

Framework for Estimating Vapor Transport Parameters through Intact Geomembranes by the Dusty Gas Model (DGM)

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ABSTRACT

A framework based on the Dusty Gas Model (DGM) for estimating gas/vapor transport parameters through geomembranes has been suggested to address the limitation in the commonly used method for the analysis of Water Vapor Transmission (WVT) tests which is based on Fick's first law of diffusion. Currently, it is assumed that gas or vapor transport through geomembranes occurs via molecular diffusion and thus Fick's law of diffusion is suitable for carrying out the analysis. The main limitation, however, stems from the fact that geomembranes have extremely low permeability, varying from 10^{-12} to 10^{-15} cm/sec, that renders other gas transport flux mechanisms such as Knudsen diffusion significant and in many cases is more important than molecular diffusion. This indicates that Knudsen diffusion dominates the gas transport mechanisms through geomembranes and that the adequacy of Fick's law to model gas transport through geomembranes is questioned. The DGM, on the other hand, incorporates Knudsen diffusion, as well as: viscous flow, bulk diffusion (non-equimolar and molecular diffusion), surface diffusion and thermal diffusion. Analysis of reported measurements of Water Vapor Transmission (WVT) test using the suggested framework resulted in estimates of the Knudsen diffusion coefficient, rather than the effective molecular diffusion coefficient that is commonly obtained when applying Fick's law. The obtained Knudsen diffusion coefficients are then used to estimate the average pore radius of the geomembranes. For the sake of contrasting the two methods, the reported WVT results were also analyzed based on the commonly used method, and the results were used to estimate the porosity of the geomembranes. The estimated value of the porosity is unreasonable and supports the argument presented in this study which states that Knudsen diffusion is the dominant mechanism for transport through geomembranes and not molecular diffusion. Finally, a new set of geomembrane parameters, permeance and permeability is introduced. The newly defined parameters are theoretically rigorous since they are based on the dominant mechanism and assume no equivalence of isobaric and non-isobaric systems.

Keywords: Framework, Geomembrane, Dusty Gas Model (DGM), Knudsen Diffusion, Equivalent Permeability.

1. INTRODUCTION

The use of geomembranes in environmental applications has been rapidly growing in the United

States and Germany due primarily to the governmental regulations that started in the early 1980s (Koerner, 2005). The same trend is expected to spread to other parts of the world soon. Geomembranes are used primarily for lining and covers of liquid and/or solid storage facilities. This includes all types of landfills, reservoirs, canals and other containment facilities. For a lined landfill, for

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example, the final cover system is mainly designed to control infiltration and methane migration and usually consists, from top to bottom, of: a 0.15 m topsoil for growing vegetation; 0.75 m compacted soil; a drainage layer consisting of soil and/or a geosynthetic drainage product; a low-permeability hydraulic barrier; a geomembrane; a gas-venting layer; and a graded landfill surface.

A geomembrane is defined (ASTM 4439) as a very low permeability synthetic membrane liner or barrier used with any geotechnical engineering related material so as to control fluid (or gas) migration in a human-made project, structure or system. Geomembranes are usually made of relatively thin continuous polymeric sheets and in some cases are made from the impregnation of geotextiles with asphalt or elastomer spray or as a multilayered bitumen geocomposite. Currently (Koerner, 2005), the most widely used geomembranes include: High Density Polyethylene (HDPE); Linear Low Density Polyethylene (LLDPE); and Polyvinyl Chloride (PVC). Somewhat less widely used geomembranes are: Flexible Polypropylene, non reinforced (FPP) and reinforced (FPP-R); Chlorosulfonated Polyethylene, Reinforced (CSPE-R); Ethylene Propylene Diene Terpolymer: Nonreinforced (EPDM) and Reinforced (EPDM-R); and Ethylene Interpolymer Alloy, Reinforced (EIA-R).

Geomembrane characteristics, such as water vapor transmission, permeance and diffusion (permeability), are experimentally estimated using the Water Vapor Transmission (WVT) test. In this test, a geomembrane specimen is sealed over an aluminum cup with water (i.e., 100% relative humidity) in it and a relatively low relative humidity outside; a weight loss over time is monitored. The required time varies from 3 to 40 days. Water vapor transmission, permeance and diffusion (permeability) are then calculated. The same test can be performed with different solvent vapors replacing the water in the cup.

