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Self-sealing of argillaceous media in the context of geological disposal of radioactive waste – a perspective from past and ongoing studies



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Abstract: A review of discontinuity self-sealing mechanisms in argillaceous media is presented. Argillaceous rocks cover a range of siliceous materials generally defined by their particle size (<0.063 mm), composition, and the quantity and type of the clay minerals present. These properties have a strong influence on their material, rock mass and hydrogeological behaviour. In the context of geological disposal of radioactive waste in England and Wales currently (2024), the Mercia Mudstone Group and Ancholme Group are being considered as potential host rocks. These rocks are potentially suitable for the isolation and containment of radioactive waste due to their low primary permeability and potential for discontinuities to self-seal, thereby reducing secondary permeability. The ability of discontinuities to self-seal is a function of lithology, mineralogy, strength, stiffness, groundwater, groundwater chemistry and the *in situ* state of stress. These properties are strongly affected by depositional provenance, geological history and depth of burial. The processes and mechanisms that lead to self-sealing include swelling, creep, shear, slaking and precipitation of minerals. Self-sealing, therefore, is an important factor influencing the suitability of argillaceous media as a potential host rock for geological disposal of radioactive waste as part of the multi-barrier system.

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The UK has been generating radioactive waste from various sources, including power generation, industry, medicine and defence, for more than 60 years (Nuclear Waste Services 2022). There is international consensus that geological disposal is the safest permanent solution to manage the most hazardous radioactive waste (Nuclear Waste Services 2018). Various rock types are being considered as potential host rocks by waste management organizations, with argillaceous rocks being considered as potential host rocks by waste management organizations that include Andra (Callovo-Oxfordian Argillite) and Nagra (Opalinus Clay: Zhang and Talandier 2023), NIRAS/ONDRAF (Boom Clay, Kortrijk and Tielt formations: Turner et al. 2023), and the BGE (molasse and claystones: BGE 2024). Research being carried out at the University of Leeds is considering the geomechanical and hydrogeological properties of Mercia Mudstone Group and Ancholme Group rocks, specifically their ability to self-seal.

Self-sealing was defined by Tsang and Bernier (2005) as 'the reduction of fracture permeability (transmissivity) by any hydromechanical, hydrochemical or hydrobiochemical processes'. Self-sealing in argillaceous media is an important mechanism for many engineering applications, including disposal of radioactive waste (NEA 2010; Zhang and Talandier 2023), mining (Nagra 2020) and hydrocarbon seals (Fisher *et al.* 2021). The coupled thermal–hydromechanical–chemical (THMC) behaviour of argillaceous media, which includes self-sealing processes, has long been an important factor in determining their suitability as a potential host rock for the geological disposal of radioactive waste, along with very low material permeability, transport predominantly by diffusion, a strong retention capacity and continuity of deposits (Norris 2019). Comprehensive reviews of self-sealing have previously been

published (e.g. Horseman 2001; NEA 2010); however, there is a need for a short summary of the current 'state of knowledge' of self-sealing behaviour from the perspective of rock types of potential interest in the UK's ongoing Geological Disposal Facility (GDF) siting programme. In this paper, we attempt to progress delivery of this requirement – we intend this paper to provide an introduction to self-sealing concepts, coupled behaviour (including the influence of the engineering) and rock properties, and to act as a signpost for further reading.

This review paper initially provides a summary of the formation and classification of argillaceous media, and discontinuities, as well as the development of our understanding of how fluids and gasses flow through rocks. Later sections discuss self-sealing processes and mechanisms; present a short synopsis of the Mercia Mudstone Group and Ancholme Group rocks; and present preliminary experimental self-sealing data for Mercia Mudstone Group rocks. We do not present self-sealing data for the Ancholme Group as this experimental work is still ongoing. Finally, we discuss the importance of self-sealing mechanisms, how they interact (coupled behaviour), how they change over time, differences between the Mercia Mudstone Group and Ancholme Group, and the importance of the evolution of the excavation damage zone (EDZ).

Argillaceous media

A wide variety of argillaceous media from soft, plastic and poorly indurated clays to brittle, hard mudstones or shales are being considered as potential host rocks for the geological disposal of radioactive waste. The rock material properties refer to the intact rock between discontinuities. A discontinuity is a generic term for any

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mechanical break in the rock material with little or no tensile strength across the break and can be of natural or anthropogenic origin.

Material

The term 'argillaceous media' is used to describe a range of siliceous soils and rocks composed of predominantly silt- and clay-sized particles. There are several systems that can be used to classify these materials based on grain size, proportion of grains of specific sizes, state of lithification or metamorphism, and sedimentary structure (Ingram 1953; Hallsworth and Cox 1999; Horseman *et al.* 2005; NEA 2010; BSI 2018, 2020). There is no universally accepted classification scheme for these materials, which is further complicated by stratigraphic unit names: for example, the Oxford Clay, which is geomechanically a rock in its fresh state. For the purposes of this paper, the broadly accepted classification scheme presented in Table 1 has been adopted.

It should be noted that term 'mudstone' is absent from Table 1 as it is often used as a higher-order descriptor for argillaceous material regardless of grain size. This is inconsistent with engineering practice that uses the term 'mudstone' to describe lithified material where more than than 50% of grains are less than 2 μm , and the term 'claystone' that refers to nodular concretions commonly found in argillaceous media (Norbury 2016). As such, where referenced material uses inconsistent terminology we will clarify this in parentheses.

The simplified geological and stress history of argillaceous rocks comprises phases and processes of deposition, burial, diagenesis, induration and degradation, starting with the deposition of predominantly silt- and clay-sized fragments in aeolian, fluvial, lacustrine or marine environments (Cripps and Taylor 1981; Jeans and Merriman 2006). These sediments are buried and compacted, causing water to be squeezed out of the formation and the alignment of grains to be normal to the overburden pressure; the alignment is accentuated by clay minerals platy morphology. Post-depositional mechanical, chemical and mineralogical changes in the material such as cementing and illitization can occur, collectively referred to as diagenesis at low temperatures (<200°C) (Jeans and Merriman 2006). Post-deposition processes such as those associated with plate tectonics can lead to a realignment of mineral grains or the 'overprinting' of a fabric onto the rock (e.g. slate). The final stage in the geological and stress evolution of argillaceous media under consideration for a GDF, and often the reason why the materials are within the depth range of most engineering projects (i.e. in the shallow subsurface), is uplift, which causes destressing of the rock by unloading, erosion and weathering.

In terms of their uniaxial compressive strength (UCS) (and other geotechnical properties), argillaceous media have characteristics that range from soils to rocks (Cripps and Taylor 1981). There has been debate concerning what UCS value should be used to determine the boundary between very high strength soils and extremely weak rocks (ISRM 1978; BSI 2018). Various UCS values

Table 1. Classification of argillaceous media

Characteristic	More than 50% of grains between 2 and 63 μm	More than 50% of grains less than 2 μm
Unlithified	Silt	Clay
Lithified	Siltstone	Claystone
Fissile	Shale	
Lightly metamorphosed	Argillite	
Moderately metamorphosed	Slate	

Source: Ingram (1953); Hallsworth and Cox (1999); Horseman *et al.* (2005); NEA (2010); BSI (2018, 2020).

have been adopted as the boundary between soil and rock, ranging between 0.25 MPa (ISRM 1978) and 0.6 MPa (BSI 2018). For the purpose of this paper, we would suggest a value of 0.6 MPa be adopted as the boundary between soil and rock but, following the convention set out in BSI (2018, 2020), descriptions should be based on the anticipated engineering behaviour.

Moisture content has a significant impact on rock strength; Lu et al. (2017) and Liu et al. (2021) presented data that showed a 76 and 61% reduction in strength when the water content was increased by between 2 and 6%, respectively. Turner et al. (2023) noted that argillaceous media with a UCS normal to bedding that ranges from less than 1 to c. 30 MPa are being considered as host rocks for several European GDF programmes. Groundwater and porewater chemistry can also influence behaviour, specifically salinity that can increase strength (Geng et al. 2022).

Clay mineralogy

The quantity and type of the clay minerals exert a strong influence on the material, mass and hydrogeological behaviour of argillaceous media. Jeans and Merriman (2006) stated that clay minerals are predominantly phyllosilicates (or sheet silicates) formed by direct precipitation from solution or via crystallization from glassy volcanic ash (neoformed), by chemical weathering of existing minerals, or 'transformed' during diagenesis and/or low-grade metamorphism. Clay minerals are often transported to sedimentary basins with little modification (inherited) (Jeans and Merriman 2006). Clay minerals are composed of three basic structures (building blocks) arranged in sheets (illite, smectite and chlorite), chains (palygorskite and sepiolite) or amorphously (allophane) (Kumari and Mohan 2021). The basic classification of clays is based on the ratio of silicon to oxygen tetrahedral layers and octahedral cation-oxygen hydroxide layers (see Table 2). A significant property of clay minerals is their ability to absorb and expel water, leading to shrink-swell behaviour.

