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CO2 storage: threshold capillary pressure estimate in a remoulded caprock specimen / Vespo, Vincenzo Sergio; Stavropoulou, Eleni; Volonté, Giorgio; Messori, Alessandro; Ferrari, Alessio; Musso, Guido; Laloui, Lyesse. - ELETTRONICO. - 1:(2023), pp. 1-2. ( Symposium on Energy Geotechnics (SEG23) Delft (NL) 03.10.23 - 05.10.23) [10.59490/seg.2023.566].

*Availability:*

This version is available at: 11583/2982852 since: 2024-11-08T10:29:27Z

*Publisher:*

TU Delft Open

*Published*

DOI:10.59490/seg.2023.566

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Peer-reviewed Conference Contribution

# CO<sub>2</sub> storage: threshold capillary pressure estimate in a remoulded caprock specimen

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The underground permanent storage of CO<sub>2</sub> is one of the actions promoted by the European Union to achieve neutrality in terms of carbon emissions by 2050. It is the only readily available option for so-called hard-to-abate sectors such as cement, steel, chemical plants, where a significant proportion of carbon dioxide emissions are linked to the industrial process and therefore cannot be avoided by means of electrification or renewables [1].

Ideal geological storage should be able to store CO<sub>2</sub> for millions of years and maintain 99% of stored CO<sub>2</sub> for at least a thousand years. A necessary condition for not having leakage is that the caprock has an adequate sealing capacity, ensured only if the threshold capillary pressure of caprock  $p_c^*$  is larger than the capillary pressure  $p_c$  due to the CO<sub>2</sub> injection. The threshold capillary pressure is experimentally determined through tests in which the non-wetting fluid is forced to penetrate into water-saturated specimen [2].

This work reports the results of two threshold tests performed on remoulded caprock specimens using CO<sub>2</sub> in supercritical conditions (scCO<sub>2</sub>) and air as non-wetting fluid.

The tested material is a non-plastic silty clay with liquid limit  $w_L = 50\%$  and plastic limit  $w_P = 25\%$ . It comes from a disturbed core extracted from a deep formation in the Po Valley (Italy). The material was mixed with distilled water at  $w = 1.2 w_L$  and then consolidated in oedometric conditions up to a vertical effective stress of 1.1 MPa. The porosity at this stress state is equal to 0.43.

Direct determination of the threshold capillary pressure in the laboratory is achieved by a step-by-step increase of the non-wetting fluid pressure at the inlet of a water-saturated sample, while the water pressure is kept constant at the outlet. When the difference between the non-wetting fluid pressure and the water one is higher than the threshold capillary pressure, water drainage occurs, and the non-wetting fluid can break through the sample [3]. Depending on the permeability, several days might be required for one measurement of  $p_c^*$  for a tight rock. The magnitude of non-wetting pressure steps plays a key role in this methodology, since small pressure increments result in long test times, while large pressure increments result in lower accuracy of the threshold capillary pressure estimation. To be able to perform the test using scCO<sub>2</sub> as a non-wetting fluid, it is necessary to set the CO<sub>2</sub> pressure greater than 7.4 MPa and maintain the temperature  $T$  of the entire experimental apparatus greater than 31.1 °C.

