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## Full Length Article

# Mechanical and hydraulic properties of carbonate rock: The critical role of porosity

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## ABSTRACT

Carbonate rocks are extensively used in civil infrastructure and play a critical role in geoenergy geo-engineering, either as hydrocarbon reservoirs or potential repositories for CO<sub>2</sub> geological storage. Carbonate genesis and diagenetic overprint determine the properties of carbonate rocks. This study combines recent data gathered from Madison Limestone and an extensive dataset compiled from published sources to analyze the hydraulic and mechanical properties of limestone carbonate rocks. Physical models and data analyses recognize the inherently granular genesis of carbonate rocks and explain the strong dependency of physical properties on porosity. The asymptotically-correct power model in terms of  $(1-\phi/\phi^*)^\alpha$  is a good approximation to global trends of unconfined stiffness  $E$  and unconfined compressive strength  $UCS$ , cohesive intercept in Mohr-Coulomb failure envelopes, and the brittle-to-ductile transition stress. This power model is the analytical solution for the mechanical properties of percolating granular structures. We adopted a limiting granular porosity  $\phi^* = 0.5$  for all models, which was consistent with the loosest packing of monosize spheres. The fitted power model has exponent ( $\alpha = 2$ ) in agreement with percolation theory and highlights the sensitivity of mechanical properties to porosity. Data and models confirm a porosity-independent ratio between unconfined stiffness and strength, and the ratio follows a log-normal distribution with mean  $(E/UCS) \approx 300$ . The high angle of internal shear strength measured for carbonate rocks reflects delayed contact failure with increased confinement, and it is not sensitive to porosity. Permeability spans more than six orders of magnitude. Grain size controls pore size and determines the reference permeability  $k^*$  at the limiting porosity  $\phi^* = 0.5$ . For a given grain size from fine to coarse-grained dominant carbonates, permeability is very sensitive to changes in porosity, suggesting preferential changes in the internal pore network during compaction.

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## 1. Introduction

Carbonate bio-genesis and post-depositional compaction, crushing, dissolution, and remineralization combine to define the porosity, fabric and inherent heterogeneity of carbonate rocks. In turn, these micro-scale characteristics affect flow and all physico-mechanical properties including stiffness, strength and brittleness. Carbonate heterogeneity and variable engineering properties challenge the design and construction of civil infrastructure and lead to conservative approaches. For example, the capacity of

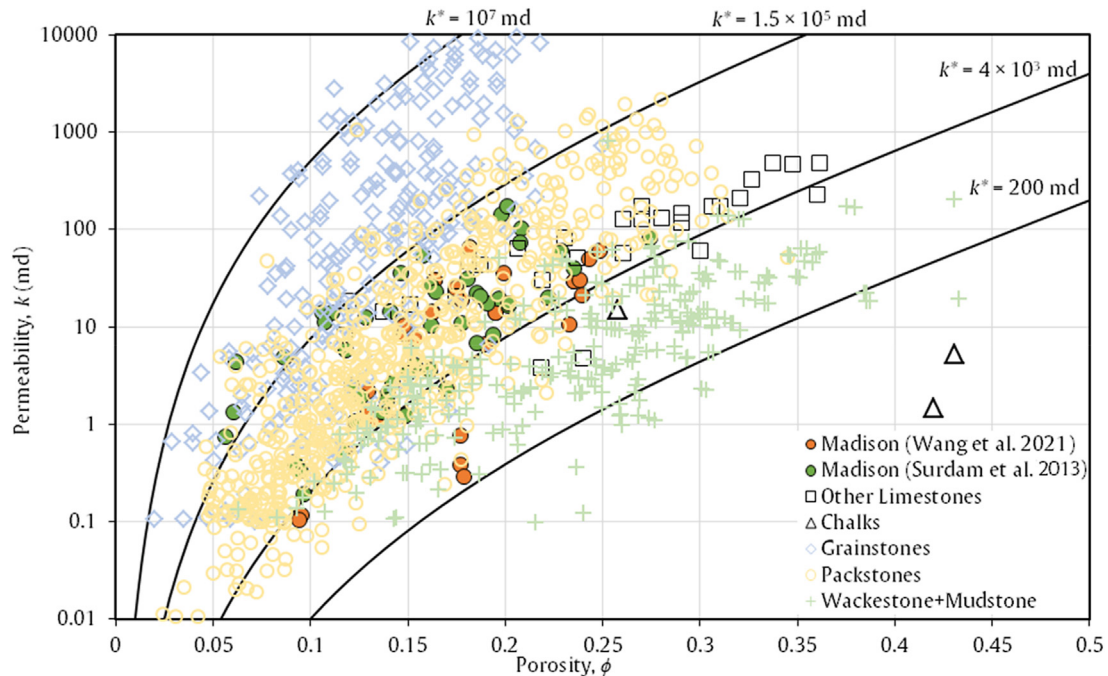
driven piles resting on soft carbonate rocks is usually under-predicted during the design stage, leading to large discrepancies between estimated and measured pile resistances (Ng et al., 2015; Ng and Sullivan, 2017).

Carbonates play an important role in energy geoengineering as well. In fact, carbonate reservoirs contain half of the world's proven oil reserves and contribute 60% of the total oil and gas production worldwide. The regimes in the Middle East (e.g. Saudi Arabia, Libya), Europe (e.g. Russia), Asian (e.g. Republic of Kazakhstan), and North America, host some of the largest carbonate reservoirs (Roehl and Choquette, 1985; Shepherd, 2009). Heterogeneity and associated uncertainty in mechanical and fluid flow properties affect drilling operations, reservoir modeling, production management, and optimization.

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**Fig. 1.** Permeability versus porosity. Data sources: databases in Lucia (1995), Lindsay et al. (2006), and Cardona and Santamarina (2020); new data from Fabre and Gustkiewicz (1997), Vanorio and Mavko (2011), Surdam et al. (2013), Wang (2017), Yu (2018), and Wang et al. (2021). Trends shown as continuous lines correspond to Eq. (1) for the same exponent  $\beta = 4.5$ ; the reference permeabilities  $k^*$  correspond to porosity  $\phi^* = 0.5$ .

More recently, porous carbonate rocks have been considered as candidate repositories for CO<sub>2</sub> geological storage. This is the case of the Rock Springs Uplift in southwest Wyoming, USA (Surdam and Jiao, 2007) where the 76-m thick high-porosity Mississippian Madison Limestone sits beneath the low-permeability Cretaceous shale seal. Spatial variability and heterogeneity were detected during the extensive characterization program of the Madison Formation (Surdam et al., 2013; Shafer, 2013; Wang, 2017; Yu et al., 2019; Ng et al., 2019; Wang et al., 2021). These characteristics must be properly considered in the design of drilling and injection operations to anticipate flow and storativity.

The study combines recent data gathered from Madison Limestone and an extensive dataset compiled from published sources for carbonates around the world. We emphasized porosity as a valuable index property to assess spatial variability and to obtain first-order estimates of material properties in order to guide field characterization and design strategies. Clearly, a more detailed analysis would require pore-scale information related to the geometry of pores and the nature of interparticle bonding. As porosity is systematically reported in the literature, it is available for all carbonates in our database.

We tried to improve our understanding and estimation of permeability, unconfined stiffness and strength, stress-dependent strength, and confining stress at the brittle-to-ductile transition. Observations and analyses recognize the inherent granular genesis of carbonates (applied to all carbonates, from grainstones to mudstones), and build on contact mechanics and percolation theory, rather than on pore-fracture analyses.