Discontinuities

A discontinuity refers to any mechanical break in a rock mass with zero or low tensile strength. Discontinuity is the collective term for most types of fractures, joints, bedding planes, schistosity planes, weak rock material zones and faults (ISRM 1978), as outlined in Table 3.

Discontinuities form and develop through a variety of processes relating to burial, extrusion, orogenesis, loading, unloading and weathering (Hudson and Harrison 2000; Palmström and Stille 2015; Hencher 2014). These processes result in different states of stress, leading to deformation or fracture formation, or both. Repeated cycles of stress change may result in multiple phases of deformation and fracturing, with the impact of anthropogenic activity also resulting in the formation or modification of fracture systems (Hencher 2014) (see Table 4).

Discontinuities formed by changes in stress occur when the forces of compression, tension or shear exceed the tensile or shear strength of the rock (material). Fracture initiation commences at microscopic weaknesses and heterogeneities in the rock (e.g. at grain boundaries) when the stress becomes greater than the strength. Continued application of stress leads to coalescence into microfractures and eventually macro-discontinuities (Hudson and Harrison 2000).

The mechanical behaviour of discontinuities is governed by the mechanical properties of the rock, the stress conditions and the nature of the discontinuities (Palmström and Stille 2015). Discontinuities can be described in terms of orientation, spacing, persistence, aperture, roughness, wall strength, infilling and water (ISRM 1978; BSI 2018). It is common practice to describe the

Table 2. Clay minerals

Group	Structure	Exchange capacity (meq/100 g)	Surface area (m ² g ⁻¹)	Swelling potential
Kaolinite	1:1 Al:Si	3–15	10–70	Low
Illite Mica Smectite Vermiculite	$2:1$ Si : Al ($\pm Mg,Fe,K$) : Si \pm interlayer ions, water, and hydrated and unhydrated cations	10–15 5–20 80–140 100–150	80–100 50–200 800 750	Low-medium Low-medium High High
Chlorite	2:1:1 Si: Al (±Mg,Fe,K): Si: gibbsite or brucite	20	10–55	Low

Source: Battey (1986); Moore (1991); Barnes (2000); Horseman et al. (2005); Kumari and Mohan (2021).

Table 3. Classification of discontinuities

Туре	Discontinuity description
Fracture and joint	A break of the geological origin in the continuity of a body of rock along which there has been no visible displacement
Fissure	Fissures are synonymous with joints but are predominantly applied to soils or extremely weak rocks
Fault	A fracture or joint along which there has been recognizable displacement
Shear	A fissure along which there has been recognizable displacement
Bedding	A break along the bedding (bedding is a surface parallel to the plane of deposition)
Cleavage	A break along a set of parallel planes of weakness often associated with mineral realignment
Induced	A discontinuity of non-geological origin (e.g. brought about by coring, core handling, blasting and excavation)
Incipient	A discontinuity that retains some tensile strength, which might not be fully developed or which might be partially or fully cemented

Source: ISRM (1978); BSI (2020).

Table 4. Discontinuity-forming processes

Process	Description
Burial stress	Stress increases with depth due to the weight of the overburden
Fluid and thermal stress	Increases in fluid pressures can initiate fracture formation (hydrofracture). Burial and/or contact with a heat source (i.e. intrusive igneous rocks) can impose a thermal stress that can cause discontinuity formation, as well as induce pore pressures due to the thermal expansion of water
Tectonics	On a regional scale, vertical stress can be quite modest when compared to high horizontal stresses associated with tectonic plate movements, particularly at convergent and transform plate margins
Uplift and erosion	Uplift and erosion lead to the relief of overburden stresses; the effect of this de-stressing on geomechanical behaviour can lead to discontinuity formation, the degree of influence depending on the lithology, geological and stress history.
Deglaciation	Similar to uplift and erosion, deglaciation can produce discontinuities (Horseman <i>et al.</i> 2005); in addition, glacial and periglacial processes and climates, specifically the influx of glacial melt water and periglacial freeze—thaw processes, can lead to an increase in groundwater pressures and hydraulically initiated fracture formation (quarrying) (Iverson 1991)
Weathering	Weathering processes that can lead to discontinuity development include, wetting (swelling of clay minerals), drying (contraction crack formation) and freeze-thaw processes (water expansion/jacking)
Anthropogenic	Discontinuities can be caused deliberately or as a by-product of many anthropogenic activities such as drilling, blasting, excavation, heating and fluid/gas injection
Other processes	Induced stresses caused by chemical reactions and or physical changes in the rock can induce discontinuities. Desiccation is the drying of materials in the context of fine-grained sedimentary rocks; as water is removed from rocks by evaporation, the material reduces in volume (shrinks). The reduction in volume is due to negative porewater pressure overcoming the tensile strength of the material and, as such, the rock cracks often in a distinctive polygonal form (NB: this is a localized effect rather than penetrative persistent discontinuity creation)

Source: Hudson and Harrison (2000); Hencher and Richards (2015); Palmström and Stille (2015).

mechanical behaviour of discontinuities in terms of shear strength (cohesion, friction and other parameters) using failure criteria such as Barton (1976) and rock masses using various failure criterion such as Mohr–Coulomb and Hoek–Brown (Hencher and Richards 2015).

Friction along discontinuities can be considered on many scales such as first-order field-scale 'waviness' and second-order laboratory-scale roughness composed of discrete peaks and ridges of various sizes that are collectively known as asperities (Wyllie and Mah 2004). The amplitude and geometry of such asperities influence the strength, stiffness and permeability of the discontinuity (Ficker and Martišek 2016; Zhang and Chai 2020; Barton *et al.* 2023). True cohesion occurs when there are intact rock bridges across a discontinuity (e.g. from the precipitation of minerals in an

open discontinuity) or in incipient discontinuities. Apparent cohesion is an artefact of testing associated with joint roughness, interlocking of asperities and the condition of joint walls as discontinuities with no tensile strength do not have any true cohesion (Barton 2013; Rullière *et al.* 2020).

Self-sealing mechanisms

Background

Discontinuity formation and development can increase secondary permeability. Laboratory tests have shown that discontinuities in argillaceous media can self-seal, reducing secondary permeability to primary permeability values (NEA 2010; Fisher *et al.* 2013; Zhang and Talandier 2023). Research has been carried out into self-sealing and the self-sealing behaviour of argillaceous media for many decades by nuclear waste disposal organizations in countries including Belgium (ONDRAF/NIRAS), France (Andra), Switzerland (Nagra), Germany (GRS), Japan (NUMO) and Canada (NWMO) (NEA 2010; Bernier *et al.* 2011; Delay *et al.* 2014; Delay 2019; Bossart 2021).

The processes and mechanisms that lead to self-sealing are well understood and have been demonstrated by several authors including Labiouse *et al.* (2009), Lanyon *et al.* (2014), De La Vaissière *et al.* (2015), Harrington *et al.* (2017) and Wang *et al.* (2024). A comprehensive summary is presented in NEA (2010). Horseman *et al.* (2005) identified around 25 self-sealing mechanisms; arguably the most significant self-sealing mechanisms include swelling, creep, shear, changes in stress, slaking and precipitation (see Fig. 1).

Calculating flow on a discontinuity

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Conceptually, discontinuity (fracture) flow is based on Darcy's law, which assumes steady-state (non-turbulent) flow of a single-phase incompressible fluid through a porous media (Fisher *et al.* 2023), see equation (1):

$$Q = -KA \frac{\mathrm{d}h}{\mathrm{d}l} \tag{1}$$

where Q is the flow (m³ s⁻¹), K represents the hydraulic conductivity (m s⁻¹), A is the cross-sectional area (m²) and dh/dl represents the dimensionless hydraulic gradient in the direction of decreasing head. Using the Darcy equation as a basis, flow on fractures can be calculated for simplified conditions of flow between two parallel discontinuity walls, of uniform aperture, and assuming

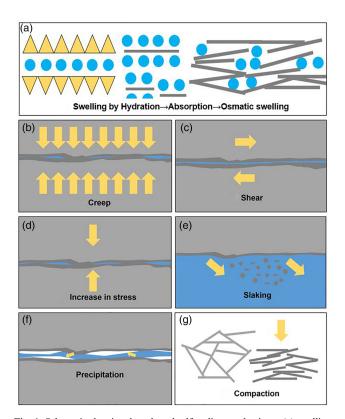


Fig. 1. Schematic showing the selected self-sealing mechanisms: (a) swelling by hydration→absorption→osmatic swelling; (b) creep; (c) shear; (d) increase in stress; (e) slaking; (f) precipitation; and (g) compaction.

that flow is laminar (i.e. the fracture aperture and flow velocities are small) (Hiscock 2005), see equation (2):

$$Q_{\rm f} = -2bwK_{\rm f}\frac{{\rm d}h}{{\rm d}l} \tag{2}$$

where $Q_{\rm f}$ is the flow through a fracture (m³ s⁻¹), $K_{\rm f}$ represents the hydraulic conductivity of the fracture (m s⁻¹), 2b is the discontinuity aperture (m), w is the discontinuity width (m) and dh/dl represents the dimensionless hydraulic gradient in the direction of decreasing head. The fracture hydraulic conductivity can then be calculated (Hiscock 2005), see equation (3):

$$K_{\rm f} = \frac{\rho g(2b)^2}{12\mu} \tag{3}$$

where $K_{\rm f}$ is the hydraulic conductivity of a fracture (m s⁻¹), ρ is the fluid density (kg m⁻³), g is gravitational acceleration (m s⁻²), 2b is the discontinuity aperture (m) and μ is the fluid viscosity (kg m⁻¹ s⁻¹).

