Permeability and physical properties of semi-compacted fine-grained sediments – A laboratory study to constrain mudstone compaction trends

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\textbf{ABSTRACT}

Permeability and physical properties of fine-grained clastic sediments show a wide range of variations. Despite rather intensive research, the impact of grain size distribution and mineralogical composition of individual rock constituents is not thoroughly investigated. We performed mechanical compaction of brine-saturated reconstituted borehole cuttings and synthetic quartz-clay mixtures to study the evolution of properties in fine-grained clastic sediments during burial. The primary objective was to examine whether the hydraulic and physical properties of fine-grained sediments could be described and constrained by binary quartz-clay mixtures. The synthetic binary mixtures were prepared by mixing quartz with non-swelling (kaolinite) and strongly-swelling (smectite) clays, which can represent the endmember properties within the clay minerals. In addition to vertical permeability, physical and seismic properties, stress-dependence of permeability, and two-phase relative permeability of brine-oil system were investigated. Experimental results show that grain size distribution and mineralogical composition control the vertical permeability. A well-constrained porosity-permeability bound is defined, where the compaction trends of pure quartz and quartz-smectite 15:85 (wt %) mixtures describe the maximum and minimum boundaries, respectively. The quartz-clay mixtures, however, fail to provide bounds to constrain the broad range of variations in physical and seismic properties of reconstituted aggregates, and consequently natural mudstones. It is crucial to incorporate microstructure into the permeability prediction models because the experiments indicated that the microscale characteristics control the macroscale fluid flow properties.

1. Introduction

Fine-grained clastic sediments are the most abundant deposits of sedimentary basins, and yet among the least investigated sedimentary rocks. Because of the markedly distinct petrophysical characteristics of mudstones and shales compared to the coarser clastic rocks such as sandstones, they are of fundamental importance as caprocks for anthropogenic-related storage sites such as geological CO\textsubscript{2} sequestration and waste repositories (Mallants et al., 2001; Song and Zhang, 2013; Nooraiepour et al., 2018b). Moreover, argillaceous deposits have a profound significance for geological processes, geotechnical applications, and conventional and unconventional petroleum-related activities.

In the first 2–2.5 km burial depth, equivalent to temperatures less than 70–80 °C depending on the geothermal gradient, effective stress controls changes in properties of fine-grained clastic sediments (Bjerlykke and Høeg, 1997). The effective stress is the difference between the overburden load of sediments and fluid pore pressure. Mechanical compaction, hence, governs changes in physical and hydraulic properties of semi-compacted mudstones. The compaction trends for different mudstone properties are, however, difficult to study in natural settings, because various factors such as mineralogy, grain size, depositional environment, excess pore pressure, biogenic content, maximum burial, and exhumation collectively impact the mudstone properties (Revil and Cathles, 1999; Schneider et al., 2011; Bjerlykke, 2015; Daigle and Screaton, 2015; Nooraiepour et al., 2017a,b; Nooraiepour, 2018). Regardless of these complexities, it is necessary to define boundaries, in which physical and hydraulic properties may vary.

For decades, researchers have studied permeability evolution of fine-grained clastic sediments during burial. Several authors have investigated variations in permeability of mudstones as a result of grain size composition (e.g., Dewhurst et al., 1998, 1999; Yang and Aplin,
Laboratory compaction experiments of reconstituted samples are the best way to study the evolution of properties in uncemented mudstones. The research outcomes may provide insights into the evolution of properties in uncemented mudstones in natural settings. As Nooraiepour et al. (2017a) documented, the reconstituted aggregates may show close petrophysical and acoustic properties to mudstones that are still in the mechanical compaction domain. The experiments can simulate compaction of sediments in a natural setting. The dynamic relaxation measurements were performed at the Norwegian Geotechnical Institute (NGI). We conducted mechanical compaction experiments using constant rate of strain protocol at the Norwegian Geotechnical Institute (NGI). We increased effective vertical stress to 25MPa to study changes in permeability and physical properties. All experiments were performed with drained loading condition at room temperature of approximately 19–21°C.

2. Materials and methods

2.1. Sample preparation and characterization

A total of 21 brine-saturated specimens, 12 reconstituted aggregates of borehole drill cuttings and 9 synthetic samples of quartz-clay mixtures, were tested. The reconstituted aggregates were originally the drill cuttings of several mudstone and shale caprock sequences of Triassic-Jurassic-Cretaceous age in the south-western Barents Sea, which were penetrated by wells 7220/10–1 (Salina discovery) and 7122/7–3 (Goliat field). For further details regarding the sampled formations and depth intervals, the reader is referred to Nooraiepour et al. (2017a). After collecting about 100 g of unwashed drill cuttings, all the samples were soaked and rinsed with a suitable solvent (deionized water, Milli-Q) to remove remains of drilling mud and salt from the cuttings. The wells 7220/10–1 and 7122/7–3 were drilled by a commercial water-based mud (seawater) and K/Na polymer mud, respectively (NPD, 2017). To reconstitute the aggregates, the collected samples were sieved, transferred to a separating column, and subsequently freeze-dried. The details of washing, sieving, and freeze-drying to reconstitute mudstone and shale aggregates were presented in Nooraiepour et al. (2017a, b).

The synthetic samples were prepared by mixing quartz grains with non-swelling (kaolinite) and strongly-swelling (smectite) clay minerals. The quartz grains composed of silt-sized (4–63 μm) and very fine sand-sized (63–125 μm) aggregates. The quartz-clay samples were obtained by mixing quartz with different weight percentages of kaolinite or smectite. The quartz-clay weight percentages were 100:0, 85:15, 50:50, and 0:100. Four quartz-clay mixtures were, thus, tested for each of kaolinite and smectite minerals in addition to the pure quartz (100%) sample.

