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Carbonate rocks: Matrix permeability estimation

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Abstract:

Carbonate rocks store half of the world’s proven oil reserves. Genesis and post-depositional diagenetic processes define the porous network topology and the matrix permeability. This study compiles a database of porosity, specific surface, mercury porosimetry and permeability values extracted from published sources, and complements the database through a focused experimental study. Specific surface and porosity combine to estimate the pore size $D_{sur}$. Permeability vs. $D_{sur}$ data cluster along a single trend with a slope of 2 in a log-log scale in agreement with the Kozeny-Carman model. Discordant data points correspond to samples with dual porosity or broad pore size distributions with long tails, where flow channels along larger interconnected pores. Indeed, the detailed analysis of all the porosimetry data in the database shows that permeability correlates best with the pore size $D_{80}$, i.e. the 80th percentile in pore size distributions. Once again, the best fit is a power function in terms of $(D_{80})^2$, analogous to Kozeny-Carman. The prediction uncertainty using $D_{80}$ is one order of magnitude, and has the same degree of uncertainty as more complex models and analyses. This observation suggests an irreducible uncertainty of one order of magnitude in permeability estimation from index properties such as porosity, mercury porosimetry and specific surface, probably due to specimen preparation effects, inherent physical differences in permeation vs. invasion, and difficulties in data interpretation. These estimates of permeability are most valuable when specimens are limited to small sizes, such as cuttings.
INTRODUCTION

The world energy demand has steadily increased during the last century, with an additional 30% increase in demand predicted by the year 2040 (BP, 2018). Fossil fuels satisfy 81% of the current global energy consumption (IEA, 2018). While its share of the total consumption will decrease to 75% by mid-century, the actual consumption of non-renewable sources will continue to increase. Hydrocarbons comprise half of the global energy mix (IEA, 2015).

The rock porosity and permeability determine the quality of hydrocarbon reservoirs: porosity implies storage capacity while permeability is needed for flow and recovery (Dullien, 1992; Tiab and Donaldson, 2012). Natural and induced fractures control the overall flow in carbonate reservoirs (Golf-Racht, 1996; Gale et al., 2004; Ortega et al., 2010), however, the rate at which stored hydrocarbons exit the matrix into fractures depends on the matrix permeability.

Permeability relates the average flow velocity to the driving total energy gradient. The flow velocity for Newtonian fluids in laminar flow through cylindrical tubes is proportional to the square of the tube diameter (Hagen-Poiseuille equation). The Kozeny-Carman model considers the porous medium as a set of parallel cylindrical tubes and uses the Hagen-Poiseuille equation to compute the effective flow velocity (Kozeny, 1927; Carman, 1937). Then, the resulting permeability $k$ [m$^2$] is proportional to the square of the pore diameter $D$ [m] and the porosity of the porous medium $\phi$ [m$^3$/m$^3$]

$$k = \frac{\phi}{32\tau^2} D^2$$

(1)
where $\tau [m/m]$ is the tortuosity. However, pore size is not constant, and the largest interconnected pores are responsible for most of the flow. This is confirmed by network model studies of flow behavior at the pore scale which show the coupling between pore size, spatial variability, and connectivity on flow patterns (Jang et al., 2011). Semi-empirical factors added to the Kozeny-Carman equation attempt to take these processes into consideration, often through a generic tortuosity factor (Equation 1).

Carbonate rocks store half of the world’s proven oil reserves (EIA, 2015). Genesis and post-depositional diagenetic processes define the pore structure in carbonate rocks (Moore and Wade, 2013). The intraparticle porosity, high friability, and chemical reactivity of carbonate sediments affect their evolution during burial (Croizet et al., 2013; Moore and Wade, 2013), and leads to features such as dual and occluded porosity (Figure 1 - see also Poursoltani and Gibling, 2011; Saner and Sahin, 1999). Experimental data show that the pore size in carbonates varies by more than 6 orders of magnitude (Nelson, 2009) while the permeability varies by $\sim$10 orders of magnitude (Nelson, 1994), in overall agreement with the power-2 dependency anticipated by Equation 1.

The purpose of this study is to enhance the understanding of carbonate permeability using a physics-inspired yet data-driven approach. The following section describes the database compiled for this study.