Self-sealing causes fracture closure, which reduces flow; the reduction in flow is described by the 'cubic law': that is, flow is proportional to the cube of aperture, and the flow rate is directly proportional to the pressure gradient, fracture aperture³/12 (Hiscock 2005; Zimmerman 2012).

Self-sealing by the swelling of clay minerals

Water can have a significant impact on the behaviour of argillaceous media. A change in the moisture content can result in a volume change (shrinkage or swelling); Cripps and Czerewko (2017) presented data that indicated axial swelling strain of up to 7.8% for UK mudstones (cf. argillaceous media). However, experiments carried out by Alzamel *et al.* (2022) demonstrated swelling strains of in excess of 300% for bentonite–sand mixtures with a 70:30 ratio, with distilled water and a density of 2 g cm⁻³. The extent of the volume change depends on the volume and composition of the fluid, as well as type and quantity of clay minerals present. Swelling is an important process as it can close discontinuities, leading to self-sealing (Horseman *et al.* 2005; Tsang and Bernier 2005; NEA 2010)

Water can be associated with clay minerals in the following ways (NEA 2010):

- free water in pore spaces between clay minerals;
- capillary water on the edges of clay minerals (cf. the meniscus of water on the edge of a container, sometimes referred to as loosely adsorbed);
- adsorbed or absorbed water on the surface layers of clay minerals (sometimes referred to as strongly adsorbed); and
- crystalline water held interlayer as part of the clay mineral structure.

Clay minerals, a significant component of argillaceous media, are composed of tetrahedral (one silica ion surrounded by four oxygen ions) and octahedral (one aluminium/magnesium/iron/other ion surrounded by six oxygen/hydroxyl ions) layers (Murray 2007). Although the tetrahedral layers are neutral, the octahedral layers are negatively charged, which makes them susceptible to substitution of, for example, Al³⁺ for Si⁴⁺ and Mg²⁺ for Al³⁺, (Barnes 2000), as well as susceptible to the adsorption of water, which results in swelling. Swelling is the ability to adsorb water between the layers, resulting in strong repulsive forces and clay expansion (Hensen and Smit 2002).

Smectite group minerals, such as montmorillonite, are particularly susceptible to swelling due to their layer structure and interlayer bonding. These swelling clay minerals have very large surface areas: for example, smectite has a surface area of $c.\ 800\ {\rm m}^2\ {\rm g}^{-1}$ compared to illite, which has a surface area of

between 80 and 100 m² g⁻¹, and thus has a relatively greater capacity for water adsorption (Barnes 2000). In addition, swelling clay minerals have high cation-exchange capacity (the capacity of the material to hold exchangeable cations): for example, smectite has a base exchange capacity of 80–140 meq/100 g compared to illite, which has a base exchange capacity of 10–15 meq/100 g, and thus the cations and water can be more readily exchanged (Barnes 2000; Reeves *et al.* 2006; Butscher *et al.* 2016).

Based on the structure, swelling in clays occurs in two principle ways (Ohazuruike and Lee 2023). First, osmotic swelling occurs as water is attracted to clay minerals to balance charges in the material, the movement is driven by 'flow' along cation concentration gradients to the clay minerals until an equilibrium is reached. Secondly, crystalline swelling occurs with hydration on the external particle interfaces and in the interlayer spaces, and the adsorbed water results in increased interlayer spacing. Horseman *et al.* (2005) refers to these processes as mechanical and physio-chemical swelling mechanisms. Pore and groundwater chemistry can influence the behaviour of clay minerals and is discussed below.

Self-sealing by creep

Creep is the time-dependent distortion of a material at a constant volume, under a constant state of stress that is less than the short-term strength of the material (Paraskevopoulou 2016). This makes creep a significant process for self-sealing in argillaceous media given the low strength of the rock and the potential state of stress around underground excavations (Horseman *et al.* 2005; NEA 2010; Herrmann *et al.* 2020). Creep is influenced by the mineralogy (strength of the ionic bonds), clay content, temperature and state of stress (Fabre and Pellet 2006). Paraskevopoulou (2016) and Fisher *et al.* (2021) noted five micro-mechanical processes that result in creep:

- Diffusion: movement or diffusion of atoms on vacancies, imperfections or impurities in the crystal lattice.
- Dislocation glide: weak bonds on mineral surfaces facilitate
 gliding or slipping perpendicular to stress. Translation
 occurs on discrete crystallographic planes of weakness,
 defects or inclusions, and propagates through the material.
 The main influence on this behaviour is the strength of ionic
 bonding and temperature. This behaviour is also referred to
 as grain-boundary sliding.
- Dislocation creep: grain-scale plasticity at high temperatures
 with propagation along favourable crystallographic planes;
 recrystallization (growth of strain-free new grains) prevents
 'pile ups' from occurring. This repeating cycle of behaviour
 can accommodate high levels of deformation (superplasticity). The main influence on this behaviour is the strength of
 ionic bonding and temperature.
- Pressure solution: a stress-driven chemical corrosion process at high-stress grain boundaries and subsequent aqueous solution precipitated at lower-stress grain boundaries. The main influence on this behaviour is stress and the presence of fluids.
- Microfracture and cataclasis: fracturing occurs as brittle
 deformation at a grain/crystal scale; when the microfracturing becomes more pervasive it can coalesce into
 bands of cataclastic flow (frictional sliding on interconnected microcracks), leading to grain-scale reduction. The
 main influence on this behaviour is stress and the presence of
 fluids.

Creep mechanisms in argillaceous media include dislocation glide and creep facilitated by phyllosilicate orientation, 'bending' of weak materials, shearing of clay flakes, grain sliding and rotation (dislocation, aggregate deformation and granular creep), crack growth, stress corrosion, and cataclastic creep (Fabre and Pellet 2006; Sone and Zoback 2013). Creep behaviour shows three distinct phases:

- primary creep: characterized by elastic (reversible), decelerating strain rate due to strain-hardening (Paraskevopoulou 2016):
- secondary creep: elasto-plastic (partially reversible), quasiconstant strain. In brittle rocks, secondary creep is rare and the rocks typically transition from primary creep straight through into tertiary creep; however, Fabre and Pellet (2006) carried out tests in the Oxfordian argillite and the Tournemire argillite that clearly showed the existence of a secondary creep phase; and
- tertiary creep: characterized by plastic (irreversible), accelerating strain related to brittle yield micro-crack/fracture mechanics and evolution (Innocente et al. 2021).

These descriptions are for an idealized creep condition, and the duration and magnitude of each stage can vary (or be absent) depending on the lithology, mineralogy and hydrogeological regime.

Self-sealing by shearing

Self-sealing through shearing results in the closure of a discontinuity by the removal of asperities (discontinuity surface roughness) and/or the creation of fault gouge (cataclasite) that infills the discontinuity, which can make the fracture functionally impermeable (Fisher *et al.* 2021; Pirzada *et al.* 2023). The ability of an argillaceous media to self-seal through shear is dependent upon the shear strength of the discontinuities, the magnitude of the *in situ* stress field and the orientation of the discontinuities in relation to the stress field.

The shear strength of a discontinuity is the resistance of two surfaces to shear stress, and is a function of the mobilized friction and cohesion. For shear to occur, the shear strength of the discontinuity needs to be exceeded (Hudson and Harrison 2000). The shear strength of discontinuities, in terms of the linear relationship with normal stress, can be calculated using a reduced version of the Mohr–Coulomb failure criteria (Hudson and Harrison 2000), see equation (4):

$$\tau = \sigma_{\rm n} \tan \left(\phi \right) \tag{4}$$

where τ is the shear strength, σ_n is the normal stress and ϕ is the friction angle. Patton (1966) recognized the importance of roughness on joint surfaces and adapted the Mohr–Coulomb failure criteria to include a term for the basic friction angle plus an asperity angle to account for joint roughness (Hudson and Harrison 2000), see equation (5):

$$\tau = \sigma_{\rm n} \tan \left(\phi_{\rm b} + i \right) \tag{5}$$

where τ is the shear strength, σ_n is the normal stress, ϕ_b is the basic friction angle and i is the angle of asperity (sawtooth) in the direction of movement. The addition of the term for the angle of asperity introduces the concept of dilation. At low normal stresses, displacement can cause discontinuity surfaces to open as discontinuity surfaces ride over each other. If the shear loading continues, the discontinuity surfaces can become damaged and asperities are sheared off, resulting in a basic friction angle of the discontinuity surface.

