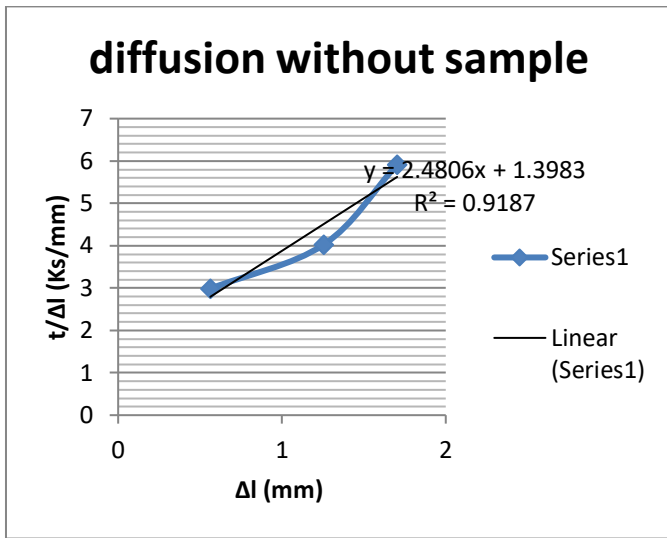


Figure 10. diffusion without sample

Diffusion calculation in sample.

In this case, the diffusivity of the sandstone sample and the lime-stone sample and this was calculated after placing the samples inside the capillary tube (Arnold cell) and with the same liquid used (ethanol) and at the same conditions.

Sand stone sample calculations.

Table.4. This table contains the results diffusion with sample Sandston.

Time(min)	Time(ks)	ΔL(mm)	t/ΔL
10	0.6	0.196	16.66667
20	1.8	0.374667	11.11111
30	3.6	0.548	8.333333
40	6	0.766667	6.666667
50	9	0.955333	5.555556
60	12.6	1.133	4.761905
70	16.8	1.355	4.166667

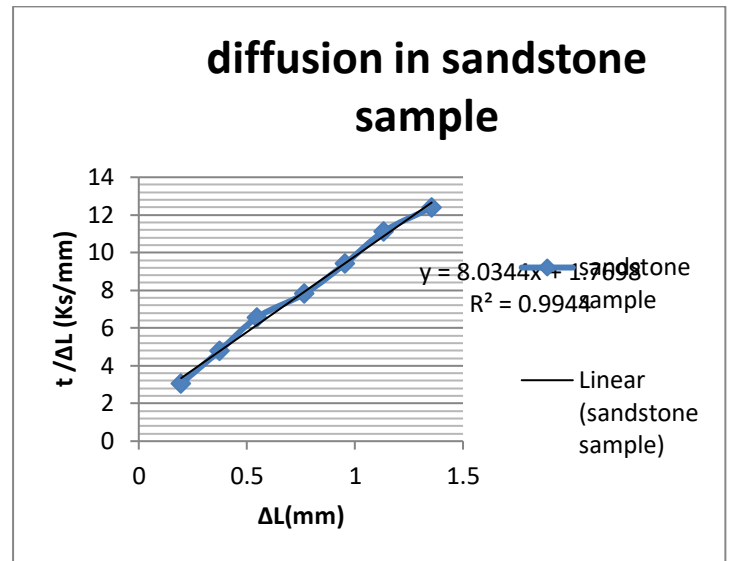


Figure 11. diffusion in sand stone sample

sample

DAB = 4.82225E-08 M² / S

DAk,e sandstone = 6.97705E-08 M² / S

(4.82225E-08 M² / S) This is the product of the total ethanol diffusivity that happen in air with the sandstone sample in Arnold cell

(6.97705E-08 M² / S) this is the product of the effective Knudsen diffusivity that happen only in the sandstone sample.

Lime-stone sample calculations.

Table.5. This table contains the results diffusion with sample lime-stone.

Time(min)	Time(ks)	ΔL(mm)	t/ΔL
10	0.6	0.19	3.157895
20	1.8	0.36	5
30	3.6	0.518	6.949807
40	6	0.743	8.07537
50	9	0.92	9.782609
60	12.6	1.07	11.7757
70	16.8	1.33	12.63158

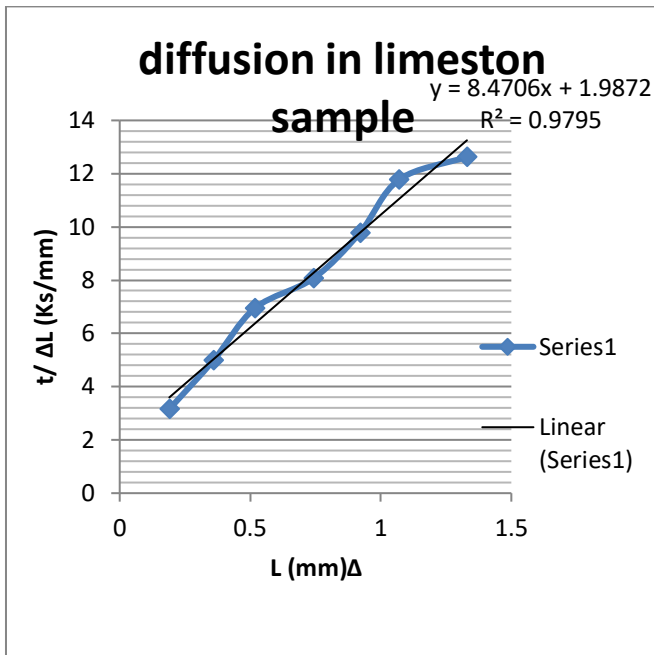


Figure 12. Diffusion in lime-stone sample.

DAB = 4.38122E-08 M²/S

DAK limestone = 6.09006E-08 M² / S

(4.38122E-08M²/S) This is the product of the total ethanol diffusivity that happen in air with the limestone sample in Arnold cell.

(6.09006E-08 M² / S) this is the product of the effective Knudsen diffusivity that happen only in the limestone sample

V. Conclusion and Recommendation:

In this search calculations by selecting a group of rocks and calculated their properties (density and porosity) and from that selected two types of rocks and calculated the Knudsen diffusion without a sample and the Knudsen diffusion in the two samples using methanol fluid.

Developing this experiment by using highly volatile hydrocarbon fluids such as gasoline and using other types of rocks with different properties.

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