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Effects of specific surface area and porosity on cube counting fractal dimension, lacunarity, configurational entropy, and permeability of model porous networks: Random packing simulations and NMR micro-imaging study

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SUMMARY

Despite the importance of understanding and quantifying the microstructure of porous networks in diverse geologic settings, the effects of the specific surface area and porosity on the key structural parameters of the networks have not been fully understood. We performed cube-counting fractal dimension (D_{cc}) and lacunarity analyses of 3D porous networks of model sands and configurational entropy analysis of 2D cross sections of model sands using random packing simulations and nuclear magnetic resonance (NMR) micro-imaging. We established relationships among porosity, specific surface area, structural parameters (D_{cc} and lacunarity), and the corresponding macroscopic properties (configurational entropy and permeability). The D_{cc} of the 3D porous networks increases with increasing specific surface area at a constant porosity and with increasing porosity at a constant specific surface area. Predictive relationships correlating D_{cc}, specific surface area, and porosity were also obtained. The lacunarity at the minimum box size decreases with increasing porosity, and that at the intermediate box size (~0.469 mm in the current model sands) was reproduced well with specific surface area. The maximum configurational entropy increases with increasing porosity, and the entropy length of the pores decreases with increasing specific surface area and was used to calculate the average connectivity among the pores. The correlation among porosity, specific surface area, and permeability is consistent with the prediction from the Kozeny–Carman equation. From the relationship between the permeability and the D_{cc} of pores, the permeability can be expressed as a function of the D_{cc} of pores and porosity. The current methods and these newly identified correlations among structural parameters and properties provide improved insights into the nature of porous media and have useful geophysical and hydrological implications for elasticity and shear viscosity of complex composites of rock, glasses, melt, and fluids as well as transport behavior of ground water.

1. Introduction

Quantification of the network structure of porous media is important for understanding and predicting their transport and macroscopic properties, including the permeability of fluids in partially molten rocks (Takei and Holtzman, 2009), transport behavior of ground water (Gladden et al., 1995; Sahimi, 1993), electrical conductivity and relaxation times for transport in porous media (Avellaneda and Torquato, 1991) and seismic wave attenuation (Bourbié et al., 1987; Li et al., 2001; Takei, 2002, 2005). Such quantification can be used for improving oil recovery from reservoir rocks, predicting pollutant migration in soil, and for the underground storage of CO_2 and nuclear waste. To quantify and characterize irregular and disordered porous networks, diverse structural parameters other than porosity have been introduced. These parameters include fractal dimensions (e.g., Dathe and Thullner, 2005; Ghilardi et al., 1993; Giménez et al., 1997b; Hansen and Skjeltorp, 1988; Tarquis et al., 2008), generalized dimensions (multifractals) (e.g., Muller et al., 1995; Posadas et al., 2003), lacunarity (e.g., Pendleton et al., 2005), and configurational entropy (Andraud et al., 1994) (see Sections 1.1–1.3). Often, more than two parameters of the porous networks are used to gain insights into the nature of porous media. For example, fractal and multifractal (e.g., Bird et al., 2006), fractal and lacunarity (e.g., Armatas et al., 2002), multifractal and configurational entropy (e.g., Tarquis et al., 2006), and lacunarity and configurational entropy (e.g., Chun

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et al., 2008) analyses have been used to characterize diverse natural porous networks. Particularly, the fractal dimension for the 3D pore space has been used to explore the degree of disorder in soil (Moreau et al., 1999; Perret et al., 2003; Tarquis et al., 2008), pumice stone, and quartz sand (Klemm et al., 1997; Müller et al., 1995). These parameters can also be useful for quantifying the fluid distribution with varying degrees of fluid saturation into the porous network. For example, a recent analysis of a water distribution with a 3D fractal dimension and configurational entropy (entropy length) allowed us to quantify the degree of water saturation in the porous model sands with immiscible fluids (Lee and Lee, in preparation).

Despite these key previous studies, the systematic relationships among the structural parameters have not yet been established because most fractal dimension, lacunarity, and configurational entropy analyses of porous networks have been performed on natural systems (e.g., soil) that are characterized by intrinsic heterogeneity in density, composition, and phase. Consequently, the general relationship among these parameters for random networks with well-controlled particle sizes and shapes have not been available thus far. Studies of model porous networks prepared using random packing simulations or model glass beads can facilitate easy control over the diameter and shape of the particles in porous media (Coelho et al., 1997; Garcia et al., 2009; Schwartz and Banavar, 1989). The potential results enable us to establish the systematic and quantitative relationships among these structural parameters. Furthermore, the macroscopic properties of porous media, such as permeability, depend on these structural parameters (Bear, 1972; Dullien, 1979; Johnson et al., 1987). Because previous studies often used 2D images of porous networks rather than 3D networks, the relations among specific surface area, other structural parameters, and permeability remain to be established.