DATABASE - CHARACTERIZATION AND POTENTIAL PITFALLS

This study compiles a database of permeability values extracted from published sources for carbonate rocks in the United States, Russia, the Middle East and Europe (Data sources:
Brooks and Purcell, 1952; Chilingarian et al., 1990; Lucia, 1995; Mortensen et al., 1998; Lindsay et al., 2006; Fabricius et al., 2007; Clerke, 2009; Alam et al., 2011; and Vincent et al., 2011). The 286 entries include mostly binary data in terms of permeability, rock formation, porosity, specific surface, and/or pore size distribution (See dataset as AAPG Datashare). Permeability and specific surface data span several orders of magnitude. While data sources use similar measurement methods (gas adsorption for specific surface and helium expansion for porosity), differences in test protocols, devices and data analyses add variability to the dataset.

Only thirteen entries have all three, porosity, pore size distribution and specific surface (Paris Basin - Vincent et al., 2011). This research conducts a focused experimental study designed to extend this dataset using eleven commercially available carbonates cores (Kocurek Industries), some with multi-modal pore size distributions (refer to Table 1). Test details and potential pitfalls follow.

Porosity. Weight change upon liquid saturation provides the accessible porosity (API, 1998). The saturation procedure involved five steps: (1) vacuum, (2) CO\textsubscript{2} injection cycles to replace the residual air inside the specimen, (3) vacuum, (4) injection of deaired-deionized water into the vessel, and (5) several vacuum-pressure cycles. The specimen dry weight $W_{\text{dry}}$ [g] and saturated weight $W_{\text{sat}}$ [g] combine to determine the porosity $\phi$ using the mineral specific gravity $G_s$

$$\phi = \frac{(W_{\text{sat}} - W_{\text{dry}})G_s}{W_{\text{dry}} + (W_{\text{sat}} - W_{\text{dry}})G_s} \quad (2)$$

Measured porosities vary between $\phi=0.11$ to $\phi=0.53$ (Table 1).
**Specific Surface.** Several liquid and gas-based methods were tested to determine specific surface. Whereas liquid adsorption measurements rely on gravimetric changes after lengthy equilibration times (Cerato and Lutenegger, 2002), gas adsorption with krypton emerged as the most adequate characterization procedure given the relatively low specific surface area of carbonates (Micromeritics ASAP 2420 - Beebe et al., 1945). The measured specific surface areas range from $S_s=0.5$ to $1.3\ \text{m}^2/\text{g}$. This coincides with reported values for carbonate rocks (Chilingarian et al., 1990; Vincent et al., 2011).

Tests were conducted with carbonates crushed to two different sizes. Results summarized in Table 1 show that the measured specific surface depends on crushed particle size, even though the external surface area is negligible in all cases (e.g., the external surface is $0.03\ \text{m}^2/\text{g}$ for $70\ \mu\text{m}$ grains). This suggests that sample crushing gives access to occluded porosity and creates new gas pathways (Note: calibration tests showed equipment variability of less than 4%).

**Pore Size Distribution.** Mercury intrusion porosimetry MIP measures the volume of mercury that invades the specimen as a function of pressure (Giesche, 2006). Mercury invades along percolating paths, and occluded porosity remains untested. The Young-Laplace equation relates the measured pressure to pore throat size (León y León, 1998) while injected volumes correspond to pore bodies. Consequently, large pores may be assigned to small pore throats, i.e., the ink-bottle effect (Diamond, 2000; Moro and Böhni, 2002).

Pore size distributions obtained from mercury injection porosimetry tests are usually presented in terms of pressure $P\ [\text{Pa}]$ and the logarithm of the differential intrusion $g(D)$ for a given saturation $S\ [\text{m}^3/\text{m}^3]$.
\[ g(D) = \frac{dS}{d(lnP)} = P \frac{dS}{dP} \]  

(3)

This definition emphasizes dual porosity systems and amplifies the contribution of large pores. However, the “physical” pore size density function \( f(D) \) relates pressure to capillarity in terms of the surface tension \( \gamma \) [N/m] and the contact angle \( \theta \) [rad] (Lenormand, 2003).

\[ f(D) = \frac{P^2}{2 \gamma \cos(\theta)} \frac{dS}{dP} = \frac{P}{2 \gamma \cos(\theta)} g(D) = \frac{2}{D} g(D) \]  

(4)

Therefore, the commonly used distribution \( g(D) \) has a pore size-dependent amplification of the true pore size distribution \( g(D) = D \cdot f(D)/2 \). Figure 2 shows the pore size distributions \( g(D) \) and \( f(D) \) obtained for the eleven specimens tested in this study. The estimated mean pore sizes computed from \( g(D) \) are significantly smaller than the mean pore sizes obtained from \( f(D) \). These results highlight profound differences in the potential interpretation of these data.