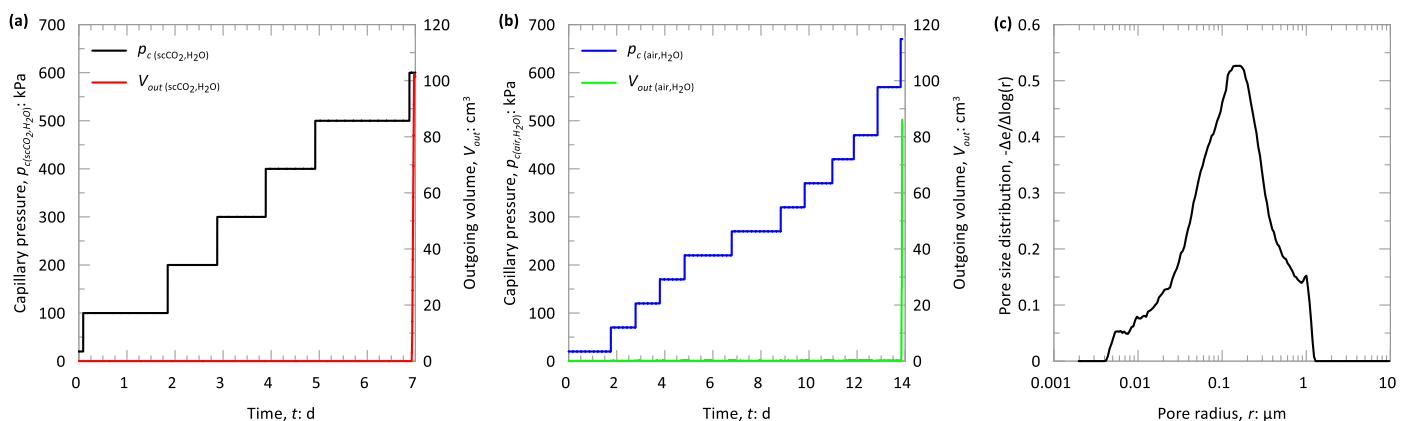
A high-pressure oedometric cell [4] is used to perform the step-by-step tests. This device allows the study of specimens with a diameter of 35 mm and a height of 12.5 mm. Water can be injected at both sides of the sample, two different pumps are used to control pressure up to 16 MPa. A third pump connected to the upstream side is used to inject the non-wetting fluid with a range up to 25 MPa. The axial stress is applied with a hydraulic jack that allows the application of a maximum total vertical stress equal to 100 MPa. The vertical displacements are measured by three LVDTs, which measure the relative displacement of the cell with respect to the piston.

Two tests were performed to determine the threshold capillary pressure of the remoulded caprock specimen in relation to the following fluid systems: scCO<sub>2</sub>/H<sub>2</sub>O and air/H<sub>2</sub>O. Each threshold test consisted of the following stages: at the beginning the sample

was saturated with water (under constant volume) after which a total axial load was applied. After the consolidation was completed, the intrinsic water permeability was assessed ( $k = 5.2 \cdot 10^{-18} \text{ m}^2$ ). Then followed the injection stage of non-wetting fluid. In both tests the effective vertical stress was set equal to 1.1 MPa. The water backpressure and the initial value of non-wetting pressure were set equal to 10 MPa in the scCO<sub>2</sub> test and 1 MPa in the air test. The former test was conducted at  $T = 35 \text{ }^\circ\text{C}$ , the latter at  $T = 25 \text{ }^\circ\text{C}$ .

Figure 1(a) and Figure 1(b) show the trend of capillary pressure  $p_c$  and outgoing volume  $V_{out}$  during the scCO<sub>2</sub> and air injection stage, respectively. The capillary pressure range for which there was a net increase in the outgoing volume was  $500 \div 600 \text{ kPa}$  in the scCO<sub>2</sub> test and  $570 \div 670 \text{ kPa}$  in the air one. The magnitude of outgoing volume implies the overcoming of the threshold capillary pressure  $p_c^*$  and the establishment of a convective flow of non-wetting fluid through the sample. In both tests, no deformation of the specimens occurred during the injection stage.

Therefore the difference between the threshold capillary pressure values determined for the two tests, carried out at an effective vertical stress of 1.1 MPa, is about 70 kPa. Instead, using the tangent method to indirectly estimate the threshold capillary pressure, through the pore size distribution of the remoulded caprock (Figure 1 (c)) and the Washburn-Laplace equation, the difference for the two fluid systems would be equal to about 170 kPa ( $\approx 2.5$  times the real experimental value). This would suggest that the tangent method can only be used for a rough estimate and the threshold capillary pressure is not only a function of the pore size distribution, but also largely depends on the actual pore interconnectivity. For a deeper analysis, further experimental tests are in progress.



**Figure 1: (a) and (b) trend of  $p_c$  and  $V_{out}$  during the injection phase of non-wetting fluid, (c) pore size distribution.**

### Contributor statement

Writing original draft, review & editing: V. S. Vespo, E. Stavropoulou, G. Volonté, A. Messori, A. Ferrari, G. Musso, L. Laloui; Conceptualization: V. S. Vespo, E. Stavropoulou, A. Ferrari, G. Musso; Investigation, visualization: V. S. Vespo; Supervision: A. Ferrari, G. Musso, L. Laloui.

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