Patton's work was further refined by Barton (1976), who highlighted that the mechanical performance of a discontinuity is influenced by the roughness of the surface (joint roughness coefficient and the compressive strength of the discontinuity

surfaces (joint (wall) compressive strength), see equation (6):

6

$$au = \sigma_{\rm n} \tan \left({
m JRC} \, \log_{10} \left({
m \frac{JCS}{\sigma_{\rm n}}} \right) + \phi_{\rm b}
ight)$$
 (6)

where τ is the shear strength, σ_n is the normal stress, JRC is the joint roughness coefficient, JCS is the joint (wall) compressive strength and ϕ_b is the basic friction angle. Barton and Choubey (1977) later replaced the basic friction angle with the residual friction angle. In the context of argillaceous media, self-sealing through shear is strongly influenced by weak wall-rock materials and low discontinuity roughness, which are common in these materials (Cripps and Czerewko 2017).

The influence of rock type on friction reduces with increasing *in situ* stresses. At low stresses found in most engineering works, the influence of friction can be significant. However, in higher stress regimes, the types of rock have no significant effect on friction (Byerlee 1978). Self-sealing through shear is dominated by the lithology and strength of asperities, and the stress acting on the discontinuity surface. NEA (2010) presented a table showing the influence on fracture wall roughness on various self-sealing mechanisms (including shearing), which is partially reproduced in Table 5.

The shear force required to cause displacement is dependent upon a number of factors that include the scale, geometry and distribution of asperities, the strength of the asperities, the initial stress regime, and the redistribution of stresses after initial asperity failure (Barton and Choubey 1977; Lei 2003). Laboratory tests show that, with favourable conditions, asperities can shear at displacements on the scale of millimetres or centimetres commensurate with, or less than, the displacements that might be expected during underground construction or during natural geological processes such as faulting (Barton and Bandis 1982; Haggert *et al.* 1992; Pereira and De Freitas 1993; Yang *et al.* 2010).

There have been a number of case studies presented in literature that discuss self-sealing by shearing; Pereira and De Freitas (1993) presented data which show that the displacements required to dilate discontinuities are measured in millimetres or less. They concluded that asperities were destroyed predominantly by abrasion and wearing at lower normal stresses, by brittle failure and by the subsequent comminution of the resulting gouge at higher normal stresses. Shear box tests tend to indicate that residual friction is mobilized at displacements of the order of 5–10 mm. The nature and extent of gouge produced by the shearing of asperities may impact on the self-sealing potential of the discontinuity.

The formation of clay gouges is a function of shear forces, displacement, *in situ* stresses and mineralogy. Data from the hydrocarbon industry have shown that sealing clay gouges form in faults at relatively small displacements even when claystone or shale is a minor constituent (Holland *et al.* 2006). Fisher *et al.* (2013) described fault gouge formation by mechanical fragmentation that created a more porous gouge but, with increasing displacement, a pure clay gouge formed with a lower permeability, which resulted in sealing. Clay smear and cataclasis are both active mechanical processes in fault zones that can result in the sealing of faults, thereby reducing the permeability of these potential pathways to the point where they become hydrocarbon reservoir seals. The primary difference being the lithological composition of the faulted rock

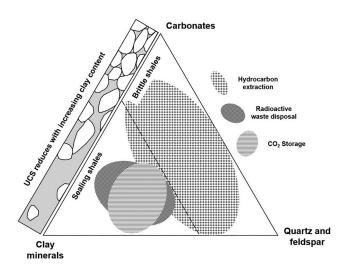


Fig. 2. Ternary diagram showing the relationship between mineralogy, strength and geomechanical behaviour. Source: simplified from Bourg (2015).

being either quartz-rich cataclasites developed from pure sandstones or phyllosilicate (clay) smears developed from shales (Knipe et al. 1997).

Experiments carried out by Bourg (2015) on fractured shale indicate that minor amounts of shearing and crushing can result in a six orders of magnitude decrease in fracture permeability if the effective normal stress is greater than the UCS of the rock (see Fig. 2). Given sufficient displacement and loads, asperities on discontinuity surfaces at potential repository depths could crush to the size of their constituent grain sizes and smear along the planes of movement leading to self-sealing (Fisher *et al.* 2013).

The heat produced by thermogenic wastes in a GDF could cause thermal overclosure due to prior higher stress or temperature in rougher discontinuities (Barton 2020), or the potential opening of smoother discontinuities that allow other self-sealing processes to become more dominant (Barton 2007).

Self-sealing by change in normal stress

Sealing of discontinuities can be achieved by an increase in normal effective stress either by a reduction in the fluid (or gas) pressure or an increase in normal stress (Horseman *et al.* 2005; Ishii 2021). Bandis (1980) in Barton *et al.* (2023) noted that the maximum closure of an open discontinuity was approximately the same as the initial aperture, and the initial aperture was determined by the JCS, which could in turn be correlated to the JRC. As such, the larger the initial aperture and the weaker the JCS, the larger the maximum closure. The ratio of JCS and initial aperture is a useful indicator of behaviour, with low JCS/initial aperture ratios giving larger values of maximum closure.

Although discontinuities in argillaceous media are relatively smooth, the load on a discontinuity surface can still be borne by a few asperities and the strength of these point contacts must be overcome before closure can occur (Cripps and Czerewko 2017). Failure of these asperities under increasing normal stress can be achieved by crushing, plastic deformation and tensile failure

Table 5. Influence of the fracture wall roughness on selected self-sealing mechanisms

Morphology	Swelling	Creep	Shear	Change in stress	Slaking
Smooth	High	Medium	Medium	High	Low
Rough	High	Medium	high	Medium	Medium

Source: NEA (2010).

(Pereira and De Freitas 1993). If the discontinuity is saturated, the increase in available clay minerals for the water to react with can weaken the asperities and the wall rock.

Another way of considering self-sealing by changes in stress is discussed in Horseman (2001) and in Cuss *et al.* (2012), who suggested that critical state concepts could be used to describe argillaceous rock behaviour. The original critical state model was developed for soils, and careful application of this method is required to account for features in rocks that influence behaviour such as lithification and cementing. The critical state concept defines material behaviour in terms of mean effective stress (p'), deviatoric stress (q') and specific volume (v). This allows the definition of stress states where the material will either show dilation, consolidation or shear.

Cuss et al. (2012) presented data in p'-q' space that showed on the dry side of the critical state line (the Hvorslev surface), deformation leads to a reduction in porosity, including along fractures, leading to a reduction in permeability and self-sealing (permeability will reach a minimum when it reaches the critical state). On the wet side of the critical state line (the Roscoe surface), shear leads initially to dilation (and an increase in permeability), and eventually to a reduction in permeability through the processes of compaction/consolidation and cataclasis. It should be noted that the critical state model assumes uniform plastic deformation of the whole sample; however, rocks shear along a plane or thin zone, with the material in the shear zone reducing to a residual shear strength and the material outside of this zone less affected and not in a critical state (Cuss et al. 2012). The additional complication noted by Chui and Johnston (1984) is the onset of dilatancy during shear that results in higher void ratios if the material shears rather than just the discontinuity.

Self-sealing by slaking

Repeated cycles of wetting and drying lead to swelling and shrinkage, which in turn can lead to the disaggregation of clay minerals and movement away from the discontinuity wall in a process known as slaking (Horseman *et al.* 2005; NEA 2010). Moriwaki (1975) suggested the following slaking mechanisms:

- Body slaking: an internal process by which a specimen will rapidly disaggregate into connected 'blocks' (to give the appearance of paving or brick work) with no apparent deterioration of the material between cracks.
- Surface slaking: loss of rock material through the loss of individual particles or groups of particles without causing cracking in the underlying material.
- Dispersion: clay-sized fragments lost from the rock by moving into suspension in the groundwater.

Slaking causes self-sealing by smoothing out fracture surfaces and allowing the closure of discontinuities as surface roughness is removed and redeposited elsewhere, potentially blocking flow paths (NEA 2010).

Self-sealing by precipitation

Precipitation of minerals along discontinuity surfaces is a well-understood mechanism for self-sealing and is an important self-sealing mechanism in higher strength rocks (Hakami and Winell 2022). However, unlike higher strength rocks where precipitation is strongly associated with the circulation of mineral-rich geothermal fluids, the properties of argillaceous media inhibit the circulation of groundwater (Fisher *et al.* 2013). NEA (2010) noted that most discontinuity infill, such as celestite veins in the Callovo-Oxfordian argillites, have been interpreted as local early diagenetic features, although there is some evidence for the precipitation of minerals from palaeoseawater as it percolated through the rock over geological timescales.

