



## Full Length Article

# Influence of particle size on gas-adsorption experiments of shales: An example from a Longmaxi Shale sample from the Sichuan Basin, China



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## HIGHLIGHTS

- Low-pressure CO<sub>2</sub> and N<sub>2</sub> gas adsorption experiments were performed on shale samples with particle sizes from 4 to 0.058 mm.
- The influence of particle size on low-pressure CO<sub>2</sub> and N<sub>2</sub> gas adsorption experiments of shales was analyzed.
- A particle size of 0.113 mm (130 mesh) is optimum for shale samples used in low-pressure CO<sub>2</sub> and N<sub>2</sub> adsorption experiments.

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## ABSTRACT

To explore the optimum particle size range for low-pressure gas adsorption experiments of shales, a sample of the Longmaxi shale from the Sichuan Basin, China, was crushed into various particle sizes between 4 and 0.058 mm. Low-pressure CO<sub>2</sub> and N<sub>2</sub> adsorption experiments were then performed on these crushed samples, and the pore volume, surface area, and pore size distribution (PSD) were determined from isotherms of gas adsorption. Variations in the volume of sorbed gas, pore volume, surface area, and PSD were recorded across the two series of experiments. In the CO<sub>2</sub> adsorption experiments, in which particle size ranged from 4 to 0.83 mm, sorbed gas volume, pore volume, and surface area increased with decreasing particle size. No obvious changes were found in sorbed volume, pore volume, surface area, or PSD for the samples with particle sizes from 0.83 to 0.058 mm. In the N<sub>2</sub> adsorption experiments, the sorbed gas volume and surface area increased with decreasing particle size in samples from 4 to 0.113 mm, whereas no obvious variations were observed in the 0.113–0.058 mm samples. Similarly, pore volume and PSD showed clear changes with decreasing particle size for the 4–0.113 mm samples. The results show that 0.113 mm (130 mesh) is the optimum particle size for low-pressure CO<sub>2</sub> and N<sub>2</sub> adsorption experiments that seek to analyze the characteristics of shale reservoirs.

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## 1. Introduction

Shale gas is an important unconventional gas resource that has been developed commercially in North America and China [1,2]. Due to its potential impact on gas storage and production, shale reservoir properties have been extensively studied. Scanning electric microscopy (SEM) [3–6], atomic-force microscopy (AFM) [7], mercury intrusion porosimetry (MIP) [8], computed tomography (CT) scanning [9,10], nuclear magnetic resonance (NMR) [11], low-pressure gas adsorption (LPGA) [12], and small angle X-ray and neutron scattering are all used to evaluate the reservoir poten-

tial of shales [13,14]. Compared with other methods, LPGA experiments are cost-effective and can provide quantitative information on the pore system in shale. Therefore, LPGA measurements are used extensively when studying the reservoir potential of shale.

In LPGA experiments, shale samples need to be crushed before measurement; however, there is no standard particle size for the crushed shale. Although 0.25 mm (60 mesh) is most widely used [15–18], previous studies have also used 8 mm [19], 0.25–1.4 mm [20], 0.38 mm [21], and 0.15 mm [22]. Given that particle size can influence the pore volume, surface area, and PSD results obtained from LPGA experiments [23], it is important that a consistent particle size range be used to obtain reliable and comparable results of pore characteristics. Chen et al. [23] performed CO<sub>2</sub> and N<sub>2</sub> LPGA experiments on New Albany Shale samples with particle

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sizes of 4.75, 0.83, and 0.25 mm, revealing that pore volume increases with decreasing particle size. This trend is ascribed to incomplete equilibration of the interior of larger shale particles during LPGA experiments [24]. Accordingly, Chen et al. [23] suggested a particle size below 0.83 mm for LPGA experiments. However, the particle size range used in their experiments was relatively narrow, and the trend of pore volume in shale samples with smaller particle sizes may be different from that observed for sizes of between 4.75 and 0.25 mm. Therefore, the influence of particle size on LPGA experiments across a wider range of particle sizes needs to be explored.

In this study, a sample of Longmaxi Shale from the Sichuan Basin was crushed to particle sizes ranging from 4 to 0.058 mm. Low-pressure gas adsorption experiments (CO<sub>2</sub> and N<sub>2</sub>) were then conducted with the aim of assessing the influence of particle size on shale micro- and meso-pore parameters such as pore volume, specific surface area, PSD, and sorbed gas volume. Finally, the optimal particle size ranges for shale samples used in low-pressure CO<sub>2</sub> and N<sub>2</sub> adsorption experiments are suggested.

## 2. Sample and methods

### 2.1. Shale sample

The Longmaxi Shale is the most important shale gas target in China [25]. In this study, a sample of the Lower Silurian Longmaxi Formation was collected from a shale gas well in the Sichuan Basin, Southwestern China. A CS-230 elemental analyzer (Leco Corporation, USA) was used to determine total organic carbon (TOC) content and a D/MAX 2500 X-ray diffractometer was used to measure mineral compositions. The reflectances of the solid bitumens were measured using a MPV-III microphotometer.

The results of the TOC measurements show that the selected sample is organic-rich and has a TOC content of 5.36 wt.% and a eqR<sub>o</sub> value [26] of 1.59% (Table 1). Mineralogical analyses indicate the sample consists mainly of clay minerals (38.5 wt.%) and quartz (35.3 wt.%) with minor feldspar (9.1 wt.%), dolomite (8.6 wt.%), pyrite (7.4 wt.%) and calcite (1.1 wt.%). Mixed-layer illite/smectite and illite form the dominant clay minerals in the studied sample (Table 1).