2.2. Experimental setup and procedure

To measure the evolution of permeability and physical properties, we conducted mechanical compaction experiments using constant rate of strain protocol at the Norwegian Geotechnical Institute (NGI). We used an aqueous solution of 0.6M (35g/l) sodium chloride (NaCl, EMSPURE®) in distilled water to prepare the brine-saturated samples. Approximately 55 g of freeze-dried reconstituted and synthetic aggregates were used to prepare each brine-saturated sample. The brine-saturated samples were tested using a high-stress oedometer equipped with acoustic measurement transducers. The oedometer cell allows an increase of vertical stress to 100 MPa. The cross-section of the utilized high-stress oedometer and a schematic of the experimental setup are shown in the appendix. Constant rate of strain (CRS) experiment was performed to measure compressibility of the brine-saturated samples (Wissa et al., 1971; ASTM, 2006; Gutierrez et al., 2015). Aiming to simulate a close to hydrostatic pressure condition, we selected a strain rate of 0.67% per hour, which corresponds to initial deformation of 0.2 mm/h for an initial sample height of 30 mm. In these experiments, we increased effective vertical stress to 25 MPa to study changes in vertical permeability and physical properties. All experiments were performed with drained loading condition at room temperature of approximately 19–21°C.

The dynamic elastic moduli were extracted from the bulk density (ρ) and ultrasonic wave velocity measurements. The high-stress oedometer is equipped with two piezoelectric transducers with a resonant...
frequency of 500 kHz to generate and receive high-quality ultrasonic signals. To measure compressional ($V_p$) and shear wave ($V_s$) velocities, we used the pulse transmission technique. A detailed description of the experimental setup and derivation of physical properties during compaction experiments is given in the appendix.

### 2.3. Absolute and relative permeability measurement

The constant rate of strain (CRS) compaction technique offers a direct method for computing vertical hydraulic conductivity continuously during the test. Fig. 1 demonstrates the applied stress and drainage condition for the performed uniaxial CRS compaction experiment. While the non-moving pedestal was kept undrained, the moving piston allowed the pore fluid to drain to atmospheric pressure (Fig. 1). To calculate the vertical permeability of the samples, we followed Wissa et al. (1971) and ASTM (2006) procedure. The permeability values were computed continuously at 5 min time intervals during the compaction experiments, and the best-fit lines are presented here. To ensure reliability of the CRS-driven permeability curves, each specimen was also subjected to single-phase flow direct permeability measurement. To measure absolute permeability, we performed steady-state flow-through experiments at a constant pressure gradient condition and calculated vertical permeability using Darcy’s law (Nooraiepour et al., 2018a).

The unsteady-state two-phase relative permeability of brine-oil (polysiloxane fluid, SF1147 Momentive) system was measured for one of the reconstituted specimens, which showed high TOC content (12.4%). It was selected to represent an uncedmented organic-rich mudstone in the mechanical compaction domain of the sedimentary basin. We conducted relative permeability ($k_r$) measurements only for one specimen to show capability of the technique and possibility of measuring unsteady-state $k_r$ curves for uncedmented fine-grained sediments. The aggregates were collected from the Hekkingen Formation, which was penetrated by well 7122/7–3. The drainage and imbibition cycles were conducted to investigate displacement characteristics of wetting and non-wetting phases. To mimic the primary flow direction that would occur in most of the caprocks, the drainage and imbibition phases were conducted in a vertical displacement mode.

The reader is referred to the appendix for further explanations and details of the experiments.

### 3. Results

#### 3.1. Characterisation of sample properties

Fig. 2 presents grain size distribution and mineralogical composition of whole-rock (bulk) and clay mineral fractions. The ternary plot of Fig. 2a demonstrates grain size classes of reconstituted aggregates and synthetic mixtures. The contents of clay-sized particles (< 4 μm according to the Wentworth classification) in the reconstituted samples vary from 21% to 57%, and silt-sized aggregates (4–63 μm) span 34%–63% (Fig. 2a). The sand-sized fractions (> 63 μm) vary between 7% and 20%. Fig. 2b shows that the whole-rock mineralogy of reconstituted aggregates comprises quartz, clay minerals,feldspars(plagioclase and K-feldspar), carbonates, and small amounts of pyrite. The content of quartz and feldspars, and clay minerals are 34–69% and 24–50%, respectively (Fig. 2b). The carbonates and pyrite compose 5–17% of the whole-rock mineralogy (Fig. 2b). The clay minerals consist mainly of varying amounts of kaolinite and illite in addition to small amounts of chlorite (less than 6%) in most of the reconstituted aggregates. Fig. 2c presents the content of the non-swelling (kaolinite) and weak-swelling (illite and chlorite) clays for the reconstituted aggregates within the total clay content, and they are 26–90% and 10–74%, respectively. The strongly-swelling clays (smectite) were not identified within the reconstituted aggregates (Fig. 2c).

In addition, Fig. 2 demonstrates the grain size distribution and mineralogical composition of the quartz-clay mixtures. The synthetic specimens rich in kaolinite and smectite minerals are shown by blue and red triangles, respectively.

#### 3.2. Evolution of mudstone properties during compaction

Experimental compaction results of brine-saturated reconstituted aggregates and synthetic quartz-clay mixtures are presented in Figs. 3 and 4, respectively. Figs. 3 and 4 demonstrate the evolution of (a) vertical permeability, (c) bulk modulus, (d) shear modulus, and (e) Poisson’s ratio as effective vertical stress was increased in the laboratory to 25 MPa. Changes of mentioned properties are also cross-plotted against total porosity during compaction in Figs. 3 and 4. Color code of the reconstituted aggregates is consistent throughout the paper. Changes in pore pressure during the tests are shown in the appendix.