Self-sealing by precipitation in argillaceous media in an EDZ is likely to be caused by a change in environmental conditions such as drying of the excavation walls that can either drive groundwater towards the free surface or through the introduction of moisture/humidity from the tunnel atmosphere (Valès *et al.* 2004; Millard *et al.* 2017). Both of these processes can lead to the precipitation of minerals through evaporation or oxidation: for example, the oxidation of pyrite that can in turn lead to expansion and chemical attack of concrete, and which can be problematic for tunnel lining systems (Cripps and Czerewko 2017).

Potential argillaceous media host rock in England and Wales

Background

Sedimentary rocks are very common globally, covering c. 75% of the land surface, with clays (including shales) forming more than 50% of the Earth's sedimentary rocks (Reeves et al. 2006). In the UK, argillaceous rocks are widely distributed geographically at the surface or beneath younger materials and within the influence of engineering projects. Argillaceous rocks are also distributed throughout the stratigraphic record from the Precambrian through to the present day (the older rocks are generally more indurated, stronger and stiffer, some have been metamorphosed to slate, phyllite and schist) (Jeans and Merriman 2006; Reeves et al. 2006; Cripps and Czerewko 2017). Figure 3 shows the presence of lower strength sedimentary rocks within the 200-1000 m depth interval of interest in England and Wales shown out to the 12 nautical mile limit (the limit of UK territorial sea), superimposed on a digital elevation model (Turner et al. 2023). Lower strength sedimentary rocks is a term used to describe fine-grained, sedimentary rocks with a clay high content that are low permeability and are mechanically weak, so that open fractures cannot be sustained (Radioactive Waste Management 2016). Lower strength sedimentary rocks include argillaceous media but not all lower strength sedimentary rocks will be suitable for hosting a GDF.

This section discusses two examples of argillaceous rocks from the UK: Mercia Mudstone Group rocks and rocks of the Ancholme Group. These rocks are currently rock types of interest in the ongoing UK government project to site a GDF for the UK's most hazardous radioactive waste (more on this initiative can be found in BEIS 2018).

Mercia Mudstone Group

The Mercia Mudstone Group is Triassic in age (Ansian-latest Rhaetian, c. 247–201 Ma) (Howard et al. 2008; Cohen et al. 2013). Argillaceous media of the Mercia Mudstone Group were deposited in a series of fault-bounded basins in the interior of Pangaea, north of the Variscan mountain belt (Hobbs et al. 2002; Howard et al. 2008). At the time the UK was at a similar geographical location to northern Africa (20°-30° N) with a monsoonal climate, resulting in high-intensity precipitation events followed by extended periods of aridity (Hobbs et al. 2002; Jeans and Merriman 2006). Fluvial deposition of the underlying Sherwood Sandstone Group gave way to subaqueous hypersaline and evaporitic mudflat environments, leading to four main depositional processes (Howard et al. 2008): settling of clay- and silt-sized particles in saline water bodies; sheetwash deposition of silt and sand during flash floods; aeolian deposition of predominantly fine sand and silt- and clay-sized particles on wet mudflats; and precipitation of evaporites such as halite and gypsum from water bodies and groundwater.

Post-deposition, the rocks were buried to a depth of between 2000 and 2500 m in the East Irish Sea Basin, for example, and were heated to c. 80°C (Jackson and Mulholland 1993; Holford et al.

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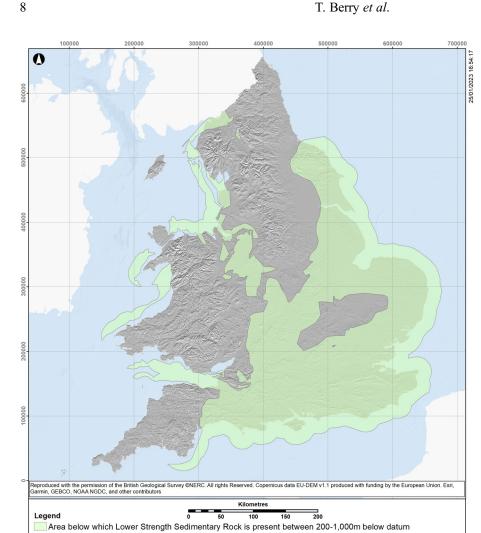


Fig. 3. Distribution of lower strength sedimentary rocks within the 200-1000 m depth intervals of interest in England and Wales, superimposed on a digital elevation model. Source: Turner et al. (2023).

2009; Armitage et al. 2016). Post-burial exhumation during the Cenozoic Era and erosion of up to 3 km of post-Early Jurassic overburden in the East Irish Sea Basin and its margins resulted in Mercia Mudstone Group rocks subcropping beneath Quaternary deposits (Armitage et al. 2016; Holford et al. 2009).

Onshore, the Mercia Mudstone Group was deposited in eight basins from the Wessex Basin in the south through to the East Midlands and NE England basins in the NE, and the West Lancashire and Carlisle basins in the NW (Hobbs et al. 2002). The Mercia Mudstone Group has been divided into the following five lithostratigraphic units (A–E) that are mappable onshore both at the surface and in the subsurface, with their East Irish Sea Basin equivalents (Howard et al. 2008):

- A. Tarporley Siltstone Formation: the East Irish Sea Basin equivalent units include the Ormskirk Sandstone Formation and part of the Leyland Formation (including the Rossall Halite Member, where present).
- B. Sidmouth Mudstone Formation: the East Irish Sea Basin equivalents include the Leyland Formation (including the Mythop Halite Member), the Preesall Halite Formation, the Dowbridge Mudstone Formation and part of the Warton Halite Formation.
- C. Arden Sandstone Formation: the East Irish Sea Basin equivalent is part of the Warton Halite Formation.
- D. Branscombe Mudstone Formation: the Elswick Mudstone Formation in the East Irish Sea Basin is contemporaneous with the Branscombe Mudstone Formation of the East Midlands Shelf.

E. Blue Anchor Formation: the East Irish Sea Basin equivalent is the Blue Anchor Formation.

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The Mercia Mudstone Group is generally composed of brown, redbrown, calcareous clays and mudstones (cf. claystones), with subordinate, sometimes impersistent, beds of siltstone, fine-grained sandstone and evaporites predominantly composed of halite and gypsum (Hobbs et al. 2002; Howard et al. 2008). Clay minerals common in the Mercia Mudstone Group include detrital mica, smectite, kaolinite, illite and chlorite, as well as authigenic (forming



Fig. 4. Branscombe Mudstone Formation of the Mercia Mudstone Group, Bantycock Mine: Source: reproduced with the permission of Saint-Gobain.



Fig. 5. Peterborough Member, Oxford Clay, Formation, Ancholme Group, Must Farm Quarry. Source: reproduced with the permission of Forterra.

in situ) mixed-layer smectite—chlorite and smectite—illite (Hobbs *et al.* 2002; Jeans and Merriman 2006). In its unweathered state the mudstone (cf. argillaceous media) is jointed and 'weak', with strong subordinate sandstone beds (Hobbs *et al.* 2002). An example of evaporite-rich argillaceous rocks of the Mercia Mudstone Group is presented in Figure 4.

Ancholme Group

The Ancholme Group refers to Jurassic-age rocks (Callovian–Kimmeridgian, c. 166–152 Ma) that developed between Norfolk and Humberside (Woods et al. 2022). The Ancholme Group was deposited in a marine environment within the East Midlands Shelf when the UK was in a similar geographical location to the Mediterranean Sea (30°–40° N) with a warm and humid climate (Barron et al. 2012).

Green *et al.* (2018) suggests that Jurassic deposits reached their maximum burial depth of c. 1700 m around the end of the Mesozoic. Post-burial exhumation of c. 800 m during the Cenozoic Era included a secondary shorter phase of reburial and reexhumation. Grant *et al.* (2021) attributed the uplift to a far-field effect of the Alpine Orogeny and the opening of the Atlantic Ocean.

The Ancholme Group comprises the following formations onshore and offshore the coast of Lincolnshire (Woods et al. 2022):

- · Kellaways Formation;
- Oxford Clay Formation;
- · West Walton Formation;

- · Ampthill Clay Formation; and
- Kimmeridge Clay Formation.

The surface expression of the Ancholme Group has been mapped in a north–south-orientated wedge between Hull in the north (where it is c. 2.5 km wide) and between Sleaford and west of Skegness (where it is c. 40 km wide). The Ancholme Group occurs extensively beneath younger soils and rocks beneath Lincolnshire, Norfolk and the North Sea.