Permeability. The permeability of all eleven specimens was measured using a gas permeameter (MetaRock SSK-300). Ends remained unpolished to avoid fines clogging near the inlet face of the cylindrical specimens (diameter: 25 mm [1 in]; length: 50 mm [2 in]), and limited pressure gradients prevented non-linear effects. Values of \( \text{N}_2 \)-permeability measured at different mean pressures were used to correct for Klinkenberg’s effect. Table 1 includes the measured permeability values.
DATA ANALYSES

Porosity and Carbonate Classification. Empirical models for carbonate permeability focus on porosity as a predictive parameter (Jennings and Lucia, 2003; Babadagli and Al-Salmi, 2004; Lucia, 2007). The inherent limitation in empirical models that are based exclusively on porosity is highlighted by the contrast between the very narrow range in porosity (say, 0.1<ϕ<0.6) versus the 10 orders of magnitude in the permeability range (Nelson, 1994).

Additional information can be included, such as carbonate classification in terms of textural features and particle size, as these features provide information about genesis and ensuing pore topology (Pemberton and Gingras, 2005; Boggs, 2009; Uddin et al., 2017). Dunham’s classification distinguishes (Dunham, 1962): (a) coarse-grained dominant carbonates (grainstones: dolostatones and large crystalline grainstones) (b) carbonates with a coarse-grained structure but with fines in pores (packstones), and (c) fines-dominant carbonates (wackestone, mudstone and fine crystalline limestones and dolostones). Then, the empirical permeability-porosity power-model (Lucia, 1995),

\[ k = a \phi^b \]  \hspace{1cm} (5)

relates the \(a\)-factor and \(b\)-exponent to carbonate rock type. Figure 3A superimposes two datasets for non-vuggy carbonate reservoirs in the United States and in the Middle East (Lucia, 1995; Lindsay et al., 2006). Lucia’s model highlights the importance of rock type and the role of fines or “mud” on pore networks and permeability, yet predictions have more than one order of magnitude in uncertainty, in part due to potential differences in pore structure (see thin section based analyses in Weber et al., 2009).
Porosity, Pore Size Distribution and Pore Structure. Other models relate permeability to pore size distributions inferred from mercury porosimetry (Swanson, 1981; Katz and Thompson, 1986; Glover et al., 2006; Rezaee et al., 2006; Zhiye and Qinhong, 2013). Data analyses reveal that the largest modal element or “porositon” $M_{max}$ [µm] determines the matrix permeability [md] in carbonates with multimodal pore size distributions (Figure 3B – Clerke et al., 2008; Clerke, 2009)

$$\log(k) = -1.54 + 1.2 \log(M_{max}) + 7.3 \phi \quad (6)$$

More detailed analyses assume an internal pore structure such as fractal, consider critical path analysis, and/or apply percolation theory (Charlaix et al., 1987; Friedman and Seaton, 1998; Hunt and Gee, 2002; Buiting and Clerke, 2013; Daigle, 2016). For example, Buiting and Clerke (2013) match mercury porosimetry data with one or more Thomeer hyperbolas and extract three parameters: the maximum invaded volume $\phi^*$, pressure at first invasion $P_d$ [kPa], and pore geometry factor $G$. Through mathematical analysis, these three parameters combine to predict the rock permeability (Assumes tortuosity ~2 and fractal dimension ~1.56 – See resemblance with the earlier empirical models by Swanson [1981] and Thomeer [1983]):