Experimental results show approximately five orders of magnitude reduction in vertical permeability during compaction, two to three orders of which occurred between 1 and 25 MPa effective vertical stresses (Figs. 3a and 4a). A rapid increase of bulk modulus is observable in the early compaction stages (1–10 MPa) (Figs. 3c and 4c). The rapid increase is attributed to the significant porosity loss at low-stress levels as the bulk modulus indicates how incompressible the samples are. The rate of the compressibility decreases after 10 MPa effective vertical stress and the bulk modulus continues to increase almost linearly afterward. This linear gradient is mainly associated with rearrangement, reorientation and closer packing of the grains. Therefore, similar to the Velde (1996) results, an exponential and a linear sub-stage can be considered for the mechanically dominated compaction. The shear modulus or rigidity shows a steady and gentle increase during mechanical compaction (Figs. 3d and 4d). The Poisson’s ratio shows a considerable drop throughout the experiments. The decline is more notable in the first 10 MPa effective vertical stress (Figs. 3e and 4e). When specimens resemble extremely soft water-saturated sediments or suspension of particles in a fluid, the values of Poisson’s ratio are around 0.45 and approaching 0.5. As effective vertical stress increases, the Poisson’s ratios experience an instant fall and continue to decrease with a gentler slope toward the end of the tests (Figs. 3e and 4e). The scatter in mechanical properties of the tested specimens is relatively small at early stress levels, but it increases with the increase in effective vertical stress (Figs. 3 and 4).

Comparison between the reconstituted aggregates and synthetic quartz-clay mixtures (Figs. 3 and 4) indicates that the reconstituted aggregates of mudstone and shale caprock sequences show higher bulk moduli, higher shear moduli, and lower Poisson’s ratios compared to the synthetic quartz-clay mixtures. Moreover, the reconstituted aggregates show higher compaction and lower total porosity (Figs. 3a and 4a). It is, however, the synthetic quartz-clay mixtures that span a wider permeability range and indicate the lowest and highest permeability.
bounds (Fig. 4a).

For the reconstituted aggregates (Fig. 3), the endpoint values at 25 MPa effective vertical stress span approximately the following ranges: vertical permeability 0.0001–0.01 mD, total porosity 9–25%, bulk modulus 11–15 GPa, shear modulus 2.4–4.3 GPa, and Poisson’s ratio 0.35–0.42. Fig. 4 shows a distinct compaction behavior for porosity and permeability of quartz-kaolinite and quartz-smectite mixtures. The quartz-kaolinite mixtures are more prone to compaction, show lower total porosity (19–29%) but higher vertical permeability (0.009–0.003 mD) compared to quartz-smectite mixtures (26–40% porosity and 0.00001–0.0001 mD permeability). The pure 100% quartz sample, a mixture of silt-sized and very fine sand-sized quartz, demonstrates the highest incompressibility, total porosity (24.5%) and vertical permeability (0.3 mD). The quartz-kaolinite mixtures have higher bulk and shear moduli and lower Poisson’s ratios compared to the quartz-smectite samples (Fig. 4c–e). For the synthetic quartz-clay mixtures (Fig. 4), the endpoint values at 25 MPa effective vertical stress vary approximately in the following ranges: bulk modulus 6.2–11.2 GPa, shear modulus 0.75–2.1 GPa, and Poisson’s ratio 0.41–0.46.

3.3. Stress-dependence of absolute permeability

Fig. 5 presents the changes in vertical permeability (k_v) of reconstituted aggregates as a function of applied vertical stress and pore pressure during the loading and unloading (compression and decompression) cycles. The direct permeability measurements in addition to best-fit regressions are demonstrated here. As shown in Fig. 5a, the vertical permeability decreases rapidly and by approximately three orders of magnitude during the loading cycle for two different reconstituted samples. The rate of the change decreases as applied vertical stress increases. During unloading, however, vertical permeability changes slightly from 30 to 10 MPa applied stress, and a more notable increase is only observable when the applied stress declines from 10 to 1 MPa. The increase in vertical permeability during unloading is less than half order of magnitude (Fig. 5a). Fig. 5b shows vertical permeability measurements of a reconstituted sample during unloading at three different pore pressure levels (0.5, 1 and 2 MPa). Fig. 5b indicates that the measurements with the highest pore pressure (2 MPa) have produced the highest increase in permeability during the reduction of applied vertical stress. The difference between the permeability measurements at different pore pressures increases with the decrease in applied stress (Fig. 5b).

3.4. Two-phase relative permeability

Fig. 6 shows two-phase relative permeability of brine-oil system for a sample of reconstituted organic-rich aggregates. The relative permeability curves are plotted in linear and semi-logarithmic scales in Fig. 6a and b, respectively. The vertical permeability of the specimen was 0.1 microDarcy. The endpoint permeability to non-wetting phase (oil) was calculated to be in the 50 to 100 picoDarcy range. The permeability to the non-wetting phase (oil) at irreducible brine saturation, thus, was approximately one-tenth of the measured brine permeability at 100% brine saturation.

The irreducible brine saturation during the drainage cycle is around 58%. Brine relative permeability is higher during imbibition than during the drainage cycle. Conversely, the oil relative permeability, as the non-wetting phase, is higher during drainage compared to imbibition. Because displacement of brine saturation inside the pores (by injected oil) depends on the interfacial tension (IFT) between the two fluid phases (Bachu and Bennion, 2008), it is expected that at a lower IFT the non-wetting fluid passes through the pore throats more easily and show higher relative permeability. The imbibition relative permeability curves suggest that small amounts of trapped oil can have a large influence on brine permeability. It might be caused by the subtle pores of the mechanically compacted aggregates and considerable capillary forces between the two immiscible fluids in such a tight medium.