Direct imaging of pore structures is essential to yield information of fractal dimension, lacrunarity, and configurational entropy. Nuclear magnetic resonance (NMR) imaging, or magnetic resonance imaging (MRI), is a nondestructive technique yielding 3D network structure. NMR imaging enables the analysis of different chemical species in fluid phases (as opposed to solid skeletons probed using X-ray based techniques), exploring species with varying relaxation times (e.g., spin–lattice relaxation time and spin–spin relaxation time), and mapping of the flow velocity vectors in porous media (see Blümich, 2000; Callaghan, 1991 and references therein). Therefore, this method has been used to explore distribution of fluids in rocks (e.g., Davies et al., 1994; Dereppe et al., 1991; Doughty and Tomutsa, 1996), chemical reactors (e.g., Gladden, 2003; Lysova et al., 2005), and cements (e.g., Balcom et al., 2003; Faure et al., 2005; Gussoni et al., 2004). Particularly, NMR micro-imaging refers to NMR imaging in which a high spatial resolution (<100 μm) is obtained. 2D NMR micro-imaging studies of porous media have been conducted, including flow velocity in computer-generated percolation clusters (Kossel and Kimmich, 2005), fluid flow and dispersion in random packing of spheres (Manz et al., 1999), self-diffusion maps through chemical products (Gladden et al., 2004), and a fixed bed of ion-exchange resin saturated with methanol (Gladden, 2003). Whereas the resolution in the MRI image for porous media can be more enhanced with the development of high magnetic field and high field gradient NMR imaging equipment (Ciobanu et al., 2003; Lee et al., 2001), it is in general difficult to obtain high resolution MRI image (e.g., <50 μm) for porous media because porous materials often possess low fluid contents and short spin–spin relaxation times. These contribute to poor-quality NMR images (Blümich, 2000). Furthermore, little attention has been devoted to the 3D characterization of porous networks using NMR micro-imaging as most of progress has been made from the 2D imaging. We demonstrate that NMR micro-imaging can be used to obtain 3D images of fluids in various porous media composed of model sands with a spatial resolution of less than ∼50 μm. Model sands can also be generated using random packing simulations where the particle sizes, shapes, and packing density of porous networks can be controlled (Jia and Williams, 2001). This method would allow us to systematically explore unambiguous relationships between changes in network structures and the corresponding transport and macroscopic properties.

As direct probing of 3D structure of pores and fluid in the porous network has become available, we note that the fractal dimension, lacunarity, and configurational entropy should be regarded as intrinsic (and fundamental) properties of random and heterogeneous media in addition to porosity and specific surface area. Therefore, in the current manuscript, we attempt to establish the relationships among these parameters: specific surface area, porosity, and the transport properties.

In this study, diverse porous networks obtained by NMR micro-imaging and random packing simulations have been parameterized into cube counting fractal dimension, lacunarity, and configurational entropy as a function of porosity and specific surface area. Based on these analyses, the current results aim to provide the previously unknown systematic relationships among the porous networks, these key structural parameters, and the corresponding transport and macroscopic properties. Before describing the methods and discussing the results of this study, we provide a brief introduction to the parameters considered in the current study in the following subsections.

1.1. Cube-counting fractal dimension of model porous networks

Fractal dimension—a measure of the degree of complexity of an object—is a noninteger dimension that originates from the power-law relationship between the length scale and the types of objects (e.g., pore space, solid mass, and interface) (Mandelbrot, 1982). The 3D box counting fractal dimension (D_{cc}) used in this study can be described as (Mandelbrot, 1982)

$$\ln N = D_{cc} \ln(1/r) + \ln k, \tag{1}$$

where N is the number of cubes covering the object, r is the side of a cube, and k is a constant. The D_{cc} is the slope of the linear part within the cutoff lengths in the log–log plot.

Table 1 presents the results of fractal dimension analyses from previous studies for the pore space in 3D images of various porous media obtained mostly using X-ray computed tomography (CT) and NMR imaging. The fractal dimension obtained from the 3D images of soil ranged from 2.05 to 2.94 (Moreau et al., 1999; Perret et al., 2003; Tarquis et al., 2008), and those for glass beads, body-centered cubic (bcc) packed spheres, or glass ballotini ranged from 2.42 to 2.52 (Klemm et al., 1997; Müller et al., 1995; Sederman et al., 1997). Fractal dimension for percolation clusters, pumice stone, and quartz sand were 2.53, 2.90, and 2.70, respectively (Klemm et al., 1997; Müller et al., 1995). It has been shown that an increase in the fractal dimension may indicate an increase in the complexity of the network structure (Perret et al., 2003), irregularity of the fractal objects (Muller and McCauley, 1992; Xu and Sun, 2005), porosity (pore fractal) or area fraction (for solid mass fractal) (Perret et al., 2003; Tang and Maragoni, 2008; Yu and Liu, 2004), and the fraction of smaller grains (Avnir et al., 1985; Bartoli et al., 1999; Xu and Dong, 2004).

The fractal object was characterized by self-similarity. Natural porous media exhibit linearity within the lower and upper cutoff lengths in log (box counts)–log (box size) plots (Foroutan-pour et al., 1999; Maria and Carey, 2002). Note that it has already been well-established that fractal dimension can be a useful parameter for the system without apparent self-similarity, such as dilute sets of single-sized spheres (Ciccotti and Mulargia, 2002; Foroutan-pour et al., 1999; Hamburger et al., 1996) and porous networks of model sands, including single-sized spheres (Klemm et al.,