$$k = 24050 \frac{\phi^*}{(P_d)^2} e^{-4.43\sqrt{G}} \quad (7)$$

where permeability is in Darcy. The application of models based on pore size distribution derived from MIP is not straightforward (starting from the interpretation of pore size distribution data discussed above – Equation 4). While authors tend to highlight model predictability, results obtained as part of this study using these models against the dataset show at least one order of magnitude in uncertainty.
Porosity and Specific Surface. Permeability is a measure of the drag that a viscous fluid experiences as it traverses a porous medium. Therefore, the data compilation and the experimental dataset include specific surface and porosity. Data reported in terms of the volumetric specific surface $S_{vol} \text{[m}^2/\text{cm}^3\text{]}$ are converted to the gravimetric specific surface $S_s \text{[m}^2/\text{g}\text{]}$ as:

$$S_s = \frac{1}{(1-\phi) \rho} S_{vol}$$  \hspace{1cm} (8)

where $\rho \text{[g/cm}^3\text{]}$ is the mass density of solids. Figure 4 illustrates permeability versus specific surface on a log-log scale. Data subsets of equal porosity cluster along lines with a slope of -2 in the log-log plot.

The Kozeny-Carman equation highlights the importance of pore size on permeability (Equation 1). The specific surface $S_s$, porosity $\phi$ and mineral mass density $\rho$ combine to estimate the pore size $D_{sur} \text{[m]}$ that corresponds to the measured surface area:

$$D_{sur} = \alpha \left[ \frac{\phi}{(1-\phi) S_s \rho} \right] \hspace{1cm} (9)$$

where the $\alpha$-factor is a function of the fabric and pore topology, as shown in Figure 5. Figure 6 plots permeability values in the database as a function of the pore size estimated with Equation 9 for $\alpha=4$ (i.e., parallel cylindrical tubes – Figure 5). All data points cluster along a single trend with a slope of 2 in a log-log scale in agreement with the Kozeny-Carman model in Equation 1 (see analogous conclusions for a wide range of sediments in Ren and Santamarina, 2018). Most values fall within one order of magnitude of the main trend. The best fit line is
\[ k = 5 \left(D_{\text{sur}}\right)^2 \]  

where \(D_{\text{sur}}\) is in micrometers [\(\mu\text{m}\)] and permeability is in milidarcy [md]. This equation allows us to predict permeability from \(S_s\) and \(\phi\) values determined from small samples such as cuttings when pores are significantly smaller than the cutting size. In carbonates, the size of cuttings ranges from 1-to-10 mm long depending on drilling conditions (Archie, 1952; Dogruoz et al., 2016), therefore cuttings are \(\sim 3\) orders of magnitude larger than pores. However, cuttings impose an inherent bias as they break preferentially along more porous and weaker planes, therefore, predicted permeabilities are lower-bound estimates of the formation permeability.

**DISCUSSION**

*Representative pore size.* Out-of-trend data points in Figure 6 suggest that the pore size \(D_{\text{sur}}\) estimated from porosity and specific surface may not be an accurate predictor of the pore size that controls permeability in all cases. Discordant data points either have clear dual porosity (see the \(g(D)\) representation for Indiana-60, Indiana-70 and Indiana-200 in Figure 2), or they exhibit very broad pore size distributions with long tails (Mount Gambier, Silurian Dolomite and Winterset specimens – Figure 2).

The representative pore size that is most predictive of permeability is explored in Figure 7 where the measured permeability values are plotted against selected pore diameter percentiles from \(f(D)\) signatures (Equation 4 - Note: the representative pore size for permeability is equivalent to the concept of critical pore size in other studies - Arns et al., 2005; Nishima and Yokoyama, 2017). The dataset used for this analysis includes the eleven samples tested in this study plus thirteen carbonate samples from the literature (Vincent et al., 2011). The computed
square error and visual inspection confirm that the pore size between D70 and D85 (i.e., the 70th and 85th percentiles in porosimetry data) provides the most predictive permeability vs. pore size regression [µm] for all specimens (data range: 0.1md < k < 10,000md)

\[ k = 0.2 (D_{80})^{1.75} \approx 0.1 (D_{80})^{2} \]  \hspace{1cm} (11)

While the first equation is the best fit, the second expression has a very similar residual error and it is quadratic on the particle diameter in agreement with the Kozeny-Carman Equation 1. The pore size estimate \( D_{\text{sur}} \) is less relevant to permeability in the discordant data points as flow channels along the larger interconnected pores (i.e., D80 percentile). Further analyses show that permeability estimates using the D80 pore size have the same degree of uncertainty, one order of magnitude, as more complex models that assume fractal pore structures, critical path analysis, and percolation theory (Methods by Charlaix et al., 1987; Buiting and Clerke, 2013; Daigle, 2016).