Although the primary function of geomembranes is containment of, or barrier to, liquids or vapors, polymeric geomembranes are not absolutely impermeable, but they are impermeable when compared to geotextiles and soils (even to clay soils). Typical

values of geomembranes' permeability as measured by water-vapor- transmission tests (Haxo et al., 1984; Koerner, 2005) are in the range of 1×10^{-12} to 1×10^{-15} m/s which is six orders of magnitude lower than the typical clay liner. These low values have a strong indication on the mechanisms of transport through intact geomembranes as will be explained below.

Gas transport through an intact geomembrane (no holes or punctures) has been studied by assuming molecular diffusion mechanism and applying Fick's law of diffusion (Starck and Choi, 2005). For geomembranes with holes or openings, gas has been assumed to flow via viscous flux (Darcy's law) and molecular diffusion. On the other hand, whenever the coefficient of permeability of the porous media is less than 10^{-10} cm², which is the case here, Knudsen diffusion is important and may dominate the transport and can not be ignored. Of the three commonly used gas transport models; namely Fick's law of diffusion, Stefan Maxwell equations and the Dusty Gas Models (DGM), only the DGM incorporates Knudsen diffusion and thus it must be used in the analysis of gas flow through geomembranes. However, the use of the DGM requires a new set of transport parameters including among others the Knudsen diffusion coefficient.

This paper presents a new framework for the analysis of gas transport through intact geomembranes using the DGM and provides a method for analyzing the WVT test data in order to estimate the Knudsen diffusion coefficient.

2. GAS TRANSPORT MECHANISMS IN POROUS MEDIA

A thorough analysis of gas transport through porous media (geomembranes herein) is a complex problem that requires multicomponent analysis and the consideration of various gas transport flux mechanisms, such as, viscous flow, free-molecule or Knudsen flow, continuum or ordinary diffusion (molecular and nonequimolar fluxes), surface diffusion and thermal flow. A brief discussion of these mechanisms is given below and a

detailed discussion of these mechanisms is available in various references (Jackson, 1970; Cunningham and Williams, 1980; Mason and Malinauskas, 1983; Thorstenson and Pollock, 1989; Thorstenson and Pollock-b, 1989; Abu-El-Sha’r, 1993; Abu-El-Sha’r and Abriola, 1997; Warrick, 2002; Abu-El-Sha’r, 2006).

Viscous flow occurs when a pressure gradient is applied on the system. The damping effects due to the high rate of interaction (i.e., collisions) among gas molecules compared to the interaction between gas molecules and the boundaries of the system causes a constant viscous flux. This flux depends on the coefficient of viscosity, which for gases is independent of pressure at constant temperature. Moreover, a mixture of gases behaves the same as a single gas, because such bulk flow has no tendency to cause a mixture to separate into its components (no segregation of species). The molar viscous flux N^v in porous media is given by:

$$N^v = -\frac{P}{RT} \frac{k}{\mu} \nabla P \quad (1)$$

where k is the intrinsic permeability, μ is the dynamic viscosity, ∇P is the pressure gradient, P is the pressure, R is the gas constant, and T is the temperature.

Knudsen flux dominates when the mean free path of the moving molecules; the distance traveled between two consecutive molecular collisions, is extremely large compared to the pore radius. Then, the probability of a molecule- molecule collision is negligible compared to that of a molecule-wall collision. Since the rebounded molecules don’t collide with other molecules, the molecules that have not collided with the wall are not affected by the existence of the rebounded molecules, so for a system in the Knudsen regime, there are as many independent fluxes present as there are species. In a multicomponent gaseous system, there is a concentration gradient for each component and thus a net Knudsen flux of each component as well. The net Knudsen flux of gas i can be calculated as (Cunningham and Williams, 1980):

$$N_i^K = -D_i^K \frac{dc_i}{dz} \quad (2)$$

where N_i^K is the Knudsen molar flux of component i , c_i is the molar concentration of component i , and D_i^K is the Knudsen diffusivity, given as:

$$D_i^K = Q_p \left(\frac{RT}{M_i} \right)^{1/2} \quad (3)$$

where: Q_p is the obstruction factor for Knudsen diffusivity, T is the temperature, R is the ideal gas constant and M_i is the molecular weight of gas i .