The Ancholme Group is described by the British Geological Survey (BGS 2024) as predominantly grey, marine mudstone (cf. claystone) and silty mudstone (cf. claystone) with subordinate beds of argillaceous limestone nodules locally forming impersistent beds. Beds of siltstone and sandstone are widespread at the base of the Kellaways Formation, within the West Walton Formation and locally in the Kimmeridge Clay Formation. Shelly marl, limestone and sandy limestone are also developed locally (notably in the West Walton Formation). Clay minerals, common in Ancholme Group rocks, include detrital mica, illite, kaolin, smectite—mica—vermiculite and rarely chlorite. Marine authigenic clay assemblages include glauconite and smectite (Jeans and Merriman 2006). An example of Ancholme Group rocks is presented in Figure 5.

Preliminary self-sealing results for the Mercia Mudstone Group

Following previous self-sealing studies in argillaceous media (summarized in NEA 2010; and presented in Labiouse et al. 2009; Cuss and Harrington 2010; de Haller et al. 2014; Lanyon et al. 2014; Bossart et al. 2019; Di Donna et al. 2019; Giot et al. 2019; Ziegler et al. 2022; Zhang and Talandier 2023; Wang et al. 2024), a programme of laboratory testing was carried out at the University of Leeds. Laboratory experimentation included classification tests, quantitative X-ray diffraction (QXRD), geomechanical tests and measurements of fracture hydraulic conductivity. The following samples of Mercia Mudstone Group material were collected from the following locations (see Fig. 6): Sid Mudstone Member (of the Sidmouth Mudstone Formation), Jacobs Ladder Beach, west of Sidmouth (block samples); Branscombe Mudstone Formation (Blue Rock Muck), Bantycock Mine (operated by Saint-Gobain), SE of Newark-upon-Trent (block samples); and undifferentiated mudstone, Teesside (borehole core samples).

The sites were selected as they provided material representative of different depositional basins and stratigraphic levels in the Mercia

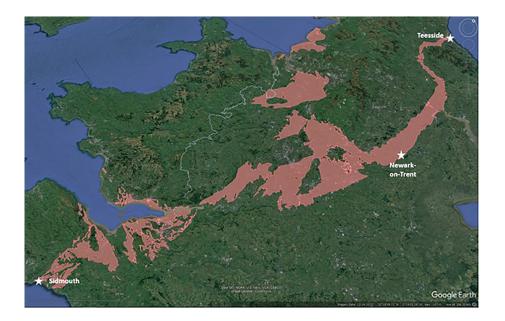


Fig. 6. Sampling locations for the Mercia Mudstone Group. Source: the British Geological Survey onshore geology 1:625 000-scale DiGMapGB625 layer for TRIA-MDSS (Triassic Rocks (undifferentiated) mudstone, siltstone and sandstone) © Google Earth, 'Contains British Geological Survey materials © UKRI 2024'.

Table 6. Preliminary laboratory test results for the Mercia Mudstone Group expressed as an average

Specimen location	Moisture content (%)	Slake durability index (I _{d2}) (%)	Point load test (MPa)
Sid Mudstone Member	5.8	55	0.8
Branscombe Mudstone Formation	2.5	39	1.2
Undivided (Teesside)	11.9	58	1.2

Mudstone Group sequence. Sampling was carried out in accordance with the guidance set out in Ulusay (2007) and BSI (2018, 2020) to deliver Category B samples, allowing definitive hydrogeological testing and non-definitive strength determinations. The specimens tested represented the finer parts of the Mercia Mudstone Group, and were described as moderately weak reddish brown, slightly fine, sandy-silty mudstone (sandy-silty claystone).

Experiments were carried out to determine the moisture content, slake durability and point load strength in accordance with the relevant standards or suggested methods (Franklin *et al.* 1979; ISRM 1979; Franklin 1985). The QXRD determinations were carried out according to the methods set out in Hillier (2000).

The preliminary results are presented in Table 6 and Figure 7.

Self-sealing tests were carried out on 50 mm-diameter, c. 50 mmlong cylindrical samples. To create the 'discontinuity', the specimens were sawed longitudinally down the centre of the core using a lapidary saw. Before installing in the triaxial cell, the discontinuity was propped open with 0.5 mm-thick spacers and glued back together. The self-sealing test was carried out in a Wykeham Farrance Tritech 50 soil triaxial rig using a 200 kPa cell pressure to ensure the water flowed through the saw-cut discontinuity. The experiment consisted of passing tap water through the propped discontinuity under a small hydraulic pressure equivalent to 1 m head of water to minimize the potential for erosion. Self-sealing was measured by recording the time taken for a volume of water to flow through the discontinuity. The fracture hydraulic conductivity was calculated using the equations presented in the 'Calculating flow on a discontinuity' subsection earlier in this paper. The results of the self-sealing experiments are presented in Figure 8.

The classification tests indicated lower moisture contents for the specimens taken from block samples than those taken from cores. This was expected as the block samples were taken from either recently exposed or quarried blocks that may have dried out. The results of the slake durability tests indicate material with a low slake durability index (Id2) and the point load test results indicate that the material is at the weaker end of the medium strong strength range.

The QXRD test results were compared with published results. Hobbs *et al.* (2002) noted that the main non-clay minerals present in the Mercia Mudstone Group include quartz, calcium and

magnesium carbonates, calcium sulfates, micas, iron oxides, halite, and feldspar; the major clay minerals are illite, chlorite, mixed-layer illite–smectite, chlorite–smectite and, less commonly, smectite. The QXRD results indicate that the composition of the material tested are consistent with the published data. The results show the dominant potential swelling clay mineral (smectite) is bound with illite (a non-swelling mineral), indicating a potential for swelling.

The results of the flow tests indicate that self-sealing is occurring in the following stages:

- Early (c. 0–24 h): initially there is a minor response to the water flow that consists of a slight reduction in the fracture hydraulic conductivity and, occasionally, even increasing (indicating a washing out of detritus in the test zone).
- Middle (c. 24–96 h): a rapid reduction in the fracture hydraulic conductivity in the middle portion of the test.
- Late (c. >96 h): the latter stages of the test indicate a slowing down in the reduction of the fracture hydraulic conductivity to a lower steady-state level with fluctuations of around one order of magnitude.

The test results so far have shown a very distinctive fracture hydraulic conductivity profile over time, specifically the sudden reduction in the fracture hydraulic conductivity that we have described as 'blooming'. Blooming tends to start c. 24 h after the start of the test and then stops c. 96 h after the start of the test. It is thought that the blooming effect could be a function of the following processes:

• The delayed response could be due to the time required for the clay mineral swelling pressures to overcome the tensile strength of the grain boundaries or cementing. Rapid fracture initiation and propagation perpendicular and parallel to the discontinuity surface lead to an increase in the volume of the fracture surface material, reducing the fracture aperture and the fracture hydraulic conductivity. In addition, the newly fractured material creates new exposures of the clay minerals for the flow fluid to react with and leads to additional swelling. Cessation of the swelling occurs when there are no

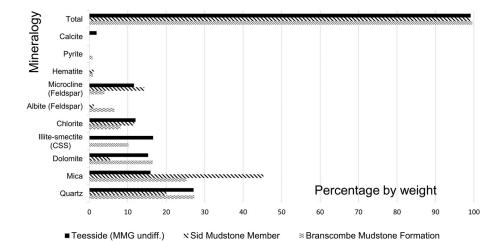


Fig. 7. Results of the QXRD testing.

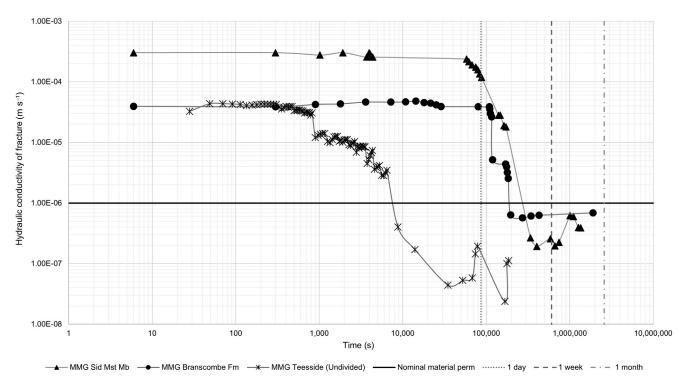


Fig. 8. Log-log plot of the hydraulic conductivity of a single sawcut discontinuity using tap water carried out in a triaxial cell.

- more available clay minerals for the water to react with and the specimen reaches a quasi-stable steady state.
- The delayed (time-dependent) response could be due to the time required to initiate the swelling mechanisms: that is, initially adsorption on the surfaces of clay minerals (Illitesmectite) and later osmotic swelling as water is incorporated between clay minerals and 'clay plugs' are formed, as described by Auvray et al. (2015) and Giot et al. (2019).