## 2. Porosity and permeability

Carbonate genesis and diagenetic overprint often produce complex multi-scale pore structures that can vary from the sub-micron scale intraparticle/interparticle/moldic pores to kilometer-scale cave systems (Saller et al., 1994; Mazzullo, 2004; Moh'd,

2009; Moore and Wade, 2013; Head and Vanorio, 2016). Interconnected interparticle pores govern permeability. Together, porosity and permeability determine injectibility, storativity and recovery (Dullien, 1992; Tiab and Donaldson, 2012).

Our database of porosity and permeability combines data gathered in our laboratories and reported in the literature including databases in Lucia (1995) and Lindsay et al. (2006). Fig. 1 shows that carbonate permeability spans more than six orders of magnitude. Given the highly variable interconnected porous network in carbonate rocks, correlations between porosity  $\phi$  and permeability  $k$  are weak.

Narrower estimates of permeability involve pore-scale information, such as pore size distribution (PSD) and pore geometry (Clerke, 2009; Buiting and Clerke, 2013). In fact, the permeability of carbonates depends on the largest interconnected pores, so it correlates best with the pore diameter that corresponds to the 80th percentile in pore-size distributions (Cardona and Santamarina, 2020).

Pore-size is a function of (a) grain size as shown in Fig. 1 where permeability is highest in grainstones and lowest in mudstones; and (b) packing density. In general, granular packings exhibit a linear relationship between permeability  $k$  and void ratio  $e = \phi/(1-\phi)$  in log-log scale, in agreement with models of fluid flow in porous media (see data and analyses in Ren and Santamarina (2017)). Mathematically, this implies:

$$\frac{k}{k^*} = \left(\frac{e}{e^*}\right)^\beta \Rightarrow k = k^* \left(\frac{\phi}{1-\phi}\right)^\beta \quad (1)$$

where  $k^*$  and  $e^*$  are reference values,  $\beta$  is the exponent value.

The second expression in Eq. (1) is obtained for a reference void ratio  $e^* = 1$  with a corresponding porosity  $\phi^* = e^*/(1+e^*) = 0.5$  that resembles the porosity of a simple cubic packing of monosized particles with  $\phi_{sc} = 0.476$ . The lines superimposed on Fig. 1 are computed with Eq. (1) for a typical exponent  $\beta = 4.5$ . It can be

**Table 1**  
Database of uniaxial compressive test results of dry carbonates compiled from published literature.

Carbonate	Location	Number of Tests	Percent calcite (%)	$\phi$	$\rho_b$ (g/cm <sup>3</sup> )	UCS (MPa)	$E$ (GPa)	$\nu$	Source
Madison	WY, USA	17	98	0.065–0.08 <sup>S</sup>	2.49–2.53	27–77	NA	NA	Surdam et al. (2013)
Indiana	IN, USA	4	99.6	0.153–0.17 <sup>#</sup>	2.23–2.26	44–59.3	29–29.6	0.18–0.2	Chitty et al. (1994); Prasad et al. (2009)
Fusselman	TX, USA	1	97	0.03 <sup>#</sup>	2.55	39	NA	NA	Handin and Hager (1957)
Wolfcamp	NM, USA	1	96	0.042 <sup>#</sup>	2.63	111	NA	NA	Handin and Hager (1957)
Carthage	MI, USA	1	99.9	0.017 <sup>#</sup>	2.62	89	71.6	NA	Prasad et al. (2009)
Bina	Israel	18	NA	0.06–0.218 <sup>#</sup>	2.19–2.63	15.4–187	10–60.45	0.15–0.4	Palchik (2010, 2011); Palchik and Hatzor (2000, 2002)
Nekarot	Israel	6	NA	0.076–0.104 <sup>#</sup>	2.42–2.49	141–184	44.4–49	0.23–0.28	Palchik and Hatzor (2002)
Yarka	Israel	4	NA	0.157–0.179 <sup>#</sup>	2.3–2.36	38.7–71	6.2–8.4	0.15–0.24	Palchik (2010, 2011)
Sakhnin	Israel	3	NA	0.071–0.132 <sup>#</sup>	2.43–2.6	89–144.6	40.7–57.3	NA	Palchik (2011)
Shmone	Israel	1	NA	0.057 <sup>#</sup>	2.64	172.4	54.4	0.28	Palchik (2011)
Yanuach	Israel	1	NA	0.161 <sup>#</sup>	2.35	35	35.4	0.32	Palchik (2011)
Solnhofen	Germany	4	97.9	0.017–0.055 <sup>&amp;#</sup>	2.08–2.73	277–310	NA	NA	Renner and Rummel (1996); Mogi (1967, 2007); Prasad et al. (2009)
Miocene	Hungary	39	NA	0.114–0.45 <sup>#</sup>	1.52–2.41	3.08–38.8	0.61–21.1	NA	Vasarhelyi (2005)
Devonian	Turkey	21	>91	0.0114–0.0412 <sup>+#</sup>	2.61–2.73	74.2–138.1	16.7–46.8	NA	Zarif and Tugrul (2003); Handin and Hager (1957)
Kirechane	Turkey	7	90–100	0.074–0.182 <sup>#</sup>	2.24–2.71	16.7–21.4	NA	NA	Ceryan et al. (2013)
Cebecikoy	Turkey	3	NA	0.021–0.023 <sup>#</sup>	2.35–2.42	34–38	NA	0.32–0.34	Kurtulus et al. (2016)
Sogucak	Turkey	3	NA	0.02–0.025 <sup>#</sup>	2.2–2.55	28–45	NA	0.29–0.38	Kurtulus et al. (2016)
Hereke	Turkey	8	NA	0.02–0.023 <sup>#</sup>	2.33–2.5	33–40	NA	0.29–0.35	Kurtulus et al. (2016)
Akveren	Turkey	10	NA	0.022–0.026 <sup>#</sup>	2.22–2.33	28–33	NA	0.33–0.38	Kurtulus et al. (2016)
Akiyoshi	Japan	2	NA	0.005; 0.009 <sup>#</sup>	2.71; 2.72	101; 75	NA	NA	Sato et al. (1981)
Soignies	Belgium	2	95	0.004	2.7	139; 170	NA	NA	Descamps et al. (2011)
Moca	Belgium	1	98.5	0.08	2.65	79	NA	NA	Descamps et al. (2011)
Asmari	Iran	63	93.4	0.0037–0.225 <sup>#</sup>	2.1–2.7	25.1–180	4.7–90	NA	Jamshidi et al. (2017); Najibi et al. (2015)
Kingston	Canada	1	92.6	0.006 <sup>#</sup>	2.72	144	90.3	NA	Prasad et al. (2009)

Note: S: Neutron-density porosity logs; #: Gravimetric method; &: Hydrostatic loading method; +: ISRM suggested method;  $\nu$ : Poisson's ratio; and NA: Not available.

observed that (a) grain size controls the reference permeability  $k^*$  at the limiting porosity  $\phi^* = 0.5$ ; (b) the exponent  $\beta = 4.5$  is higher than the power coefficients of  $\beta = 2$  and 3 proposed in the Kozeny-Carman parallel tube model where  $k \propto \phi^3/(1-\phi)^2$  (Kozeny, 1927; Carman, 1937); and (c) the high sensitivity to porosity points to preferential changes in the internal pore network structure during compaction.

Other factors influence the permeability and its spatial variability, such as mechanically induced changes (confining stress, Brace et al. (1968); deviatoric stress, Zoback and Byerlee (1975)), thermo-hydro-mechanical coupling (Summers et al., 1978), and fracture characteristics (Kranz et al., 1979; Cardona et al., 2021).

### 3. Unconfined stiffness and strength

Pores and flaws cause stress concentrations, therefore, strength and stiffness correlations often include rock porosity (Palchik and Hatzor, 2000, 2002). From a granular perspective, porosity is an indicator of contact deformation, as in sintered granular materials. In this section, we explored the correlation between porosity, stiffness and strength based on datasets compiled for carbonate formations worldwide. The carbonate formations are mostly limestone with a minimum of 90% calcite. Other formations with missing calcite content are believed to be limestone based on their formation names.