For a porous medium with a single pore size, Q_p is given by (Abu-El-Sha’r, 1993):

$$Q_p = 1.064 \bar{r} \quad (4)$$

where \bar{r} is the average pore radius.

A continuum or ordinary diffusion stems from the experimental evidence that when two gases with different molecular weights are brought into contact through a porous medium, two different fluxes develop. The first is a segregative diffusive flux for each component and the second is a non-segregative diffusive flux as a result of the difference in concentration (non-equipolarity) of species and is called nonequipolar flux. Thus, in a system with walls (porous media), the total diffusive flux of a given species consists of two components; diffusive flux (segregative) and non-equipolar flux (non-segregative).

For a binary gaseous system of non-equipolar gases i and j , the total molar diffusive flux for gas i is given by (Cunningham and Williams, 1980):

$$N_i^D = J_{iM} + x_i \sum_{j=1}^v N_j^D \quad (5)$$

where N_i^D , N_j^D are the total molar diffusive fluxes of species i , and j , respectively; J_{iM} is the molar diffusive flux of species i ; x_i is the molar fraction of species i ; and ν is the number of gas components.

The total molar diffusive flux of component i is given by (Cunningham and Williams, 1980):

$$N_i^T = N_i^D + X_i N^\nu \quad (6)$$

where N_i^T is the total diffusive flux of component i , N_i^D is the molar diffusive flux of component i and X_i is the mole fraction of component i .

Surface flux occurs when gas molecules are adsorbed on specific sites at the surface of the particles of the porous medium (geomembrane). Due to the continuous movement (vibrations) of the adsorbed molecules, each molecule transfers by hopping to other adsorption sites a number of times before it returns to the gaseous phase. Surface diffusion is usually modeled by employing Fick's law of diffusion where the concentration gradients refer to the surface concentration gradients and all the complexities of the porous medium geometry, surface structure and adsorption equilibrium are lumped into the surface diffusion coefficient. The Fickian model is useful only at low surface coverage (Mason and Malinuskas, 1983). In this paper, the number of molecules adsorbed to the geomembrane adsorption sites is assumed equal to the number of molecules leaving the adsorption sites (steady-state condition). Thus, net surface flux will not affect the total gaseous flux and is neglected.

Thermal diffusion is the relative motion of the gaseous species caused mainly by temperature gradient. However, it has been theoretically demonstrated that, for temperature differences less than 200 °C, the thermal diffusive flux is insignificant and may be ignored (Al-Ananbeh, 2003).

3. GAS TRANSPORT MODELS

Fick's first law of diffusion has been used to study gas or vapor transport through geomembranes (Haxo et al.,

1984; Koerner, 2005; Starck and Choi, 2005). However, due to the small permeability of the geomembranes, Knudsen diffusion is dominant and must be considered. The only gas transport model commonly used and that considers Knudsen diffusion is the Dusty Gas Model (DGM). These models are briefly reviewed in the following subsections.

Fick's First Law of Diffusion

Fick's first law of diffusion is generally used to predict molecular diffusion of a binary gas system into porous media under isobaric conditions. For multicomponent systems, a binary analysis is performed with all gas components, other than the gas for which the prediction is needed, assumed as one component. For non-isobaric conditions, the diffusive and the viscous fluxes are usually assumed to be independent of each other and thus the total flux is obtained by adding these two fluxes. The diffusive flux given by Fick's law of diffusion when written in the molar form is given by (Bird et al., 1960; Jaynes and Rogowski, 1983):

$$(N_i^D)_F = -D_{ij}^e C \nabla x_i \quad (7)$$

where $(N_i^D)_F$ is the Fickian molar diffusive flux of component i ; D_{ij}^e is the effective binary diffusion coefficient of gas components i and j (given by equation (8) below); C is the total molar gas concentration; and ∇x_i is the molar fraction gradient for species i . The effective binary diffusion coefficient of components i and j can be written as:

$$D_{ij}^e = Q_m D_{ij} \quad (8)$$

where Q_m is the diffusibility factor, also known as the obstruction factor and is only a function of the porous medium; and D_{ij} is the free binary diffusion coefficient of gases i and j .