Equations 10 and 11 indicate that the representative pore size D80 along the most conductive percolating paths is \( D_{80}/D_{\text{sur}} \approx 50 \) larger than the pore size inferred from specific surface \( D_{\text{sur}} \). The dataset confirms the inverse relationship between specific surface and pore size, however the pore size \( D_{\text{sur}} \) computed from specific surface correlates best with the 20-percentile D20 of the pore size distribution \( f(D) \) measured with mercury intrusion.

**Anisotropy.** Porosity, pore size distribution, and specific surface do not provide information about anisotropy. Therefore, all models based on these parameters considered permeability to be isotropic, i.e., a scalar. However, permeability is direction-dependent, i.e., a tensor. Permeability
anisotropy in carbonates originates from inherent sedimentation layering and preferentially aligned features (Dürrast and Siegesmund, 1999; Tipping et al., 2006), biogenic burrows (Pemberton and Gingras, 2005), stress anisotropy (Barton and Quadros, 2014) and ensuing stress-dependent diagenetic processes (Sibson, 1994; Rashid et al., 2015; Toussaint et al., 2018).

**Upscaling.** MIP-based predictions depend on the measurement and interpretation of pressure-volume data obtained on small specimens. The assumption of a fractal pore structure provides a convenient framework for upscaling laboratory measurements, but only within the validity of the fractal system in the rock matrix (Katz and Thompson, 1985; Pape et al., 1999; Costa, 2006). Stratigraphic features and fractures limit the upscaling size.

**Alternatives?** Analyses suggest that inherent limitations in the prediction of carbonate permeability from index properties lead to an uncertainty of at least one order of magnitude. Furthermore, porosity, specific surface and porosimetry are costly measurements. They are most valuable when specimens are limited to small sizes, such as cuttings (Swanson, 1981; Santarelli et al., 1998). However, when cores are available, quick liquid-based measurements of permeability can be less costly; these measurements avoid complex data analysis (as in gas-based measurements) (Wu et al., 1998; Tanikawa and Shimamoto, 2009; Sander et al., 2017), provide the true value of permeability rather than a correlated estimate, and test series can readily assess anisotropy and heterogeneity.
CONCLUSIONS

Matrix flow is important even in fractured systems as hydrocarbons stored in the matrix need to migrate to fractures. Carbonate type and formation history define the matrix permeability.

Permeability data plotted against the pore size estimated using porosity and specific surface cluster along a single trend with a slope of 2 (in log-log scale). This result highlights the underlying physics of permeability as captured in the Kozeny-Carman model.

Out-of-trend data points correspond to carbonates with either multi-modal or broad pore size distributions with long tails. In both cases, flow channels along the larger interconnected pores. Permeability correlates best with the pore size D80 that corresponds to the 80th percentile in the porosimetry data. This conclusion applies to all carbonates in the database, and it leads to a simple and robust permeability estimator.

More detailed analyses assume an internal pore structure and concepts such as critical path analysis. Their implementation is not straightforward; estimates of permeability using these models result in permeability values with a typical one order of magnitude in uncertainty. This variability is similar to that obtained with other simpler estimators.

All analyses suggest an irreducible uncertainty of one order of magnitude in permeability estimation from index properties such as porosity, porosimetry and specific surface. This may reflect specimen preparation effects (e.g., crushing size for specific surface measurements or inadequate saturation in porosity determinations), inherent physical differences (permeation of single-phase fluid vs. invasion of an immiscible fluid in MIP), and difficulties in data interpretation (e.g., porosimetry, gas related corrections in k-measurements).
The estimation of permeability based on specific surface and porosity is most valuable when only cuttings are available. When cores are available, simple and quick liquid-based permeability measurements should be sought: they can be less costly than specific surface, porosity, and mercury porosimetry measurements, avoid the inherent uncertainty in correlation-based estimates, and allow the assessment of anisotropy.

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**Vita**

**J. Carlos Santamarina**

J. Carlos Santamarina (PhD: Purdue University; MSc: University of Maryland; CE: UNC Argentina) explores the foundations of subsurface processes using particle- and pore-scale testing methods combined with high resolution geophysical process monitoring systems and inversion techniques. The combined experimental-numerical framework supports developments in the field of energy geotechnology, with contributions to resource recovery as well as energy and waste geostorage.