#### 3.1. Unconfined stiffness

Our database contains stress-strain data measured in 142 unconfined compression tests for 12 different carbonate samples. All specimens having a minimum of 90% calcite (i.e. limestones) had a length to diameter ratio of two, and were either air- or oven-dried. All tests were conducted at room temperature (25 °C), subjected to a typical strain rate of about  $1 \times 10^{-5}$ /s, and involved strain gauges mounted directly on rock specimens. Table 1 summarizes the

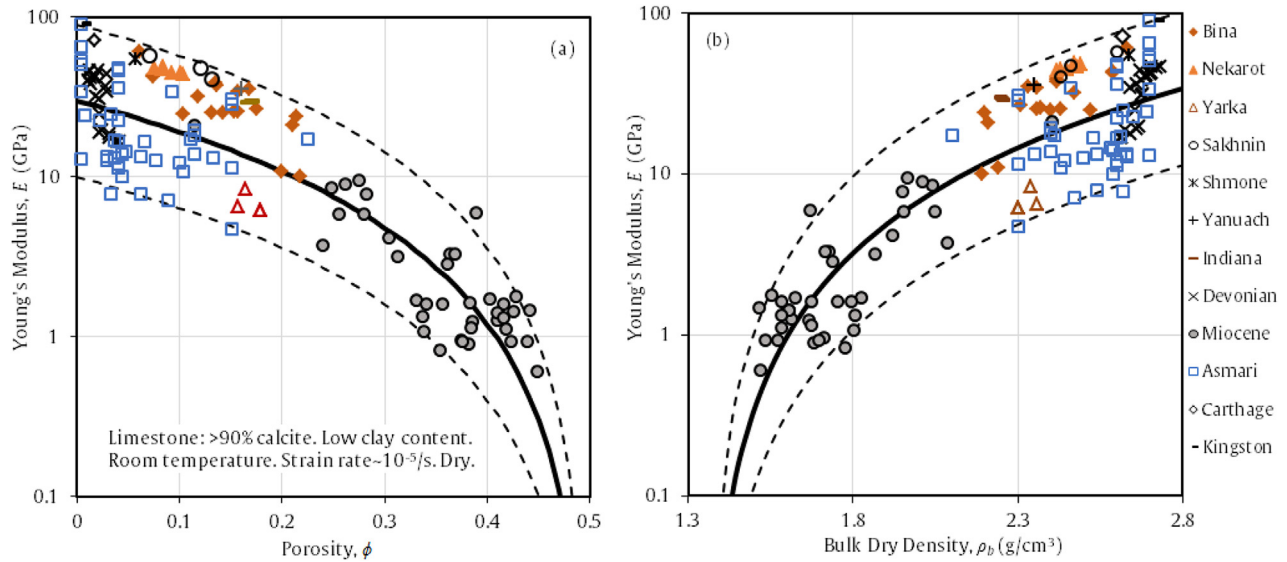
composition, porosity and dry bulk density  $\rho_b$  of the dry carbonates in the database. Porosity  $\phi$  values range from 0.004 for Asmari Limestone to 0.45 for a Hungarian Miocene Limestone. Dry bulk densities  $\rho_b$  range from 1.52 g/cm<sup>3</sup> to 2.73 g/cm<sup>3</sup> with some limestones approaching the density of calcite 2.71 g/cm<sup>3</sup>.

The reported stiffness values in terms of Young's modulus vary from  $E = 0.61$  GPa for the Hungarian Miocene Limestone to  $E = 90.3$  GPa for the Kingston Limestone. Genesis, post-depositional burial and diagenesis, and exhumation effects contribute to the observed wide range in stiffness. Fig. 2a shows the inverse relationship between Young's modulus  $E$  and porosity  $\phi$  values observed in the database. We adopted an asymptotically-correct power function to satisfy the following limits: (a) stiffness tends to  $E \rightarrow E_0$  (GPa) as porosity  $\phi \rightarrow 0$ ; and (b) there is virtually no stiffness in the initial granular packing with limiting porosity  $\phi^*$  before burial and diagenesis. This limiting porosity  $\phi^*$  can be related to the concept of critical porosity  $\phi_c$  that separates two domains: a continuous frame-supported rock medium with  $\phi < \phi_c$  and fluid-supported solid suspensions with  $\phi > \phi_c$  (Nur et al., 1998). However, very few studies investigate the  $\phi_c$  for carbonate rocks (Fournier and Borgomano, 2009), and the  $\phi_c = 0.4$  suggested for carbonate rocks by Nur et al. (1998) is close to the  $\phi^* = 0.5$  based on a simple cubic packing of monosized particles. Then, we have

$$E = E_0 \left( 1 - \frac{\phi}{\phi^*} \right)^\alpha = 30 (1 - 2\phi)^2 \quad (2)$$

The second equality in Eq. (2) shows the fitted trend superimposed on Fig. 2a with the asymptotic value  $E_0 = 30$  GPa. Nonlinear equations were fit using the statistical software RStudio to determine the asymptote and the exponent (R Core Team, 2016).

Results in Fig. 2a support the following observations: (a) the mean stiffness at zero porosity  $E_0 = 30$  GPa is significantly lower than the stiffness of a single calcite crystal due to the presence of crystal boundaries and impurities at contacts (single crystal: 60–



**Fig. 2.** Young's modulus under unconfined conditions. (a) As a function of porosity: mean trend  $E = 30(1 - 2\phi)^2$ . (b) As a function of bulk dry density: mean trend  $E = 30 \left( \frac{2\rho_b}{2.71} - 1 \right)^2$ . In both cases, the lower and upper bounds are shown in dashed lines and correspond to  $E_0 = 10$  GPa and  $E_0 = 90$  GPa, respectively.

70 GPa, see Beiki et al., 2013); (b) the limiting porosity of a granular packing  $\phi^* \approx 0.5$  is an adequate upper bound; and (c) the exponent  $\alpha = 2$  captures the sensitivity of stiffness to porosity. For a given porosity, the variability in stiffness among different carbonate formations in the database reflects the differences in formation history and geological processes such as compaction, dissociation, precipitation, and pore topology (Roehl and Choquette, 1985; Mazzullo et al., 1992; Durrast and Siegesmund, 1999).

Porosity  $\phi$  is a function of the rock dry bulk density  $\rho_b$  (g/cm³) and the mineral density  $\rho_m$  (g/cm³). Then, we can relate stiffness to dry bulk density, from Eq. (2):

$$E = E_0 \left( 2 \frac{\rho_b}{\rho_m} - 1 \right)^\alpha = 30 \left( \frac{2\rho_b}{2.71} - 1 \right)^2 \quad (3)$$

The second equality in Eq. (3) shows the fitted trend with assumed values  $\phi^* = 0.5$  and  $\rho_m = 2.71$  g/cm³ for calcite (Fig. 2b).

### 3.2. Unconfined compressive strength database

The initial porosity plays a prevalent role on the unconfined compressive strength (UCS) of cohesive media, including carbonate rocks (Rzhevsky and Novick, 1971; Farquhar et al., 1994; Chang, 2004; Fay-Gomord et al., 2016; Astorqui et al., 2017; Maryam et al., 2018). The mean grain size and mineralogy have a secondary effect on strength, except for low-porosity homogenous carbonates (Chang et al., 2006; Moh'd, 2009; Asef and Farrokhrouz, 2010; Jensen et al., 2010).