Equation (7) has the same form as Fick's law for a

liquid provided that the total concentration of the gas system is constant, *i.e.* for an isobaric system. Despite the fact that Fick's law of diffusion is widely used in soil science and environmental engineering to study gaseous diffusion, it must be remembered that, when applied to gases, it is primarily an empirical relation borrowed from studies involving solutes and shown to agree well with some observations of gaseous diffusional processes (Kirkham and Powers, 1972).

Dusty Gas Model (DGM)

The Dusty Gas Model (DGM) combines the different gas transport mechanisms in a rigorous manner and can be applied to multicomponent systems, both isobaric and non-isobaric. The physical picture behind the model is that of a dusty gas, in which the dust particles constitute the porous medium. The basic working assumptions of the DGM are: 1) the suspended particles are spherical, can be treated as a component of the gas mixture, are motionless and uniformly distributed, are very much larger and heavier than the gas molecules and are acted upon by an external force that keeps them at rest even though a pressure gradient may exist in the system and 2) no external forces act on the gas molecules.

The constitutive forms of the DGM equations for *v* gas components for isobaric isothermal conditions are given by (Cunningham and Williams, 1980):

$$\sum_{j=1, j \neq i}^n \frac{x_i (N_j^D)_{DGM} - x_j (N_i^D)_{DGM}}{D_{ij}^e} - \frac{(N_i^D)_{DGM}}{D_i^k} = \frac{1}{RT} \nabla p_i + n \sum_{j=1}^v x_i x_j \alpha_{ij} \nabla \ln T \tag{9}$$

where x_i and x_j are the molar fraction of gas component *i* and *j*, respectively; $(N_i^D)_{DGM}$ and $(N_j^D)_{DGM}$ are the total molar diffusive fluxes of

components *i* and *j* given by the DGM; D_i^k and D_j^k are the Knudsen diffusion coefficient of species *i* and *j*, respectively; p_i is the component pressure of gas *i*; *R* is the gas constant; *T* is the temperature of the system; *n* is the gas and particle density; α_{ij} is the generalized thermal diffusivity; and *v* is the number of gas components.

The summation term on the left-hand side of equation (9) is the momentum lost through molecule-molecule collisions with species other than *i* (but not the particles). The second term on the left hand-side of equation (9) is the momentum lost by species *i* through molecule-particle collisions. The first term on the right-hand side represents the concentration gradient contribution to diffusion of species *i*, and the second term represents the thermal gradient effect on diffusion, which may be neglected for isothermal systems.

Analysis of Gas transport through Geomembranes

The analysis of any gaseous system is based on material balanced equations. For binary steady state conditions with no reactions between flowing gases and the geomembrane material and where surface diffusion and thermal diffusion are ignored, the mass balance equation for a gas *i* is given by:

$$\nabla \cdot N_i^T \tag{10}$$

where, N_i^T is the total molar flux of gas component *i* which is either the diffusing gas or air.

Analysis of Water Vapor Transmission (WVT) Test Results

In this test, a geomembrane specimen is sealed over an aluminum cup with water (*i.e.*, 100% relative humidity) in it and a relatively low relative humidity outside as shown in Figure (1). The weight loss of the water in the can is monitored over a time period that varies from 3 to 40 days under isobaric and isothermal conditions. Water vapor transmission, permeance and diffusion (permeability) are then calculated as explained in subsequent sections. The currently used analysis is

based on Fick's first law of diffusion which assumes molecular diffusion of water vapor into air (assumed as one component herein) and that no other gas transport mechanisms are considered despite the fact that Knudsen flux dominates in extremely low permeability porous media. The following subsections present the commonly used analytical procedure for parameter estimation of geomembranes from the WVT test results as well as a contrast of this procedure to the analysis based on the mentioned gas transport models.

According to Fick's Law

Based on Equations (7) and (8) given above, the total mass flux of component *i* under isobaric conditions may be written as:

$$F_i^D = -Q_m D_{ij} (M_i / R T) \nabla p_i \tag{11}$$

Where F_i^D is the Fickian mass diffusive flux of component *i* (g/(m²-day); M_i is the gram molecular weigh of gas *i* (g/mole); R is the gas constant (62363.8 cm³-mmHg/mole-K^o), T is the absolute temperature (°K), ∇p_i is the pressure gradient across the geomembrane (mmHg/cm). Note that p_i is estimated from the saturation vapor pressure at test temperature (mm Hg) multiplied by the relative humidity at that temperature.