Experimental work is currently being carried out to investigate selfsealing of selected materials from the Ancholme Group; however, this experimental work is ongoing and, as such, cannot be presented at this time.

Discussion

Factors influencing self-sealing in argillaceous media

The nature and extent of self-sealing is controlled by a number of parameters that interact with each other (coupled), including the nature of the argillaceous media (material), the nature, orientation and extent of discontinuities (rock mass), the hydrogeological regime (groundwater and geochemistry), the geological history, the current state of *in situ* stress (stress regime) and the effect of the engineering (changes in stress) (see Fig. 9).

Bourg (2015) (see Fig. 2) has shown how the quantity of clay is linked to the shale strength and sealing behaviour: that is, more than 40% clay minerals generally results in 'sealing shales' in hydrocarbon reservoir cap rocks. The strength and stiffness of an argillaceous media is related to a number of factors, including the bulk composition, clay mineralogy, cementing (Armand *et al.* 2017), stress history (Hudson and Harrison 2000) and moisture content of the argillaceous media (Liu *et al.* 2021). Higher strength and stiffness inhibits the ability of argillaceous media to deform in a ductile manner, shear and, ultimately, self-seal. Reference groundwater and porewater compositions for argillaceous media in England and Wales have been proposed by Smedley *et al.* (2022). Their results suggest that porewater and groundwater in the Mercia Mudstone Group (e.g. in the East Irish Sea Basin) will be

substantially more saline than seawater, and porewater and groundwater in argillaceous media of the Ancholme Group will have a similar salinity to seawater. Salinity impedes the swelling of clay minerals, thereby reducing their ability to self-seal by inhibiting the entrance of water into the clay mineral structure and reducing the hydration of the clay minerals (de Carvalho Balaban *et al.* 2015). Although swelling can be inhibited by saline groundwater, it does not stop swelling. Results of experiments carried out and reported in Savage (2005) demonstrate that the functional swelling pressure requirements of engineered barrier systems will be met even with a groundwater salinity equivalent to 3 M NaCl (and assuming that the density of the argillaceous media is 1.9 Mg m⁻³ or greater).

The dominance of a particular self-sealing mechanism is strongly dependent on the influencing external parameters, the nature of the rock and the stress regime. As such, several self-sealing processes can be operating at any time around an excavation based on the geometry and the orientation of the excavation relative to the *in situ*



Fig. 9. Factors influencing the self-sealing processes.

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stress. In high stress regimes (e.g. as experienced in the petroleum industry), it may be that a change in stress will be the dominant process, whereas in a lower stress environment, with the development of tensile fractures, the short-term increase in fracture hydraulic conductivity may allow temporary flow such that swelling could be the dominant self-sealing process. For example, Armand et al. (2013, 2017) present figures showings the nature and distribution of discontinuities during the construction of the d'Expérimentation Trois (GET) drift in the Meuse Haute-Marne Underground Research Laboratory in France. The results show that both shearing and tensile fracturing occur at the same time at different locations around the excavation, implying that both selfsealing by shearing and potentially self-sealing by swelling could be contemporaneous. These self-sealing mechanisms also evolve over time, and active processes will change in relative importance as each process affects, and is affected by, the initiation and progress of other self-sealing processes (Parsons 2020; Birkholzer and Bond 2022).

It is accepted that there are many self-sealing mechanisms (Horseman et al. 2005; NEA 2010), and the nature and evolution of an EDZ could potentially allow different self-sealing mechanisms to operate contemporaneously and interact (Armand et al. 2013, 2017). The interactions of these self-sealing processes (coupled behaviour) are under-represented in the literature and there are uncertainties around how one self-sealing process could inhibit, stop or induce another self-sealing process. For example, the displacement required to shear the material to a self-sealed state may not be necessary as other sealing processes might become dominant: for example, as asperities potentially override each other, temporarily increasing the fracture hydraulic conductivity, and allowing water ingress and swelling. The effectiveness and significance of various self-sealing processes are also influenced by other coupled thermal-hydrogeological-mechanical-chemical processes. For example, an increase in temperature from the emplacement of heat-generating waste may change the geomechanical properties of the rock and the hydrogeological properties of fluids to create thermo-hydraulic gradients driving water flow, change the viscosity of water and potentially change the properties of clay minerals (Gens et al. 2011; Parsons 2020; Woodman 2020) (see Fig. 10). Significant research on this subject has been carried out by researchers and practitioners working on the DECOVALEX project (DEvelopment of COupled models and their VALidation against EXperiments) (https://decovalex.org/).

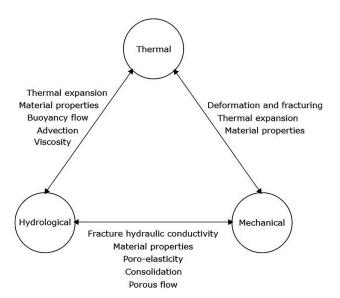


Fig. 10. Illustrative summary of the THM coupled processes. Source: Parsons (2020).

The geosphere as part of a multi-barrier system

The disposal of radioactive waste relies on a multi-barrier containment system broadly comprising the waste containers, engineered barriers in a GDF and the geosphere itself, all working together to provide suitable conditions for long-term isolation and containment (Turner *et al.* 2023). Packaging and engineered barrier systems will be optimized once the site for a disposal facility has been investigated, as the nature of the deep geological environment, including the chemistry of deep groundwater systems, significantly influences the materials and designs that will be used in the engineered barriers (Turner *et al.* 2023).

The properties that make argillaceous media a potentially effective barrier in the geosphere are the same properties that make clay (bentonite, bentonite cement and bentonite clay) a favourable material for engineered barrier systems within a disposal facility, including its self-sealing capacity (Norris 2017). There are three main ways that clay can be used in an engineered barrier system (Norris 2019):

- Buffer material: clay placed between the waste package and the lining system or host rock.
- Backfill material: clay placed in space created underground not used for disposal (i.e. access ways and shafts).
- Sealing or plugging material: clay placed to isolate sections of a disposal facility (e.g. when emplacement is complete).

Finally, the geosphere will provide a significant impediment to the migration of radioactive material and acts as the final barrier in the multi-barrier containment system. In addition to their favourable material properties, argillaceous media, such as those associated with the Ancholme Group, can provide a host rock that is laterally extensive and uniform over many kilometres and vertically homogeneous on the scale of decametres (Turner *et al.* 2023). However, construction of space underground causes disturbances in the stress field, leading to the formation of an EDZ, a limited and temporally impersistent zone of fracturing confined to a few metres around the excavation (Armand *et al.* 2013, 2017) (see Fig. 11).

Discontinuities can form around excavations (including boreholes) on multiple scales, from a microscopic scale to a decametre scale. This disruption can result in an EDZ immediately around the excavation where the hydromechanical and geochemical modifications of the rock can induce significant changes in flow and transport properties (Tsang and Bernier 2005). The EDZ can contain new discontinuities that allow the movement, and reopening, of pre-existing discontinuities which can act as temporary pathways around the disposal facility and enhance the secondary permeability of the rock. The properties of these discontinuities could change through time, affected, for example, by self-sealing mechanisms.

The self-sealing potential of the Mercia Mudstone Group and Ancholme Group argillaceous media

The primary differences between argillaceous media of the Mercia Mudstone Group and the Ancholme Group are their lithology and mineralogy (influenced by their depositional environment), and their geological and stress history. The secondary differences between the groups (i.e. strength, permeability and self-sealing potential) arise from these primary factors. A high-level summary of selected parameters is presented in Table 7. The result of the variation in grain size of the argillaceous particles is that the Mercia Mudstone Group is often referred to as a mudstone (being composed of a mixture of clay- and silt-sized particles) and the Ancholme Group is referred to as a claystone.

The geological and stress history of argillaceous media can have a significant impact on behaviour. For example, the Mercia Mudstone

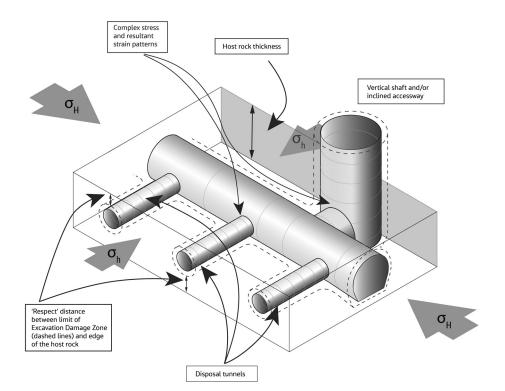


Fig. 11. Schematic perspective view of shafts and tunnels and their associated EDZ. $\sigma_{\rm H}$ = arrows indicate the orientation of the maximum horizontal stress. $\sigma_{\rm h}$ = arrows indicate the orientation of the minimum horizontal stress. Source: Turner *et al.* (2023).