4. Discussion

4.1. Permeability variations with grain size distribution and mineralogical composition

In Fig. 7, vertical permeability of reconstituted aggregates and synthetic quartz-clay mixtures at 20 MPa effective vertical stress are plotted versus grain size distribution and mineralogical composition. These values may represent the range of permeability variations in mudstones at around 2 km burial depth before the onset of chemical compaction and cementation. The first row in Fig. 7 shows vertical...
permeability against three grain size classes, namely, clay-, silt- and sand-sized fractions (Fig. 7a–c). The second row in Fig. 7 demonstrates vertical permeability versus bulk mineralogical content (Fig. 7d–f). At effective vertical stresses equivalent to 2 km burial depth, the reconstituted aggregates show vertical permeabilities varying from approximately $10^{-2}$ mD to $10^{-4}$ mD (Fig. 7). The synthetic quartz-clay mixtures range approximately between $10^{-2}$ and $10^{-5}$ mD (Fig. 7). The highest and lowest permeabilities among binary mixtures are recorded for the quartz-kaolinite 85:15 and quartz-smectite 15:85 (wt %) specimens, respectively.

In Fig. 7a, there is a negative correlation between the vertical permeability of reconstituted aggregates and the content of clay-sized particles, which is consistent with the previously published research (e.g., Gamage et al., 2011; Schneider et al., 2011; Daigle and Screaton, 2015). The correlation is positive between the silt-sized fractions and the permeability (Fig. 7b). In other words, the overall trend demonstrates that lower amounts of clay-sized particles and higher silt-sized fractions yield higher permeability in reconstituted samples. Although several studies (for instance, Yang and Aplin, 2010) proposed that samples with similar content of clay-sized particles show the same permeability, the results of present study suggest that it may not be a universal observation (Fig. 7a). In particular, Fig. 7 demonstrates one order of magnitude scatter in permeability of reconstituted aggregates and two orders of magnitude difference between quartz-kaolinite and quartz-smectite mixtures at the same range of clay-sized particles (Fig. 7a). The synthetic quartz-clay mudstones show an overall decrease
in permeability with the increase of clay-sized content. The relationship, however, is not one-to-one. In other words, a higher clay-sized content does not necessarily mean a lower permeability. It implies that it is the best-possible packing of different size classes that produces the most low-permeability porous medium. As is shown in Fig. 7c, the samples with more sand-sized particles indicate higher permeability, particularly when sand-sized content is more than 10% and clay-sized content is low. Higher sand-sized content, however, does not necessarily mean higher total porosity (Figs. 3 and 4).

It is expected that rigid grains like quartz and feldspar withstand the applied stresses and, thus, preserve total porosity, while ductile particles like clays, mica, and rock fragments bend and block the inter-particle pores. The ductile particles, thus, can significantly reduce matrix permeability (Fawad et al., 2010; Zhao et al., 2016). A positive correlation between vertical permeability and content of quartz and feldspar in reconstituted aggregates is observable in Fig. 7d. The correlation is negative between the clay mineral fractions and vertical permeability (Fig. 7e). Simply stated, permeability decreases with the increase of clay minerals and decrease of quartz and feldspar. The samples with higher content of carbonates also show lower
permeabilities (Fig. 7f). The cross-plots of vertical permeability versus bulk mineralogical content for synthetic quartz-clay mixtures (Fig. 7d–f) demonstrate that permeability of clay-rich sediments is strongly dependent on the type of clays, particularly non-swelling (kaolinite) and strongly-swelling (smectite) clay minerals. Although it is generally assumed that the higher the content of clay minerals the lower the permeability, permeability of the quartz-clay mixtures suggest that in addition to the clay type, grain mixing ratio, packing scenarios, and preferred orientation of grains is required to be taken into account. For example, quartz-kaolinite 50:50 and quartz-smectite 15:85 (wt %) synthetic mixtures produced the endmember low-permeability among the kaolinite- and smectite-rich mixtures.

As shown earlier (Figs. 4 and 7), strongly-swelling clays (smectitic) impose control on permeability, which can be associated with the grain size of these clays (Brigatti et al., 2013) and the subsequent influence of pore size and pore throat on the fluid flow. Moreover, large specific surface area of the smectite clay group (inversely proportional to grain size) can prevent pore fluid from participating in flow and, thus, leads to the reduced permeability. We have found no correlation between the vertical permeability of reconstituted aggregates and the content of kaolinite, illite, and chlorite clay minerals in the bulk mineralogy (multiplication of bulk clay content by the fraction of each clay mineral derived from clay XRD analyses). Illite and chlorite clay minerals can form pore-bridging and pore-lining morphologies in the porous medium, which can cause significant permeability loss (e.g., Neasham, 1977). However, if these minerals are not authigenic such as the content of illite and chlorite within the reconstituted aggregates, they have a far less negative influence on flow properties because they lack the pore-bridging and pore-lining morphologies.

4.2. Porosity-permeability relationships

Fig. 8 presents the semi-logarithmic cross-plot of vertical permeability as a function of total porosity. It shows variation in permeability of reconstituted aggregates and quartz-clay mixtures rich in kaolinite and smectite clay minerals. The green background illustrates published data set for the natural mudstones rich in mixed clay types (Neuzil, 1994; Gamage et al., 2011; Daigle and Screaton, 2015). The logarithmic-linear $k_r$-$\phi$ relationship indicates that the permeability decline is much faster than the porosity loss, and the decrease in permeability is more rapid at higher porosities. A relatively well-constrained porosity-permeability bound is observable, where the maximum and minimum boundaries are described by pure quartz and quartz-smectite 15:85 (wt %) compaction trends, respectively. Approximately, five orders of magnitude dispersion between the two boundaries indicates the potential variability of fluid flow properties in fine-grained sediments and mudstones within the mechanical compaction domain. The quartz-kaolinite mixtures fail to provide compaction boundaries for the

Fig. 5. Stress-dependence of vertical permeability. (a) Changes in permeability of two reconstituted samples during loading and unloading cycles. (b) Changes in permeability of a reconstituted sample during unloading cycle at three different pore pressure levels (0.5, 1 and 2 MPa).