The database compiled for this study contains 221 UCS values gathered from 24 different dry limestone samples at 13 locations from around the world (Table 1). Data show a range from  $UCS = 3.08$  MPa for the Hungarian Miocene Limestone to  $UCS = 310$  MPa for Solnhofen Limestone. Data trends in Fig. 3a confirm the inverse relationship between UCS and  $\phi$ . Once again, the data suggest a power function relation between UCS and  $\phi$  (Fig. 3), where the two asymptotes are (a) the rock strength at zero porosity  $UCS_0$ ; and (b) vanishing strength  $UCS \rightarrow 0$  as porosity approached the limiting granular porosity  $\phi \rightarrow \phi^*$ .

$$UCS = UCS_0 \left( 1 - \frac{\phi}{\phi^*} \right)^\beta = 100 (1 - 2\phi)^2 \quad (4)$$

The second equality in Eq. (4) assumes  $\phi^* = 0.5$ , and the fitted values capture the average strength at zero porosity  $UCS_0 = 95$  MPa (see also Beiki et al., 2013). The exponent  $\beta = 2$  reflects the high strength sensitivity to porosity. The variability in strength at low porosity points to the differences in mineralogy and the effect of impurities on contact strength. In terms of dry bulk density, Eq. (4) becomes (Fig. 3b):

$$UCS = UCS_0 \left( 2 \frac{\rho_b}{\rho_m} - 1 \right)^\beta = 100 \left( \frac{2\rho_b}{2.71} - 1 \right)^2 \quad (5)$$

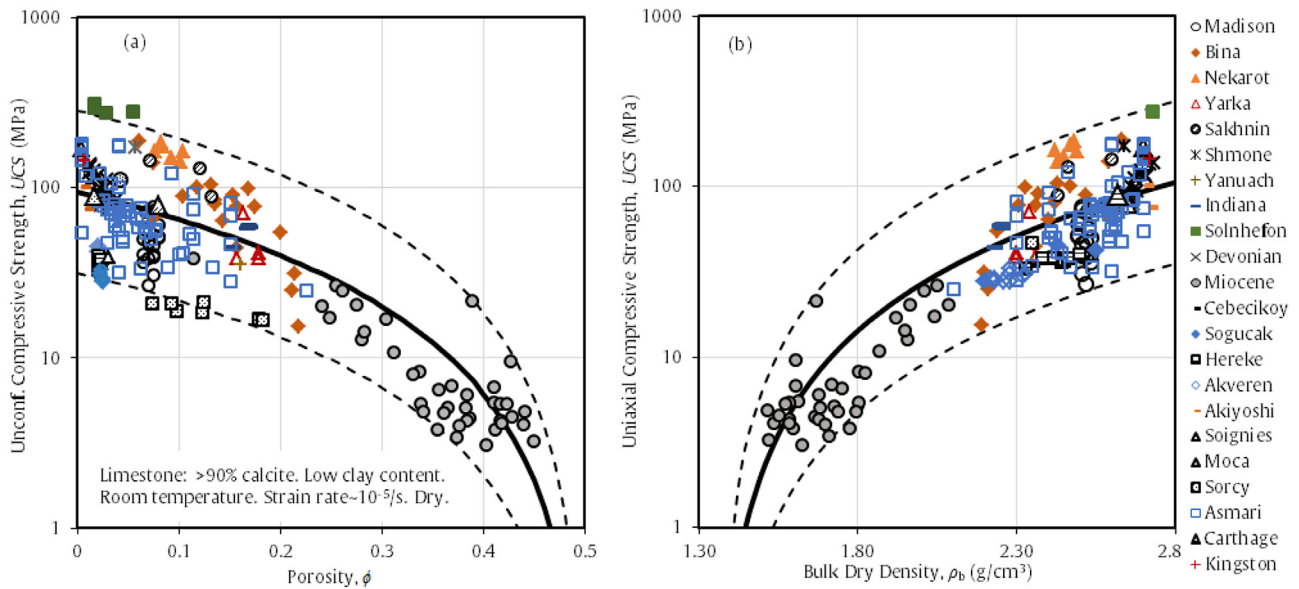
### 3.3. Relationship between unconfined stiffness, strength and porosity

Fig. 4 shows a linear relationship between  $E$  and UCS, where both parameters increase by two orders of magnitude when porosity decreases from 0.5 to 0. The log-log plot in Fig. 4 allows a clear assembly of the wide range of stiffnesses from 0.61 GPa to 90.3 GPa. Furthermore, the trend is independent of porosity  $\phi$  (Fig. 4 includes 133 entries). Indeed, Eqs. (2) and (4) or Eqs. (3) and (5) predict a porosity-independent linear relation between  $E$  and UCS:

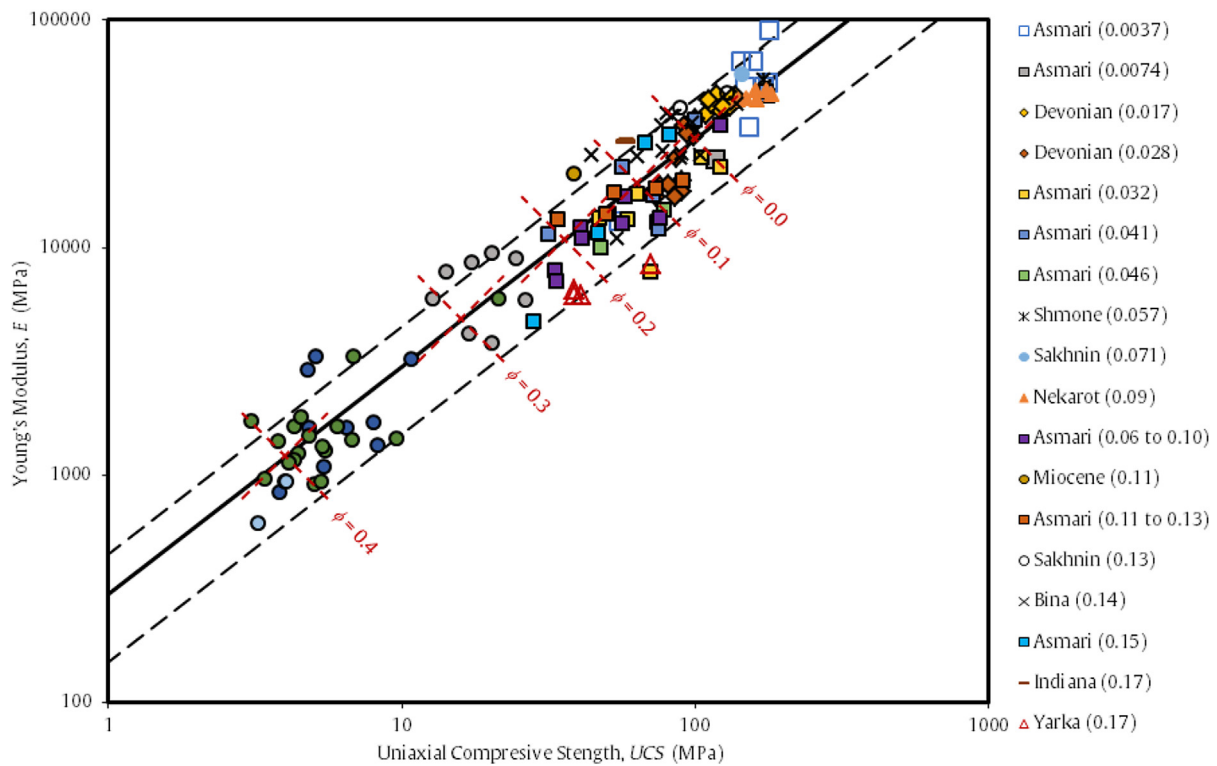
$$E = 300UCS \quad (6)$$

The linear relationship is recommended over a power relationship due to its simplicity and comparable prediction. The ratio ( $E/UCS$ ) follows a log-normal distribution with mean  $\mu[\log_{10}(E/UCS)] = 2.46$  and standard deviation  $SD[\log_{10}(E/UCS)] = 0.153$ ; the corresponding factors are 287 for  $\mu$ , and 202 and 408 for  $\mu \pm SD$ . The stiffness-strength relationship observed in carbonates resembles that for most solids, where stiffness is 2–3 orders of magnitude larger than strength.