Group in the East Irish Sea Basin was buried to depths of *c*. 2000–2500 m before exhumation. These processes have induced changes that will affect behaviour, such as the crystallization of dolomite and anhydrite, gypsum–anhydrite–gypsum reactions, the formation of carbonate cements, and changes in clay mineral assemblages (i.e. chlorite formation or illitization of smectite) (Hobbs *et al.* 2002; Armitage *et al.* 2016; Ohazuruike and Lee 2023). The Ancholme Group, however, was buried to shallower depths, up to 1700 m, and for a shorter period of time, so the effects of burial on the geomechanical behaviour of these rocks could be similar but are likely to be less significant (Green *et al.* 2018).

Rocks of the Mercia Mudstone Group were deposited in a relatively dynamic mudflat environment in temporary lakes, flash-flood sheet washes and by wind. In addition, thick sequences of halite occur in basins between Somerset and Cheshire (Hobbs *et al.* 2002). As such, the Mercia Mudstone Group is heterogeneous (Turner *et al.* 2023), and argillaceous sequences can include reprecipitation of evaporites in discontinuities, subordinate laminae to beds of silt and fine sand, as well as persistent decimetre- to metre-scale sandstone beds sometimes referred to as 'skerries' (Hobbs *et al.* 2002) (see Fig. 4). Hobbs *et al.* (2002) reported that the

onshore Mercia Mudstone Group is predominantly composed of quartz, carbonates (calcium and magnesium), sulfates, micas, iron oxides and halite, with clay minerals that include illite, chlorite, mixed-clay minerals (illite–smectite and chlorite–smectite) and, occasionally, smectite. This lithological heterogeneity results in variations in strength, discontinuity formation, permeability (horizontal and vertical) and the ability to resist stress, all of which are factors influencing self-sealing behaviour (see Fig. 9).

By comparison, the Ancholme Group was deposited in a lowenergy, shallow-marine environment, and there has been a longheld view that these deposits are more consistent both in their lateral persistence (Woods *et al.* 2022) and their internal composition (Hudson and Martill 1994; Turner *et al.* 2023). Although the lateral persistence and consistency of the stratigraphic sequence off the Lincolnshire coast have been presented in British Geological Survey maps and reports (BGS 1990; Woods *et al.* 2022), some authors (e.g. Wignall 1989; Woods *et al.* 2023) suggest that significant heterogeneity, in terms of engineering properties, exists within argillaceous media of the Ancholme Group due to storm deposits and variations in the calcium carbonate and organic content (see Fig. 5). Norry *et al.* (1994) reported that the Peterborough

Table 7. Summary of selected properties for argillaceous media of the Mercia Mudstone Group and Ancholme Group

Parameter	Mercia Mudstone Group	Ancholme Group
Age (Ma)	247–201	166–152
Deposition	Mudflat – lacustrine, sheet wash and aeolian	Shallow marine
Composition	Claystone with siltstone and evaporites	Claystone, silty and locally calcareous
Thickness (m)	330–1485	240–340
Porosity (%)	7–12	22–55
Hydraulic conductivity (m s ⁻¹)	$10^{-11} - 10^{-9}$	$1.09 \times 10^{-10} - 1.07 \times 10^{-9}$ *
Salinity	Hypersaline	Saline
Total clay (wt%)	10–40	45–49*
Density (Mg m ⁻³)	2.3–2.5	1.7–2.1
UCS (⊥bedding) (MPa)	0.2–30	0.2–6*

*Oxford Clay Formation.

Source: Horseman (2001); Smedley et al. (2022); Woods et al. (2022); Turner et al. (2023); Woods et al. (2023); BGS (2024).

Member of the Oxford Clay Formation is composed largely of quartz and illite, with minor chlorite, kaolinite, potassium-feldspar and plagioclase, with calcite and siderite. Again, this lithological heterogeneity results in variations in strength, discontinuity formation, permeability (horizontal and vertical) and ability to resist stress, all of which are factors influencing self-sealing behaviour (see Fig. 9).

The variation in lithology (and age) results in argillaceous media of the Mercia Mudstone Group often being stronger and stiffer than those of the Ancholme Group, influencing the magnitude and rapidity of self-sealing processes such as changes in stress and shearing. Groundwater and porewater chemistry is also quite different between the Mercia Mudstone Group and the Ancholme Group rocks, which will impact swelling self-sealing mechanisms.

European analogues for potential host rocks in England and Wales

European waste management organizations have been carrying out laboratory and *in situ* characterization of argillaceous media, including self-sealing experiments, for geological radioactive disposal for many decades (Bernier *et al.* 2011; Bossart *et al.* 2017; Delay 2019). The argillaceous media under consideration to host a GDF include the Middle–Upper Jurassic Callovo-Oxfordian argillite (France) and the Lower–Middle Jurassic Opalinus Clay (Switzerland). In addition, extensive research has been carried out on other argillaceous media to understand the wider geomechanical and hydrogeological behaviour of these materials: for example, the Keuper Marl, Tertiary Molasse and Jurassic Opalinuston Formation (Germany), and the Oligocene Boom Clay (Belgium).

The Callovo-Oxfordian argillite investigated by Andra is similar in age to the Oxford Clay in England, and both were deposited within the same basin and are broadly similar (Schofield *et al.* 2014). There are some differences in the depositional environment between the two units as they were deposited at different sides of the basin. The Callovo-Oxfordian is likely to have been deposited in a deeper-water environment than the shallower depositional environment of the Oxford Clay, leading to the Oxford Clay being sandier, siltier and more carbonate rich than the Callovo-Oxfordian argillite (Horseman *et al.* 1984). The similarity in age, depositional environment and lithology means that there is some value in considering Callovo-Oxfordian data analogous to the Oxford Clay.

Significant self-sealing experiments have been carried out on samples of Callovo-Oxfordian argillite in laboratories and *in situ* at the Meuse Haute-Marne Underground Research Laboratory in France (Davy *et al.* 2007; De La Vaissière *et al.* 2014; Di Donna *et al.* 2019; Giot *et al.* 2019; Zhang and Talandier 2023) that demonstrated self-sealing as an active, effective and reliable process. Despite the geographical and depositional differences, there is sufficient justification to use evidence of self-sealing in the Callovo-Oxfordian argillite as an analogue for the Oxford Clay in England for early stage (desk study phase) engineering geological ground models, and conceptual engineering design, in lieu of site-specific data.

The German waste management organization, the Bundesgesellschaft für Endlagerung (BGE), is investigating various rocks to host a GDF, including rock salt, claystone and crystalline rock (Schafmeister 2023; BGE 2024). Although the German GDF siting programme is not specifically considering the Keuper Marl, which is the approximate equivalent of the Mercia Mudstone Group (Hobbs et al. 2002; Meschede and Warr 2019), it has been of historical interest in both German and Swiss GDF siting studies. As such, there is potential to learn from the BGE and other organizations, and to leverage their knowledge and experience of the Keuper Marl into the UK GDF programme (e.g. see Seidel et al. 2024).

Summary

A review has been carried out to summarize the composition and nature of argillaceous media (material and discontinuities), self-sealing processes and potential relevant rock types currently (2024) being considered in the UK to host a GDF.

There are many self-sealing mechanisms of various significance, the most important from the perspective of geological disposal are swelling, creep, shear, changes in stress, slaking and precipitation. Self-sealing has been observed as an active and effective process in laboratory experiments, *in situ* experiments, in engineering practice, and in the natural geological environment (NEA 2010; Fisher *et al.* 2013; Nagra 2020). Self-sealing has been observed at various scales from clay particle level, through engineering scales, up to geological scales, and has been shown to operate at different rates and over different time periods from just a few hours to millions of years (Fisher *et al.* 2013; Giot *et al.* 2019; Zhang and Talandier 2023).

Excavations to create space underground cause perturbations in the stress field that could lead to the creation, activation and reactivation of discontinuities at multiple scales forming an EDZ and potentially creating pathways. The EDZ will evolve, and self-sealing by various processes is possible at these times that may seal the discontinuities. There are a number of significant factors that influence self-sealing in argillaceous media such as the lithological composition, clay mineralogy, strength, stiffness, porewater and groundwater chemistry, and *in situ* stress. The clay mineral-rich, weak nature of argillaceous media make them particularly well suited to self-sealing behaviour, particularly at the moderate *in situ* stress regimes associated with the potential depths of geological disposal facilities: for example, *c*. 500 m in France and 800 m in Switzerland.

Significant work has been carried out globally to investigate self-sealing of argillaceous media, as such self-sealing mechanisms are a well-understood phenomenon in certain geological environments. Although the fundamental questions concerning 'if' and 'how' self-sealing will occur have been substantially answered (NEA 2010), there are clear hydro-geomechanical gaps in knowledge around the rate and magnitude of self-sealing of the Mercia Mudstone Group and Ancholme Group. As such, further research is required to close these information gaps.

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