For a geomembrane with thickness *t*, ∇p_i may be approximated by $\Delta p_i/t$, and Equation (11) may be rewritten as:

$$F_i^D = -Q_m D_{ij} (M_i / R T) \Delta p_i/t \tag{12}$$

According to DGM

For a binary gaseous system, the dusty gas model (equation (9)) yields the following equation for the total molar diffusive flux of component *i*, (Cunningham and Williams, 1980):

$$N_i^D = N_i^T = - \left\{ \frac{1}{D_i^k} + \frac{1-(M_i/M_j)^{1/2}x_i}{D_{ij}^e} \right\}^{-1/2} C \nabla x_i \tag{13}$$

The terms between brackets to the right hand side of equation (13) represent the momentum exchange between the diffusing gas molecules with the particles of the geomembranes (Knudsen diffusion) and with each other (molecular diffusion), respectively. The relative importance of these two mechanisms can be estimated by comparing the values of the Knudsen effective diffusion coefficient and the effective molecular diffusion coefficient. As can be seen from equation (13), the importance of the transport mechanism is inversely proportional to its transport coefficient value. For geomembranes, the values of the Knudsen diffusion coefficients are much smaller that the corresponding values of the binary molecular diffusion coefficients. Thus Knudsen diffusion dominates, and equation (13) can be rewritten as after neglecting the molecular diffusion terms:

$$N_i^T = - \left\{ \frac{1}{D_i^k} \right\}^{-1/2} C \nabla x_i \tag{14}$$

On a mass basis, Equation (14) may be written as follows:

$$J_i^T = - \left\{ \frac{1}{D_i^k} \right\}^{-1/2} \frac{M_i}{RT} \nabla p_i \tag{15}$$

where, J_i^T is the Knudsen diffusion flux expressed on a mass basis.

For a geomembrane with thickness *t*, ∇p_i may be approximated by $\Delta p_i/t$, and Equation (15) becomes:

$$J_i^T = - \left\{ \frac{1}{D_i^k} \right\}^{-1/2} \frac{M_i}{RT} \frac{\Delta p_i}{t} \tag{16}$$

Rearranging Equation (16), the Knudsen diffusion coefficient may be estimated as:

$$D_i^k = \left(N_i^T \frac{RT}{M_i} \frac{t}{\Delta p_i} \right)^2 \quad (17)$$

Incorporating equations (3) and (4) into equation (17) and rearranging yields:

$$\bar{r} \cong 0.94 \left(\frac{N_i^T t}{\Delta p_i} \right)^2 \left(\frac{RT}{M_i} \right)^{1.5} \quad (18)$$

Equation (18) provides a practical means for estimating the average pore radius of geomembranes from the WVT tests.

Common Analysis of Water Vapor Transmission Tests Results

The results obtained from conducting the WVT are usually used to estimate the WVT, permeance and the (diffusion) permeability of the tested geomembranes. The WVT is estimated by dividing the mass change of the water in the cup used for the test by the time interval representing the test duration and the area of the cup. In other words, WVT is the mass flux of water vapor through the geomembrane indicated by the obtained units of (g/ m²-day).

The permeance on the other hand is given by:

$$\text{Permeance} = WVT / \Delta p_i = WVT / S(R_2 - R_1) \quad (19)$$

The vapor diffusion permeability following Fickian diffusion, not Darcian permeability, is defined as:

$$\text{(Diffusion) permeability} = \text{permeance} \times \text{geomembrane thickness} \quad (20)$$

Comparing equations (12) and (20) yields:

$$\text{permeance} = (D_{ij}^e / t) (M_i / RT) \quad (21)$$

Rearranging and solving equation (21) for D_{ij}^e yields:

$$D_{ij}^e = (\text{permeance} \cdot t \cdot R \cdot T) / (M_i) \quad (22)$$

On the other hand, comparing and contrasting equations (17) and (18) with equation (19) yields the following two equations, respectively:

$$D_i^k = \left(WVT \frac{RT}{M_i} \frac{t}{\Delta p_i} \right)^2 \quad (23)$$

And the average pore radius of the tested geomembranes may be estimated from the following:

$$\bar{r} \cong 0.94 \left(\frac{WVT t}{\Delta p_i} \right)^2 \left(\frac{RT}{M_i} \right)^{1.5} \quad (24)$$