**Fig. 3.** Unconfined compressive strength. (a) As a function of porosity: mean trend  $UCS = 100(1 - 2\phi)^2$ ; (b) As a function of bulk dry density: mean trend  $UCS = 100\left(\frac{2\rho_b}{2.71} - 1\right)^2$ . In both cases, the lower and upper bounds are shown in dashed lines and correspond to  $UCS_0 = 33$  MPa and  $UCS_0 = 300$  MPa, respectively.



**Fig. 4.** Young's modulus versus unconfined compressive strength under unconfined conditions. The central trend is  $E = 300UCS$ . The lower and upper bounds shown in dashed lines correspond to  $E = 150UCS$  and  $E = 450UCS$ . Porosity values of the carbonates are included in the parentheses.

#### 4. Stress-dependent strength and ductility

The previous section addressed unconfined conditions. Rock stiffness and strength are stress-dependent properties (Farquhar, 1993; Vernik et al., 1993). This section explores the effective stress-dependent strength and brittle-to-ductile transition of carbonates.

##### 4.1. Strength

The database compiles 22 triaxial compression test datasets gathered from 15 dry carbonate samples at 10 different locations (Table 2). The minimum calcite component is 90% in all samples, i.e. limestones, except Wolfcamp Limestone from Texas, USA and Saint Maximin Limestone from France. Porosity  $\phi$  values range from

0.003 for Oak Hall Limestone to 0.37 for Saint Maximin Limestone. All triaxial compression data are gathered at room temperature (25 °C).

Data analysis seeks to identify strength parameters, including a link between confined and unconfined strengths. Fig. 5 (inset) shows the Mohr circle from an axial compression triaxial test and the linear Mohr-Coulomb failure envelope both on the normal stress and shear stress plane. The effective normal  $\sigma'_f$  and shear stress  $\tau'_f$  on the failure plane are a function of the effective major  $\sigma'_1$  and minor  $\sigma'_3$  principal stresses and the angle of internal shear strength  $\varphi$ :

$$\sigma'_f = \frac{1}{2}(\sigma'_1 + \sigma'_3) - \frac{1}{2}(\sigma'_1 - \sigma'_3)\sin \varphi = p' - q'\sin \varphi \quad (7)$$

$$\tau'_f = \frac{1}{2}(\sigma'_1 - \sigma'_3)\cos \varphi = q'\cos \varphi \quad (8)$$

where  $p' = (\sigma'_1 + \sigma'_3)/2$  and  $q' = (\sigma'_1 - \sigma'_3)/2$ . The Mohr-Coulomb failure criterion relates the normal  $\sigma'_f$  and shear stress  $\tau'_f$  on the failure plane through the rock cohesion  $c$  and the angle of internal shear strength  $\varphi$ .

$$\tau'_f = c + \sigma'_f \tan \varphi \quad (9)$$

Eqs. (7)–(9) are combined to anticipate the relationship between principal stresses at failure:

$$\sigma'_1 = \frac{2c \cos \varphi}{1 - \sin \varphi} + \frac{1 + \sin \varphi}{1 - \sin \varphi} \sigma'_3 \quad (10)$$

The UCS is the effective axial stress  $\sigma'_1$  at failure when  $\sigma'_3 = 0$ . Then, Eq. (10) predicts:

$$c = UCS \frac{1 - \sin \varphi}{2 \cos \varphi} \quad (11)$$

We used the compiled triaxial dataset to determine the Mohr-Coulomb strength parameters  $\varphi$  and  $c$  (Eq. (9), Table 2). The mean angle of internal shear strength is  $\varphi = 31^\circ$  for carbonates with a minimum of 90% calcite, i.e. limestone, and a maximum porosity  $\phi \leq 0.3$  (Note: Saint Maximin Limestone with  $\phi = 0.37$  exhibits  $\varphi = 14^\circ$ ). Porosity has a negligible effect on the angle of internal shear strength (see also Scott, 1989).

Fig. 5 shows the cohesive intercept  $c$  measured in triaxial tests (red dots) and computed from unconfined compressive strength UCS assuming  $\varphi = 31^\circ$ . The fitted trend parallels with Eq. (4):

$$c = c_0 \left(1 - \frac{\phi}{\phi^*}\right)^\psi = 80(1 - 2\phi)^3 \quad (12)$$

The second equality shows the mean trend for a limiting porosity  $\phi^* = 0.5$ . The mean cohesion at zero porosity  $c_0$  is 80 MPa, and the fitted exponent  $\psi = 3$ . Analogous to the UCS, cohesion  $c$  rapidly decreases as porosity tends to  $\phi \rightarrow \phi^*$ . Finally, Eqs. (9) and (12) combine in the following mean failure criterion for carbonates based on the available data:

$$\tau_f = 80(1 - 2\phi)^3 + \sigma_f \tan 31^\circ \quad (13)$$

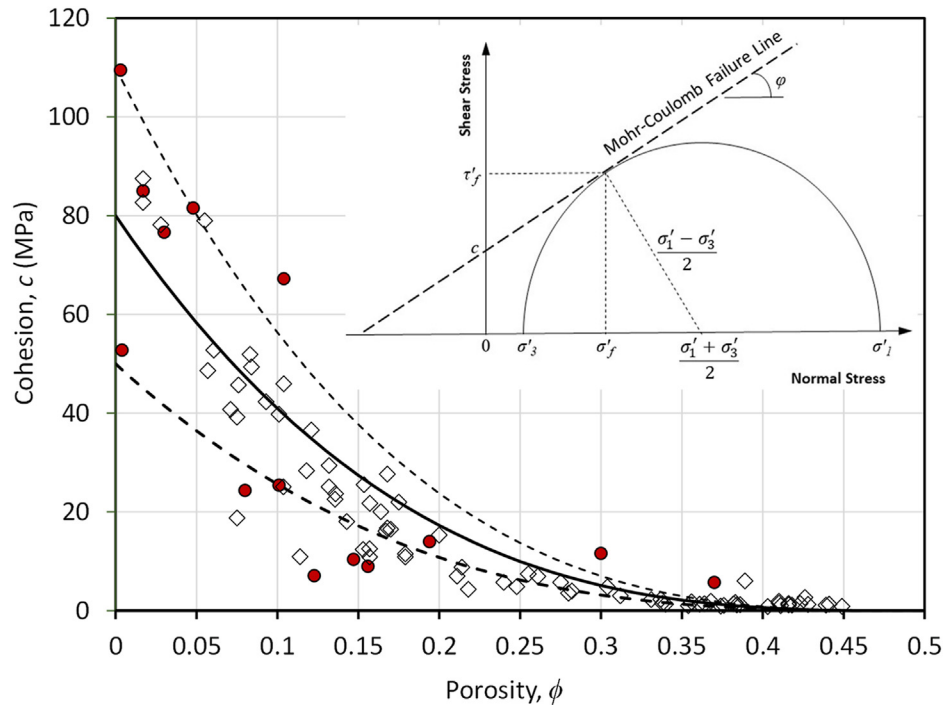
In terms of the mean effective stress  $p' = (\sigma'_1 + \sigma'_3)/2$  and the