Fig. 6. Laboratory measurement of two-phase relative permeability of brine-oil system for a reconstituted organic-rich sample in (a) linear and (b) semilogarithmic scales.
changes in porosity and also permeability of reconstituted aggregates, in particular for the lower than 30% porosity values. Two orders of magnitude dispersion in permeability of reconstituted aggregates is observable at a given total porosity (Fig. 8). Overall, the reconstituted aggregates are compacted more and show lower total porosity compared to quartz-clay mixtures. When we compare compaction trend lines for various mixtures of quartz-kaolinite and quartz-smectite specimens, it can be suggested that the best-possible packing, grain size, and specific surface area of the sediments are the controlling factor in porosity-permeability relationships (Fig. 8). The same rationale can be used to explain high permeability of pure quartz sample, which was characterized with bigger grain size and consequently bigger pores and pore throats available for fluid flow compared to quartz-clay mixtures.

According to the analyses presented in Fig. 7, grain size distribution and mineralogical composition are among the controlling factors in porosity-permeability relationships of fine-grained sediments and mudstones. To describe single-phase permeability of mudstones and shales, porosity-permeability relationships are widely proposed because porosity is a routine and usually available measurement. The empirical relationships are typically developed from laboratory measurements (e.g., Yang and Aplin, 2010), theoretical models such as the Kozeny-Carman equation (e.g., Chapuis and Aubertin, 2003) and the Hagen–Poiseille equation (e.g., Civan et al., 2011), or binary mixing models (e.g., Revil and Cathles, 1999). These approaches assume some macroscale assumptions that correlate total porosity to permeability and apply a tuning factor related to mineralogy, clay fractions, or clay surface area. The present experimental results indicate that seeking to approximate vertical permeability of fine-grained sediments and mudstones with a single macroscale equation may not provide a universal solution because multiple parameters with not a one-to-one correspondence affect flow characteristics. Incorporating microstructure characteristics of mudstones into the permeability models is a necessity because they control the macroscale fluid flow properties.

4.3. Permeability–seismic properties relationships

In Fig. 9, we have presented semi-logarithmic cross-plots of seismic and geomechanical properties of reconstituted aggregates and synthetic mixtures versus vertical permeability. It shows (a) P-wave acoustic impedance \( (I_p) \) [m/s\(^2\)g/cm\(^3\)], (b) S-wave acoustic impedance \( (I_s) \) [m/s\(^2\)g/cm\(^3\)], (c) \( \lambda \rho \) [GPa\(^2\)g/cm\(^3\)], and (d) \( \mu \rho \) [GPa\(^2\)g/cm\(^3\)].

Each compaction curve shows a strong correlation between vertical
permeability and seismic and geomechanical properties, which in turn is a function of total porosity. However, it is not possible to draw a generalized relation for all the reconstituted borehole aggregates. In contrast to porosity-permeability relationships, the synthetic quartz-clay mixtures illustrate a limited span compared to the range of variations in seismic and geomechanical properties of reconstituted aggregates (Fig. 9). The cross-plots of Fig. 9 suggest that the relationship between seismic responses and permeability, even for single-phase flow, is dependent on microstructure characteristics. In addition to mineralogy and grain size, Nooraiepour et al. (2017b) have shown that the content of the siliceous biogenic material influence the physical properties of mudstones. In particular, biogenic skeletal debris may decrease bulk density, increase $V_s$, decrease $V_p/V_s$ ratio, and consequently change other seismic and geomechanical properties. In the absence of microstructural information, the in-situ permeability of mudstones may not be reliably determined using geophysical techniques such as seismic surveys.

4.4. Two-phase flow properties

As shown in Fig. 6, brine permeability is higher during imbibition cycle than the drainage. It has, however, also been reported that in some extremely low permeability caprocks, brine relative permeability is lower during imbibition compared to drainage (Burnside and Naylor, 2014). This observation is mainly attributed to the trapping of the non-wetting phase in the extremely tight porous medium, in particular, gaseous fluids like CO₂. Comparison of present two-phase fluid flow results with the reservoir samples (e.g., Bennion and Bachu, 2013; Burnside and Naylor, 2014; Ruprecht et al., 2014) shows that mudstones have markedly distinct absolute and relative permeability characteristics, such as (a) considerably lower absolute permeability to brine; (b) smaller displacement efficiency during drainage (brine

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**Fig. 8.** Semilogarithmic cross-plot of vertical permeability versus total porosity. The circles and squares represent reconstituted aggregates collected from 7220/10–1 and 7122/7–3 wells, respectively. The synthetic quartz-clay mixtures rich in kaolinite and smectite minerals are outlined with blue and red color polygons, respectively. The yellow line represents the pure quartz sample. The notations stand for Qz = quartz, Sm = smectite, and Ka = kaolinite. The green background shows the published date set for the natural fine-grained sediments rich in mixed clay types (Neuil, 1994; Gamage et al., 2011; Daigle and Screaton, 2015). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