Calculations and Results

The Water Vapor Transmission (WVT) has been measured (Haxo et al. 1984) for several common geomembranes with different thicknesses under a constant temperature of 25 °C and a relative humidity gradient of 45%. The values of WVT are reproduced in Table (1). For the test temperature (25 °C) and a pressure of 1 atmosphere, the saturated water vapor pressure is 23.78 mm Hg, and the free diffusion coefficient of water vapor into air is experimentally determined as 0.256 cm²/sec (Arora, 1989). Using equation (22), the effective binary diffusion coefficient is calculated (Table (2)). These values are then used to estimate the obstruction factor values based on equation (8) which in turn are used to estimate the porosity of the tested geomembrane based on Abu-El-Sha'r – Abriola diffusibility model given by (Warrick, 2002):

$$Q_m = 0.435 n \quad (25)$$

Where: n is the porosity.

Estimated values of the porosity for the different membranes are given in Table (2). As can be seen, the values of porosity for several tests were above 1 which is unreasonable. This is explained by the fact that molecular diffusion is not the dominant flux mechanism and that other mechanisms should be considered.

Using the DGM, the Knudsen diffusion coefficient is calculated based on equation (23) and the average pore radius of the geomembrane is estimated based on equation (24). Results are given in Table (3).

4. CONCLUSIONS

This study reveals that gaseous transport through geomembranes is dominated by Knudsen diffusion contrary to the commonly assumed and used molecular diffusion because of the extremely low permeability of these geomembranes. This finding dictated a fundamental change in the modeling process from the conventionally used Fick's law to the DGM which incorporates the

Knudsen diffusion as well as other flux mechanisms. The suggested framework has been successfully applied to WVT test results and estimates were obtained for the Knudsen diffusion coefficients and the average pore radius of these geomembranes. Besides, when applying the conventional method for analyzing the same experimental results, some values of the calculated porosities exceeded 1, which signals an error in analysis. This conforms well with the fact that molecular diffusion is not the dominant mechanism for gas transport in geomembranes. Finally, a new set of geomembrane parameters has been rigorously defined and presented for future use.

Table (1): Summary of Water Vapor Transmission Tests Results.

Geomembrane Polymer	PVC			CSPE-R	EPDM-R		HDPE	
Thickness (cm)	0.028	0.052	0.075	0.089	0.051	0.123	0.08	0.244
WVT (g/m ² -day)	4.4	2.9	1.8	0.44	0.27	0.31	0.017	0.006
Perm-cm	1.2x10 ⁻²	1.4x10 ⁻²	1.3x10 ⁻³	0.84x10 ⁻²	0.13x10 ⁻²	0.37x10 ⁻²	0.013x10 ⁻²	0.014x10 ⁻²

Source: Haxo et al., 1984.

Table (2): Results of Analysis of WVT Tests Using a Fick's law Based Approach.

Geomembrane Polymer	Thickness (cm)	Perm-cm	D _{ij} ^e (cm ² /sec)	Q _m	Porosity, n
PVC	0.028	0.012	0.1435	0.560	1.288
	0.052	0.014	0.1674	0.654	1.503
	0.075	0.013	0.1554	0.607	1.396
CSPE-R	0.089	0.0084	0.1004	0.392	0.902
EPDM-R	0.051	0.0013	0.0155	0.061	0.140
	0.123	0.0037	0.0442	0.173	0.397
HDPE	0.08	0.00013	0.0016	0.006	0.014
	0.244	0.00014	0.0017	0.007	0.015

Table (3): Results of Analysis of WVT Tests By the Suggested DGM Framework.

Geomembrane Polymer	Thickness (cm)	Perm-cm	D_i^K cm ² /sec	\bar{r} cm
PVC	0.028	0.012	2.06E-02	1.90E-05
	0.052	0.014	2.80E-02	2.59E-05
	0.075	0.013	2.42E-02	2.23E-05
CSPE-R	0.089	0.0084	1.01E-02	9.33E-06
EPDM-R	0.051	0.0013	2.42E-04	2.23E-07
	0.123	0.0037	1.96E-03	1.81E-06
HDPE	0.08	0.00013	2.42E-06	2.23E-09
	0.244	0.00014	2.80E-06	2.59E-09

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