**Table 2**

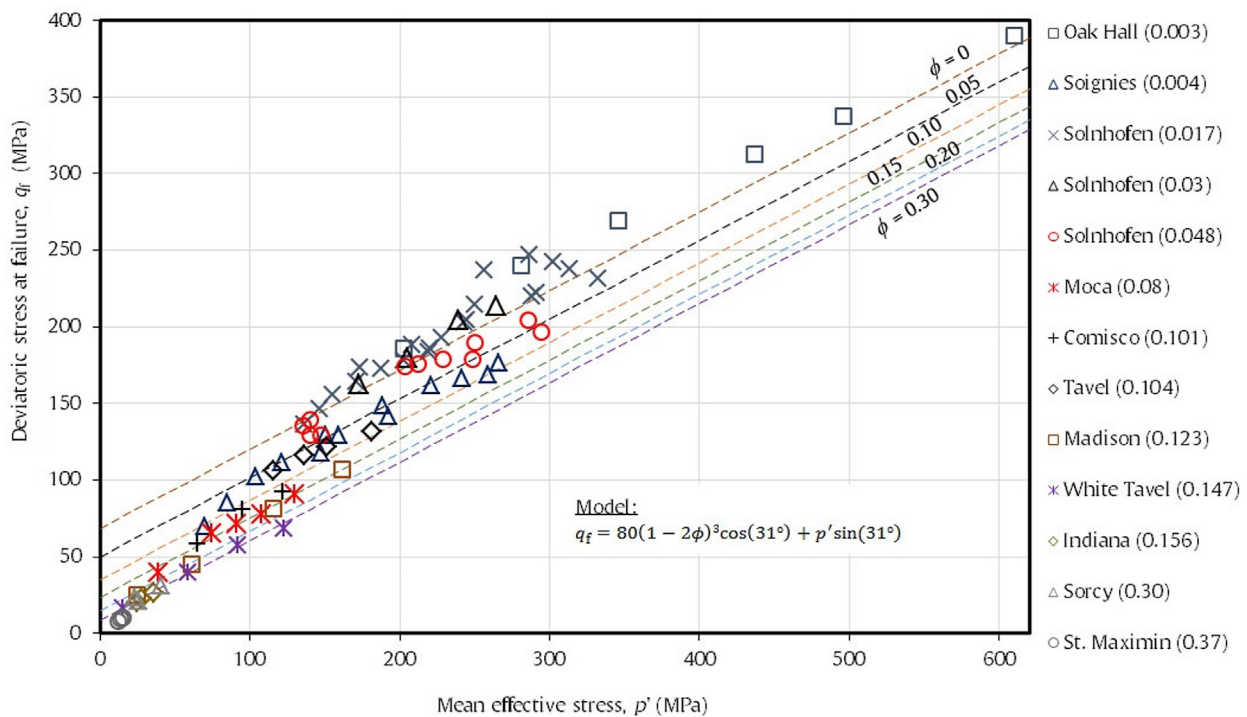
Database of triaxial compressive test results of dry carbonates compiled from published literature.

Carbonate	Location	Number of tests	Percent Calcite (%)	$\phi$	Effective confining stress (MPa)	$\epsilon_{\max}$ (%)	Mohr-Coulomb criterion		Failure Source
							$c$ (MPa)	$\varphi$ (°)	
Madison	WY, USA	3	98	0.085 <sup>§</sup>	17, 35, 55	0.65	7.15	39.4	B; T
Madison <sup>®</sup>	WY, USA	6	98	0.094 –0.23 <sup>*</sup>	7, 35, 55				B; T
Fusselman	TX, USA	3	97	0.03 <sup>®</sup>	0, 101, 203	16	NA	NA	B; T; D
Wolfcamp	TX, USA	3	75	0.042 <sup>®</sup>	0, 101, 203	4	NA	NA	B; T; D
Wolfcamp	NM, USA	3	96	0.042 <sup>®</sup>	0, 101, 203	9	NA	NA	B; T; D
Oak Hall	PA, USA	8	99	0.003	18, 41, 76, 125, 159, 220, 335, 466	4	109.6	29.3	B; T; D
Indiana	Indiana, USA	11	99	0.194 <sup>®</sup>	0, 7, 14, 21, 28, 34, 41, 48, 55, 62, 69	2.7	9	30.8	B; T; D
Indiana	Indiana, USA	6	99	0.156 <sup>®</sup>	5, 10, 20, 30, 40, 50	3.5	14	27.1	B; T; D
Soignies	Belgium	5	95	0.004	0, 30, 50, 75, 90	3	52.8	30.4	B; T
Moca	Belgium	8	98.5	0.08	0, 10, 20, 30, 40, 60, 80, 100	3.5	24.4	32.9	B; T; D
Sorcy	Belgium	6	~100	0.30	0, 5, 10, 20, 30, 60	1.5	11.7	29.7	B; T; D
Solnhofon	Germany	6	99	0.03 <sup>®</sup>	50, 100, 200, 300, 350, 435	5	76.7	35.5	B; T; D
Solnhofon	Germany	7	99	0.048	17, 37, 60, 81, 122, 204, 517	5	81.6	26.9	B; T; D
Solnhofon	Germany	7	99	0.017 <sup>®</sup>	0, 76, 101, 127, 152, 304, 507	1.9	85.1	32.9	B; T; D
Solnhofon	Germany	5	99	0.059 <sup>®</sup>	100, 200, 400, 600, 800	2	NA	NA	T; D
Solnhofon	Germany	8	99	0.015	35, 35, 69, 69, 104, 104, 138, 138	1.4	NA	NA	B; T
St. Maximin	France	5	61	0.37 <sup>®</sup>	3, 5, 6, 9, 12	1.6	5.8	14.1	B; T; D
Tavel	France	8	NA	0.104 <sup>®</sup>	10, 20, 30, 50, 100, 150, 200, 240	4.6	67.3	22.8	B; T; D
White Tavel	France	14	99	0.147 <sup>®</sup>	0, 20, 20, 35, 35, 55, 55, 70, 70, 70, 85, 85, 85	4.4	10.4	29.8	B; T; D
Majella	Italy	3	NA	0.32 <sup>®</sup>	25, 25, 25	NA	NA	NA	D
Comiso	Italy	4	97.7	0.101 <sup>®</sup>	7, 15, 30, 50	1.3	25.4	36.9	B; T; D
Devonian	Turkey	3	NA	0.023 <sup>®</sup>	0, 101, 203	20	NA	NA	B; T; D

Note:  $\epsilon_{\max}$ : Maximum axial strain attained in percentage at the maximum applied deviator stress; <sup>®</sup>: Temperature at 93 °C and brine-saturated; <sup>\*</sup>: Gas expansion method; B: Brittle regime; T: Transition regime; and D: Ductile regime.



**Fig. 5.** Mohr-Coulomb failure criterion – Cohesion intercept as a function of porosity. Red dots: Values extracted from sets of triaxial tests. White diamonds: Computed from unconfined compressive strength for mean  $\phi = 31^\circ$ . The mean trend is  $c = 80(1 - 2\phi)^3$ . The lower and upper bounds shown in dashed lines correspond to  $c_0 = 50$  MPa and  $c_0 = 110$  MPa, respectively.