**Fig. 9.** Semilogarithmic cross-plot of vertical permeability versus (a) P-wave acoustic impedance ($I_p$) [m/s·g/cm³], (b) S-wave acoustic impedance ($I_s$) [m/s·g/cm³], (c) $\lambda\rho$ [GPa·g/cm³], and (d) $\mu\rho$ [GPa·g/cm³]. The circles and squares represent reconstituted aggregates collected from 7220/10–1 and 7122/7–3 wells, respectively. The seismic properties of smectite-rich and kaolinite-rich mixtures are outlined with blue and red color polygons, respectively. The yellow line represents the pure quartz sample. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
substitution by the invading non-wetting phase), which results in high residual water saturation; and (c) high relative permeability hysteresis during imbibition once trapped non-wetting phase is present in the pore space. Although such properties are undesirable for reservoir rocks, they are favorable for the mudstone caprocks. It indicates that unless vertical thickness of the caprock sequence is very thin and it is exposed to extremely high overpressure compared to the upper side of the caprock, matrix flow of non-wetting phase through such mudstone samples would be minimal to nonexistent. The caprocks with similar characteristics have the potential to provide long-term sealing for the injected CO₂ in geological CO₂ storage projects.

4.5. Limitations and uncertainties

We have conducted mechanical compaction experiments on the reconstituted mudstone and shale aggregates collected from several caprock sequences in the south-western Barents Sea. In addition, quartz-kaolinite and quartz-smectite binary mixtures were tested as the endmember size and mineralogy components in fine-grained sediments and mudstones. Laboratory compaction of brine-saturated specimens, however, may cause reduced anisotropy and clay preferential orientation compared to natural settings. Such differences arise because fine-grained aggregates may become settled and deposited differently in nature. The texture and packing of the reconstituted aggregates may also vary from the original caprock sequences. Development of high overpressures, undrained loading, calibration and measurement precision, and violation of other guidelines set by ASTM (2006) may cause
further uncertainties in the results of a CRS test. Measuring dynamic elastic properties instead of static ones during compaction experiments can be other consideration. Nonetheless, the abundance of published literature on the laboratory compaction of fine-grained samples indicates that the technique provides valuable information for understanding natural processes. For instance, the comparison between the naturally-driven (reconstituted aggregates) and synthetic binary samples (quartz-clay mixtures) demonstrate how properties of fine-grained sediments with different composition of grain size and mineralogy may change during early burial as a result of effective stress. These experiments try to examine some of the possible natural scenarios with sample properties presenting potential boundary conditions. Moreover, packing of constituent and its impact on binary and multicomponent mixtures were investigated. As fine-grained sediments undergo mechanical compaction, they transform from mud to mudstones. The laboratory observations derived from reconstituted and synthetic mud-sized aggregates, therefore, present range of compaction behavior in fine-grained sediments and mudstones, and thus, the outcomes help to better comprehend the evolution of mudstones' properties in nature.

5. Conclusions

We have shown that grain size distribution and mineralogical composition impose control on the vertical permeability of fine-grained sediments and mudstones. The presented results focus exclusively on uncemented mudstones in the mechanical compaction domain of sedimentary basins. The samples with lower clay-sized, higher silt-sized, and higher sand-sized fractions indicate higher permeability. In particular, when sand-sized content is more than 10% and clay-sized content is low. The permeability decreases with the increase of clay minerals, increase of carbonates, and decrease of quartz and feldspar. The mentioned grain size- and mineralogy-permeability relationships, however, is not one-to-one. In other words, a higher or lower content of one constituent does not necessarily mean a higher or lower permeability. Two orders of magnitude difference between the permeability of kao-linite- and smectite-rich mixtures is observed for the entire range of size and mineralogy classes. Although several studies proposed that mudstones with similar content of size or mineralogy classes show similar permeability, the observed scatter in permeability of synthetic quartzstones with similar content of size or mineralogy classes is not one-to-one. In other words, a higher or lower content of one constituent does not necessarily mean a higher or lower permeability. Two orders of magnitude difference between the permeability of kao-linite- and smectite-rich mixtures is observed for the entire range of size and mineralogy classes. Although several studies proposed that mudstones with similar content of size or mineralogy classes show similar permeability, the observed scatter in permeability of synthetic quartz-clay mixtures and reconstituted aggregates at a given value of grain size or mineralogy suggest that it may not be a correct and universal assumption.

A relatively well-constrained porosity-permeability bound is defined, where the pure quartz and quartz-smectite 15:85 (wt %) compaction trends describe the maximum and minimum boundaries, respectively. We have documented three to four orders of magnitude dispersion in permeability at a given total porosity between the two boundaries. Type of clay minerals (i.e., strongly-swelling and non-swelling clays) influences the porosity-permeability relationship notably. While the most low-porosity low-permeability sample among the kaolinite-rich mixtures is the quartz-kaolinite 50:50, the smectite-rich mixtures show different behavior. The most low-porosity and the most low-permeability mixtures are different from each other and are quartz-smectite 50:50 and 15:85 specimens, respectively. The quartz-kaolinite mixtures cannot provide compaction boundaries for the changes in the porosity and permeability of reconstituted aggregates, in particular for total porosity values less than 30%.

Comparison of relationships between permeability, physical and seismic properties indicates that the reconstituted aggregates can have higher elastic moduli and seismic properties, higher compaction, and lower total porosity compared to quartz-clay synthetic mixtures. As a result, the quartz-clay mixtures fail to provide bounds to constrain the broad range of variations in physical and seismic properties of reconstituted aggregates, and consequently natural mudstones. The present experimental results demonstrate that seeking to approximate vertical permeability of fine-grained sediments with a single macroscale relationship may not provide an accurate solution. Incorporating microstructure characteristics into the permeability models was found to be a necessity as they control the macroscale fluid flow properties. The permeability of reconstituted aggregates also demonstrates a stress-dependent behavior, which influences flow properties during compression and decompression, or burial and uplift. Changes in effective stress, thus, need to be considered when the permeability of fine-grained sediments are being modeled and evaluated. In the absence of such information, the in-situ permeability of mudstones may not be reliably determined using geophysical techniques such as seismic surveys.