### 2.2. Low-pressure gas adsorption experiments

The studied sample was crushed to 4, 0.83, 0.38, 0.25, 0.15, 0.113, 0.106, 0.075 and 0.058 mm (corresponding to 5, 20, 40, 60, 100, 130, 150, 200 and 250 mesh, respectively) with a pestle and mortar. The samples were then dried in an oven overnight at 110 °C and degassed under high vacuum (<10 mmHg) for 12 h at 110 °C [27].

Low-pressure CO<sub>2</sub> and N<sub>2</sub> adsorption experiments were performed using a Micromeritics ASAP 2020 surface area analyzer. One to two grams of each of the crushed and degassed samples were exposed to CO<sub>2</sub> and N<sub>2</sub> at the temperatures of 0 °C and –196.15 °C, respectively. CO<sub>2</sub> and N<sub>2</sub> adsorption volumes were measured over the relative equilibrium adsorption pressure ( $P/P_0$ ) ranges of 0.0005–0.0300 and 0.050–0.995, respectively, where  $P_0$  is the condensation pressure of CO<sub>2</sub> or N<sub>2</sub> under laboratory con-

ditions, and  $P$  is the actual gas pressure. CO<sub>2</sub> molecule can access pores with diameter from 0.35 to 1.5 nm, while N<sub>2</sub> adsorption can reflect the pores with diameter between 1.2 and 100 nm. Although the molecule diameter of CO<sub>2</sub> (0.387 nm) is larger than the N<sub>2</sub> (0.315 nm) [28], CO<sub>2</sub> adsorption experiment was performed at 0 °C, the corresponding temperature for N<sub>2</sub> adsorption was –196.15 °C. At the experimental temperatures, the thermal energy of CO<sub>2</sub> molecule is greater than N<sub>2</sub>, which makes CO<sub>2</sub> molecule can penetrate smaller pores [29]. Therefore, the pores measured using low-pressure CO<sub>2</sub> adsorption experiment are smaller than low-pressure N<sub>2</sub> adsorption experiment.

The pore volume, surface area, and pore size distribution (PSD) of each sample were then calculated using the density functional theory method (DFT) [30,31]. This method was chosen because it provides a more accurate description of the thermodynamic properties of the gas in the pores [32]. For N<sub>2</sub> adsorption, the adsorption branches of the isotherms were used.

## 3. Results and discussion

### 3.1. Low-pressure CO<sub>2</sub> adsorption

#### 3.1.1. Isotherms

Fig. 1 shows the low-pressure CO<sub>2</sub> adsorption (LPGA-CO<sub>2</sub>) isotherms for each of the studied size grades. The isotherms are Type I, which indicates the studied sample is a microporous solid. The volume of sorbed CO<sub>2</sub> increases with decreasing particle size from 4 to 0.83 mm (Fig. 1a and b); however, no discernable variation in the volume of sorbed CO<sub>2</sub> is observed in the 0.83–0.058 mm samples (Fig. 1b–i). Higher sorbed CO<sub>2</sub> volumes in the 0.83–0.058 mm samples can be attributed to the higher degree of crushing of these samples. Fig. 2 shows how crushing can expose pores trapped in larger particles. In the larger particle shown in Fig. 2 (diameter = 4 mm) there is an open pore and a closed pore. The CO<sub>2</sub> can pass through the open pore but not the closed pore. In the smaller particle (diameter = 0.83 mm), CO<sub>2</sub> can pass through both pores, which are both open. The sorbed CO<sub>2</sub> volume recorded for samples with a higher abundance of smaller particles ( $\leq 0.83$  mm) is therefore greater than for the sample containing larger particles ( $\geq 4$  mm).

#### 3.1.2. Pore volume and surface area

The pore volumes obtained from the LPGA-CO<sub>2</sub> experiments vary from 0.0034 to 0.0055 cm<sup>3</sup>/g (Table 2). Surface area results calculated from the LPGA-CO<sub>2</sub> data vary from 9.99 to 16.85 m<sup>2</sup>/g (Table 2).

The relationship between particle size and each of calculated pore volume and surface area in the studied shale samples are shown in Fig. 3. The patterns observed in pore volume and surface area across the particle size range considered here match those of the volume of sorbed CO<sub>2</sub> recorded. LPGA-CO<sub>2</sub> measurements show an increase in pore volume with decreasing particle size from 4 to 0.83 mm, and only minor differences in pore volume between the 0.83 and 0.058 mm samples (Fig. 3a).

The pattern for LPGA-CO<sub>2</sub>-calculated surface area follows that of LPGA-CO<sub>2</sub>-calculated pore volume. Minor fluctuations in the vol-

**Table 1**  
TOC content and mineral composition of the studied sample.

Depth (m)	TOC (wt.%)	eqR <sub>o</sub> <sup>a</sup> (%)	Mineral composition (wt.%)							Clay mineral composition (wt.%)			
			Clay	Quartz	Feldspar	Calcite	Dolomite	Pyrite	I/S	Illite	Kaolinite	Chlorite	
3112.06	5.36	1.59	38.5	35.3	9.1	1.1	8.6	7.4	49	33	3	15	

<sup>a</sup> EqR<sub>o</sub> is calculated according to the equation established by Jacob [26]. I/S: mixed-layer illite/smectite.

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