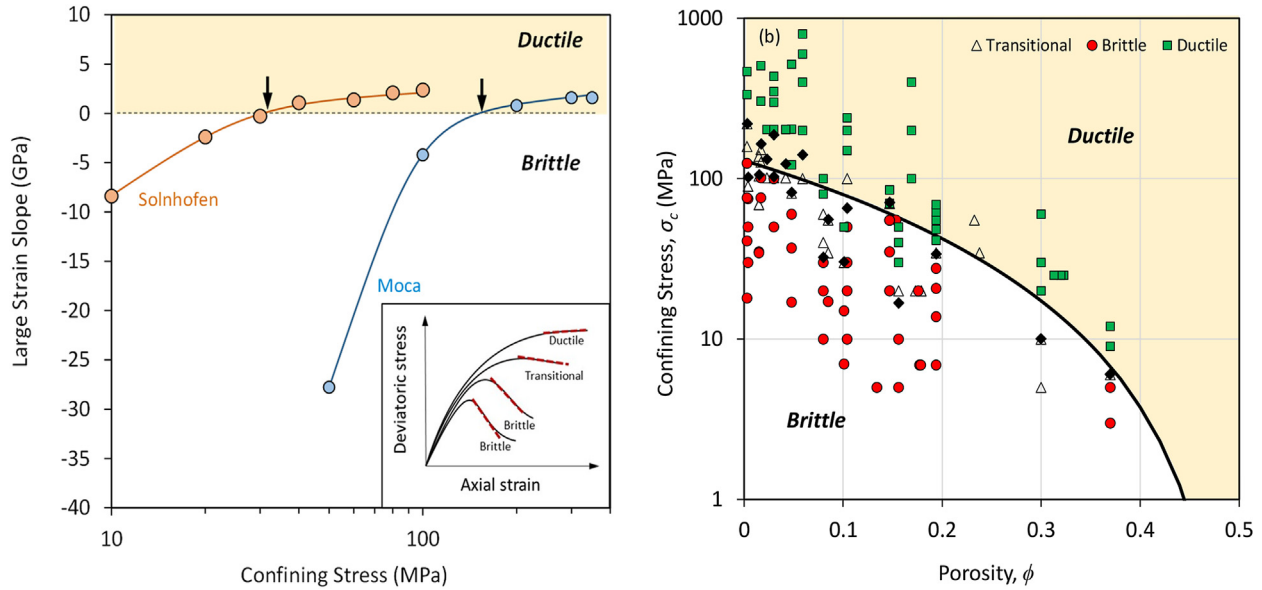


**Fig. 6.** Mohr-Coulomb failure criterion in  $q$ - $p'$  space. Data collected from the literature. All trends computed for the same parameters (Eq. (14)). Porosity values of the carbonates are included in the parentheses.

deviatoric stress at failure  $q_f = (\sigma'_1 - \sigma'_3)_{\max}/2$ , the Mohr-Coulomb failure criterion is

$$q_f = c \cos \phi + p' \sin \phi \quad (14)$$

Compiled experimental results lead to the following semi-empirical function for deviatoric stress at failure in terms of the mean effective stress (Eq. (12) and (14)):



**Fig. 7.** Brittle-to-ductile transition. (a) determination of the brittle-to-ductile transition using the large strain slope, and (b) Triaxial data versus porosity: single tests and inferred brittle to ductile transition stress (back diamonds). The dividing boundary is  $\sigma_{bd} = 130 (1 - 2\phi)^{2.2}$ .

$$q_f = 80 (1 - 2\phi)^3 \cos 31^\circ + p' \sin 31^\circ \quad (15)$$

Fig. 6 shows the predicted  $p'$  and  $q_f$  failure lines using Eq. (15) superimposed on top of triaxial test data gathered for 13 carbonates (porosities  $\phi$  from 0 to 0.37).

#### 4.2. Brittle-to-ductile transition

Rocks subjected to low confinement experience propagation of contact breakage, contiguous damage coalescence, shear localization, dilation and a sudden loss in strength (Brace, 1978; Baud et al., 2000, 2009; Vajdova et al., 2004; Nicolas et al., 2017). However, rocks under high confinement above brittle-to-ductile transition experience shear-enhanced compaction, pore collapse, crystal plasticity, distributed grain debonding and micro-cracking, grain rotation and grain-scale plastic flow (Fredrich et al., 1989; Dresen and Evans, 1993; Renner and Rummel, 1996; Paterson and Wong, 2005; Zhu et al., 2010; Dautriat et al., 2011).

The stress-dependent brittle-to-ductile transition in carbonate rocks follows similar patterns (Renner and Rummel, 1996; Wong et al., 1997; Vajdova et al., 2004; Wong and Baud, 2012; Nicolas et al., 2016). Internally, there is a transition from frictional sliding in brittle failure to cataclastic flow in ductile failure (e.g. Logan, 1987; Wong and Baud, 2012). However, the brittle-to-ductile transition in carbonate rocks is often unclear as these rocks may experience substantial inelastic strain but eventually fail in brittle mode (Jaeger et al., 2007). The extended strain hardening response reflects additional deformation mechanisms in carbonate rocks, including calcite twinning and slip (Turner et al., 1954; Griggs et al., 1960; Fredrich et al., 1989).

There is no standardized protocol to determine the confining stress at the brittle-to-ductile transition  $\sigma_{bd}$ . Previous studies considered the axial stress at volumetric strain reversal (Walton et al., 2017) and the post-yield strength and dilatancy (Wong and Baud, 2012). We followed a simple yet robust approach to estimate the  $\sigma_{bd}$  from stress-strain triaxial data (Fig. 7a): (a) determine the stress-strain slope at large-strain; (b) plot the measured slope against the confining stress; and (c) the brittle-to-ductile transition is the effective confining stress at the zero intercept, where the

slope changes from negative “strain-softening” to positive “strain-hardening” (Fig. 7a).

We repeated this methodology for published triaxial datasets obtained for 19 dry carbonates at room temperature 25 °C (Table 2). Fig. 7b shows all test results plotted on a confining stress versus porosity space. Each data point represents a triaxial test: green squares mean ductile strain-hardening response (positive slope), red circles mean brittle post-peak softening response (marked post-peak negative slope), and white triangles mean transitional response.

Black diamonds show the determined  $\sigma_{bd}$  for each dataset following the procedure described above (as in Fig. 7a).  $\sigma_{bd}$  values range from 15.5 MPa for Saint Maximin Limestone to 610 MPa for Oak Hall Limestone (see also: Heard, 1960; Rutter, 1972). Overall, the data suggest that lower-porosity carbonates exhibit brittle-to-ductile transitions at higher confining stresses (see also Logan, 1987). In agreement with models selected above, we used a power model to relate the brittle-to-ductile transition stress  $\sigma_{bd}$  (MPa) to porosity  $\phi$  (solid line in Fig. 7b),

$$\sigma_{bd} = \sigma_0 \left(1 - \frac{\phi}{\phi^*}\right)^\xi = 130(1 - 2\phi)^{2.2} \quad (16)$$

where  $\sigma_0$  is confining stress when  $\phi$  approaches zero, and the exponent  $\xi = 2.2$ .

In this case, the limiting porosity  $\phi^* = 0.5$  is in agreement with inherently ductile, very loose granular packing.

#### 5. Physical model analysis

Published empirical models for the hydraulic and mechanical properties of carbonate rocks are either exponential or power functions written in terms of density  $\rho$ , porosity  $\phi$  or solidity  $\Sigma = 1 - \phi$ . Table 3 summarizes these models and includes the new relations adopted in this study (Tables 1 and 2).

Previous studies often considered rocks as a continuum where pores and flaws are soft anomalies that cause stress concentrations and act as nucleation sites for fracture initiation and propagation. When tested against our databases, these models show adequate



**Table 3**

Carbonate rocks: Empirical equations for hydraulic and mechanical properties (See Tables A1 and A2 in Appendix A for details and fitting parameters).