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Appendix. Experimental compaction experiments

To perform mechanical compaction experiments, we placed a high-stress oedometer cell within a hydraulic load frame (VDL series, Enerpac). Fig. 10 presents schematics of the oedometer cell and experimental setup. A GDS pressure-volume controller provided precise regulation of applied vertical stress at the hydraulic load frame. Applied stress was recorded through a compaction-logging software, which was programmed to take into account hydraulic pressure drops, internal frictions, dead loads, and the false deformations. A pore pressure sensor, which was connected to one of the bottom drainage tubes of the oedometer, measured the pore pressure development during the test. In the experimental setup, stress is applied along the vertical axis (uniaxial strain), and strain is prevented in radial directions (ε2 = ε3 = 0). Two diametrically opposed LVDT displacement transducers (ACT 500A, RDP Electronics) measured changes in height during the test. The axial strain (εa) was computed using the following relationship:

$$\varepsilon_a = \frac{dH_{\text{mean}}}{H_i} = \frac{dH_1 + dH_2}{2H_i},$$

(1)

where $dH_1$ and $dH_2$ are axial deformations measured by LVDT displacement transducers, $dH_{\text{mean}}$ is the average axial deformation, and $H_i$ is the initial height of the specimen.

The initial porosity ($\phi_i$) and initial bulk density ($\rho_{bi}$) were calculated at standard ambient conditions (atmospheric pressure and room temperature):

$$\phi_i = \frac{V_i - V_{dry}}{V_i} = 1 - \frac{(m_{dry}/\rho_{dry})}{V_i},$$

(2)
\[ \rho_{bi} = \sum_{i=1}^{k} \rho_i \, \rho_r \]  
(3)

where \( V_t \) is the initial bulk volume of the sample, \( V_{dry} \) is the volume of dry aggregates, \( m_{dry} \) and \( \rho_{dry} \) mass and density of dry aggregates, respectively. The \( \rho_i \) and \( \rho_r \) are the weight fraction and the grain density of each mineral constituent in the specimen, respectively.

It is assumed that individual grain compressibility is insignificant in comparison with the bulk compressibility of the specimen (Wissa et al., 1971; Mondol et al., 2008; Nooraiepour et al., 2017a). It implies that the bulk volume changes of the specimen can be considered equal to the pore volume changes during the test. While having zero radial strain in oedometric configuration, the axial strain was used to compute changes in total porosity throughout the experiment. We used the following relationships to determine continuous changes of total porosity (\( \varphi \)) and bulk density (\( \rho_b \)):

\[ \varphi = \frac{\rho_b - \rho_i}{1 - \rho_i}, \]  
(4)

\[ \rho_b = (1 - \varphi) \rho_i + \varphi \rho_f, \]  
(5)

where \( \rho_f \) is the density of saline pore fluid. The brine-saturated samples were prepared by using 35000 ppm NaCl brine with an approximate density of 1.025 g/cm\(^3\). Preparing the brine-saturated specimens in a cold room with a humidity around 90\% minimized evaporation from the slurries. In addition, necessary precautions were taken to avoid uncertainty in the mass of the used material. The experimental setup, procedure of mounting the specimens, and testing conditions were also described in Nooraiepour et al. (2017a).

Fig. 11 shows the developed pore pressure during the compaction experiments at drained loading condition for the reconstituted aggregates and quartz-clay mixtures. The pore pressure increased rather quickly between 5 and 25 MPa applied vertical stress when the CRS test progressed relatively faster to keep the strain rate constant during the experiment (Fig. 11). The increase is more notable for quartz-smectite mixtures. However, as the tests reached 25 MPa applied vertical stress and no further vertical forces were applied, the pore pressure dissipated quickly. The compaction experiments, thus, were carried out at a close to hydrostatic pressure condition with only minor overpressures. A total of four brine-saturated experiments (two reconstituted and two synthetic mixtures) were randomly selected and repeated to check the repeatability of compaction trends. The acquired data confirmed the precision and repeatability of the compaction and acoustic results.

### Dynamic elastic properties

Through the pulse transmission technique, we measured travel time of the acoustic signals that passed a sample of known height, and then, converted the travel time to ultrasonic velocity. In Nooraiepour et al. (2017a) we have discussed development of \( V_p \) and \( V_s \) as a function of effective vertical stress and total porosity. The following relationships were subsequently used in the calculation of dynamic elastic moduli:

\[ K = \rho \left( V_p^2 - \frac{4}{3} \frac{V_s^2}{V_p} \right), \]  
(6)

\[ \mu = \rho V_s^2, \]  
(7)

\[ \nu = \frac{V_p^2 - 2V_s^2}{2(V_p^2 - V_s^2)}, \]  
(8)

where \( K \) is bulk modulus (GPa), \( \mu \) is shear modulus (GPa), and \( \nu \) is Poisson’s ratio.

The largest sources of error in velocity measurements were: (a) determination of arrival time from the raw waveform, (b) sampling interval for digitizing the waveforms in the ultrasonic recording system, and (c) measurement of the sample height. The estimated relative errors in ultrasonic wave velocity were varying from 5\% to 2\% for \( V_p \) and \( V_s \) during the experiment. Details of acoustic velocity measurements and error analyses were explained by Nooraiepour et al. (2017a). It can be claimed that the use of these isotropic equations rather than the complete anisotropic relations for a potential VTI medium is not entirely precise. However, Sone and Zoback (2013) have demonstrated that the error of using apparent dynamic modulus against the true dynamic modulus is small and the estimations are in close agreement.