**Alejandro Cardona**

Alejandro Cardona (MSc: KAUST; BS: UNAL Colombia) is currently pursuing his doctoral studies in Energy Resources and Petroleum Engineering in King Abdullah University of Science and Technology KAUST. His research focuses on the understanding of flow-related phenomena in porous media with an emphasis on fractured carbonate rocks.
Figure Captions

**Figure 1.** Scanning electron microscope image of an Indiana carbonate sample. The image confirms the presence of ~15 µm pores in agreement with mercury intrusion data. The zoomed-in picture on the right (corresponds to the white square on the left image) illustrates the sub-micron pore topology.

**Figure 2.** Normalized pore size distributions obtained from mercury porosimetry. The black solid lines show the logarithmic differential intrusion $g(D)$, and the black dashed lines correspond to the probability density function $f(D)$.

**Figure 3.** Empirical models for carbonate permeability: (A) Permeability as a function of porosity $\phi$ and carbonate type (Data from Lucia, 1995, and Lindsay et al., 2006; for comparison, the original classification used by Lindsay et al. is mapped onto Lucia’s classification). Color coding identifies rock type; triangles correspond to Lucia’s data and filled circles are Lindsay et al. data. (B) Permeability as a function of the largest porosity size $M_{max}$ measured using mercury porosimetry (after Clerke, 2009); dashed lines correspond to isoporosity values in the model (refer to Equation 6). Data points are colored to reflect the distance between model predictions and measured values in terms of standard deviation $\sigma$.

**Figure 4.** Permeability $k$ versus specific surface $S_s$ for different porosity ranges. The dashed line has a -2 slope in agreement with the Kozeny-Carman equation. The color coding distinguishes
data points according to porosity. Dataset: 286 datapoints. Data sources: Alam et al., 2011; Brooks and Purcell, 1952; Chilingarian et al., 1990; Fabricius et al., 2007; Mortensen et al., 1998; and Vincent et al., 2011.

**Figure 5.** Models to estimate the surface-related pore size $D_{sur}$ for different pore geometries, where $S_s$ [m$^2$/g] is specific surface and $\phi$ is porosity.

**Figure 6.** Measured permeability $k$ versus estimated pore size $D_{sur}$ using a model of parallel cylindrical tubes (Equation 9). Most of the data collapse onto a narrow trend. The dashed line has a 2-slope in agreement with the Kozeny-Carman equation. Data sources: Alam et al., 2011; Brooks and Purcell, 1952; Chilingarian et al., 1990; Fabricius et al., 2007; Mortensen et al., 1998; and Vincent et al., 2011. New experimental data gathered in this study are shown as yellow points.

**Figure 7.** Permeability $k$ versus different pore diameter percentiles. The dashed line has a slope value of 2. Percentiles D70-D85 provide the best regression with lowest square error with respect to the dashed line. Yellow points: this study. Gray points: published data (Vincent et al., 2011)
Indiana Limestone
Formation: Bedford
Age: Mississippian
(A) Scatter plot showing the relationship between permeability ($k$) and porosity ($\phi$) for different types of rocks: grainstones (black triangles), packstones (red circles), and wackestones, mudstones (blue triangles). 

(B) Scatter plot showing the relationship between permeability ($k$) and largest porosity ($M_{\text{max}}$) with error bars indicating standard deviations ($\sigma$) for different porosity levels: $\phi = 0.08$, $\phi = 0.17$, $\phi = 0.26$, and $\phi = 0.35$.
Surface-related pore size $D_{sur}$

\[ D_{sur} = \alpha \left[ \frac{\phi}{(1 - \phi) S} \right] \frac{1}{\rho} \]

\[ D_{sur} = 2 \left[ \frac{\phi}{(1 - \phi) S\rho} \right] \]

\[ D_{sur} = 4 \left[ \frac{\phi}{(1 - \phi) S\rho} \right] \]

\[ D_{sur} = 2 \left[ \frac{\phi}{(1 - \phi) S\rho} \right] \]

Assumptions/Comments

- General form
- $\alpha$ is the structure factor
- Simple cubic packing of cubical particles
- $D \ll L$
- Pores are parallel cylindrical tubes
- Parallel sheets