Equations	Source
Permeability $k$ $\frac{k}{k^*} = \left(\frac{\phi}{1-\phi}\right)^\beta$	This study (limiting granular porosity $\phi^* = 0.5$ )
Unconfined stiffness $E$ $E = E_0 e^{-\alpha\phi}$ $E = E_0(1-2\phi)^\alpha = E_0\left(2\frac{\rho_b}{\rho_m} - 1\right)^\alpha$ $E = a' + b \text{ UCS}$ $E = a \text{ UCS}^\theta \phi^\vartheta d_g^\omega$ (Unit of UCS in MPa)	Farquhar et al. (1994); Ameen et al. (2009); Faÿ-Gomord et al. (2016) This study (limiting granular porosity $\phi^* = 0.5$ ) Afsari et al. (2009); Palchik (2011); this study Palchik and Hatzor (2000); Chang (2004); Asef and Farrokhrrouz (2010); Najibi et al. (2015)
Unconfined compressive strength UCS $\text{UCS} = \text{UCS}_0 e^{-\beta\phi}$ $\text{UCS} = \text{UCS}_0(1-\phi)^\beta = \text{UCS}_0\left(\frac{\rho_b}{\rho_m}\right)^\beta$ $\text{UCS} = \text{UCS}_0(1-2\phi)^\beta = \text{UCS}_0\left(2\frac{\rho_b}{\rho_m} - 1\right)^\beta$ $\text{UCS} = a' - b \ln\phi$ $\text{UCS} = a' - b\phi - c\rho + dS_w$	Farquhar et al. (1994); Chang (2004); Moh'd (2009); Faÿ-Gomord et al. (2016) Rzhevsky and Novick (1971); Astorqui et al. (2017) This study (limiting granular porosity $\phi^* = 0.5$ ) Hebib et al. (2017) Maryam et al. (2018)
Stress at brittle-to-ductile transition $\sigma_{bd}$ $\sigma_{bd} = \sigma_0 \left(1 - \frac{\phi}{\phi^*}\right)^\xi = 130(1-2\phi)^{2.2}$	This study (for initial granular porosity $\phi^* = 0.5$ )
Confinement-dependent shear strength $q_f$ $q_f = c_0(1-2\phi)^\psi \cos\varphi + p'\sin\varphi$	This study

Note:  $d_g$ : mean grain size;  $S_w$ : water saturation; Fitting factors  $a'$ ,  $b$ ,  $c$ ,  $d$  and exponents  $\alpha$ ,  $\beta$ ,  $\theta$ ,  $\omega$ ,  $\vartheta$ ,  $\xi$ .

Sources: Rocks from multiple locations including Algeria, Belgium, France, Iran, Israel, Russia, Saudi Arabia, Spain, United Kingdom, and USA.

trends for low porosity carbonates, but fail to anticipate trends at high porosity.

In fact, the inherent granular genesis of carbonates readily confirmed by SEM images: (a) supports a contact-mechanics based analysis rather than a porous-continuum formulation; (b) enforces a limiting porosity  $\phi^*$  where particles jam and start forming percolating force chains; and (c) implies that mechanical properties  $P$  below the percolation porosity  $\phi < \phi^*$  follow a power function of the form (Sahimi, 1994; Kovacic, 1999; Nezamabadi et al., 2021; Ternero et al., 2021):

$$P = P_0 \left(1 - \frac{\phi}{\phi^*}\right)^\alpha \quad (17)$$

Indeed, this expression matches trends identified above for all the mechanical parameters. The Taylor expansion of Eq. (17) for  $\phi \rightarrow 0$  is

$$P = P_0 \left(1 - \frac{\alpha\phi}{\phi^*}\right)^\alpha = P_0(1 - 2\alpha\phi) \quad (18)$$

where  $P_0$  is the mechanical property at the percolation porosity; the second equality corresponds to  $\phi^* = 0.5$ . Clearly, we can always approximate trends with a linear relationship near  $\phi \rightarrow 0$  as adopted in some earlier studies where pore-scale characteristics dominate. On the other hand, percolation and contact mechanics gain relevance at high initial porosities  $\phi \rightarrow \phi^*$ .

The simple cubic packing is the loosest arrangement of mono-sized spherical particles, thus its porosity  $\phi_0 = 0.476$  is an upper bound for the porosity granular materials may attain (Note: there may be intra-particle vuggy porosity in carbonates). This bound is consistent with our database and justifies the adopted limiting porosity  $\phi^* = 0.5$ . Time and confinement promote contact yield as well as dissolution-precipitation. Under isotropic conditions, the six contacts around a particle flatten and spherical caps at contacts dissolve and re-precipitate coating free surfaces nearby. Spherical caps in  $x$ ,  $y$ ,  $z$  directions interfere when contact areas reach a radius  $a = r\sin 45^\circ$  ( $r$ : particle radius). At this point, the remaining pores become isolated and the occluded porosity  $\phi$  is 0.035, as observed in sintered materials in powder metallurgy (see Danninger et al., 1993). Detailed analysis is listed in Appendix A. In fact, most

carbonates in our database have porosities between the two boundaries identified above,  $0.035 \leq \phi \leq 0.476$ .

The rock strength and stiffness are determined by localized contact deformation and contact strength, which are function of the contact area relative to the total area. A geometric analysis and algebraic manipulations lead to the following relationship between relative contact area  $A_r$  and porosity  $\phi$  (see Appendix A)

$$\phi = 1 - \frac{\pi}{2} - 3A_r + \left[3\sqrt{\pi} \left(\frac{1}{\pi + 4A_r}\right)^{3/2}\right]^{-1} \quad (19)$$

Eq. (19) predicts a trend that is very similar to the power function  $(1-2\phi)^2$  adopted above for stiffness, strength, and brittle-to-ductile transition (with limiting porosity  $\phi^* = 0.5$ , Eqs. (2), (4) and (16), see Appendix A). Therefore, contact mechanics determines the small and large strain mechanical properties of carbonates. Note that the relationship between relative contact area  $A_r$  and porosity  $\phi$  in Eq. (9) is independent of grain size, as observed in experimental studies reported above.

Rock studies using discrete element simulations with cemented grains show that the brittle-to-ductile transition corresponds to the stress where bond breakage changes from tensile-controlled (at low confinement) to shear-controlled at high confinement (Garcia, 2020). The high angle of internal shear strength observed in carbonates  $\varphi \approx 31^\circ$  indicates delayed contact tensile failure with increased confinement (Fig. 6).

## 6. Conclusions

Results in this context show that the mechanical and hydraulic properties of limestone carbonates reflect their granular genesis and diagenetic overprint and exhibit a strong dependency on porosity. Power functions capture the effect of porosity on unconfined stiffness, compressive strength and the brittle-to-ductile transition stress. The observed power functions  $(1-\phi/\phi^*)^\alpha$  agree with percolation theory. We adopted a limiting granular porosity  $\phi^* = 0.5$  for all models which is consistent with the loosest packing of monosized spheres. Asymptotes for the mechanical properties at zero porosity are lower than the corresponding mechanical properties for a single calcite crystal due to compliance and the presence of impurities at crystal interfaces. On the other hand, the

unconfined stiffness, compressive strength and brittle-to-ductile transition stress vanish as the limestone carbonate porosity approaches the loosest granular packing  $\phi \rightarrow \phi^*$ . The fitted power equations have exponents  $\alpha = 2$  or greater in agreement with percolation theory, highlighting the sensitivity of mechanical properties to porosity.

The linear relationship between unconfined stiffness  $E$  and strength  $UCS$  resembles trends observed for a wide range of solids. In the case of limestone carbonates,  $E/UCS$  is porosity independent and follows a log-normal distribution with mean  $\mu(E/UCS) \approx 300$  and standard deviation  $SD(E/UCS) \approx 100$ .

The high angle of internal shear strength measured for limestone carbonates reflects delayed contact failure with increased confinement, and it is not sensitive to porosity. However, the cohesive intercept  $c$  decreases with porosity  $\phi$ . Consequently, failure envelopes depict parallel lines as a function of porosity in the  $\tau$ - $\sigma$  or  $q$ - $p'$  spaces.

The permeability of carbonates spans more than six orders of magnitude. Grain size controls pore size and determines the reference permeability  $k^*$  at the limiting porosity  $\phi^* = 0.5$ . For a given grain size from fine to coarse-grained dominant carbonates, permeability  $k$  is very sensitive to changes in porosity due to internal changes in pore structure and connectivity.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jrmge.2022.07.017>.

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