### Absolute permeability measurements

#### Indirect permeability measurement

According to the ASTM (2006), the underlying assumptions for interpretation of variations in hydraulic conductivity as a function of effective vertical stress are: (1) homogenous water-saturated specimen; (2) negligible compressibility of pore fluid and individual solid grains; (3) negligible compressibility of pore pressure measurement system compared to the sample; (4) vertical flow of pore fluid; (5) Darcy flow; and (6) constant ratio of hydraulic conductivity and compressibility between two separate readings (but it can vary during the experiment).

Wissa et al. (1971) have published a theoretical solution for one-dimensional linear consolidation behavior of the CRS tests. The Wissa’s relationship for the hydraulic conductivity of the specimen, modified for large strains is given as (Adams, 2011):

\[ K = \frac{e_r H_t H_i \gamma_w}{2 P_p}, \]  
(9)

where,

\[ e_r = \frac{\Delta e_r}{\Delta t} = \frac{\Delta H_t/H_i}{\Delta t}, \]  
(10)

where \( e_r \) is the imposed constant loading rate (strain rate) [T\(^{-1}\)], \( \Delta e_r \) is the change in axial strain, \( \Delta t \) is the time interval [T], \( K \) is the vertical hydraulic conductivity [L/T], \( H_t \) is the initial height [L], \( H_i \) is the specimen’s current height at a given time [L], \( \gamma_w \) is the pore fluid specific weight [M/T\(^3\)L\(^2\)], and \( P_p \) is the measured excess pore pressure at the undrained bottom of the specimen [M/T\(^3\)L].
Because of the small strain assumption, Wissa et al. (1971) proposed and entered the initial height of the specimen in the hydraulic conductivity relationship and considered that it remains constant during the test. The modified for large strain equation gives the best results when the height of the sample changes significantly during the experiment (Adams, 2011; Gonzalez, 2000). The ASTM (2006) has adopted this definition for measurement of the hydraulic conductivity in fine-grained specimens such as the specimens in this study.

By inserting hydraulic conductivity (K) of equation (9) into the following expression, vertical permeability can be calculated:

$$k_v = \frac{K_d}{\rho g L^2}$$

where $k$ is the vertical permeability [$L^2$], $\mu$ is the dynamic viscosity [M/TL], $\rho$ is the fluid density [M/L^3], and $g$ is the gravity [L/T^2]. Knowing that

$$\gamma_v = \rho_v \mu_v$$

By substituting (12) and (9) into the expression (11) and then simplifying the expression, vertical permeability ($k_v$) can be directly computed as

$$k_v = \frac{\mu \gamma_v L^2}{2P}$$

Direct permeability measurement

To measure absolute permeability, we performed steady-state flow-through experiments at a constant pressure gradient condition and calculated vertical permeability using Darcy’s law:

$$k = \frac{\mu g L}{A \Delta \rho}$$

where $\mu$ is the flow rate [$L^2/T$], $\Delta \rho$ is the pressure difference between the inlet (upstream) and the outlet (downstream) [M/TL^2], $L$ and $A$ are the length [L] and cross-sectional area [$L^2$] of the specimen, respectively.

A schematic of the experimental setup for single-phase flow measurements is shown in Fig. 10b. The experiments were performed under controlled axial stress and pore pressure. While the oedometer cell was placed on the hydraulic load frame, a computer-controlled screw pump (GDS pressure-volume controller) regulated the vertical stress over the oedometer’s top piston using the load frame. Another computer-controlled GDS pump injected the brine at a constant pressure condition via the bottom compartment of the oedometer (inlet). The pore pressure sensor monitored the lower pore pressure continuously during the permeability test. The top drainage opening (outlet) was connected to a graduated pipette at atmospheric pressure ($P_{outlet} = 0.101$ MPa) to collect and measure the displaced pore fluid. To begin with the permeability measurement, the inlet pressure was adjusted to the desired pressure using the inlet GDS controller, and then the inlet valve was opened for flow. Although reaching steady-state flow requires prolonged testing time, the flow rates were only considered for evaluation of vertical permeability once the steady-state flow conditions were reached.

Relative permeability measurements

We conducted unsteady-state flow technique to derive two-phase relative permeability of brine-oil system. The unsteady-state flooding test uses an injection of single fluid phase during the drainage and imbibition stages. So the saturation changes and steady state is never achieved during the experiment. The pressure gradient and produced fluids are monitored and recorded as a function of time, and the corresponding relative permeability curves are extracted. The experiment was conducted using the described setup in Fig. 10b.

To determine drainage relative permeability curves, brine pore fluid was displaced by injecting oil (polysiloxane fluid, SF1147 Momentive) from the bottom inlet. While the specimen was subjected to 25 MPa vertical stress, the drainage phase started by flooding oil at 12.35 MPa pressure and keeping the outlet at atmospheric pressure ($P_{outlet} = 0.101$ MPa). The dynamic viscosity of brine and oil at experiment’s pressure and temperature were considered 1.06 and 60 cP, respectively. Although production of brine pore fluid was stopped after injecting 0.5 pore volumes (PVs) following the oil breakthrough, nonetheless, oil injection continued for more than 2 PVs after the breakthrough. The imbibition phase was accomplished by injecting brine with the same pressure profile as the drainage stage. During the imbibition cycle, the oil discharge came to an end after around 2 PVs injection of brine following the brine breakthrough. However, the injection resumed to more than 5 PVs after the brine breakthrough. Subsequently, after-breakthrough immiscible fluid displacement data were interpreted using the methodology suggested by Toth et al. (2002) to derive the relative permeability curves. The resulting relative permeability curves consist of a plot of the relative permeability to the brine-oil system as a function of the sample-averaged brine saturation.

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