Pore-scale fluid dynamics resolved in pressure fluctuations at the Darcy scale

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Abstract

Complex flow dynamics have been observed, at the pore-scale, during multiphase through porous rocks. These dynamics are not captured in large scale models exploring the migration and trapping of subsurface fluids e.g., CO2 or hydrogen. Due to limitations in imaging capabilities, these dynamics cannot be observed directly at the larger, Darcy scale. Instead, by using pressure data from pore-scale (mm-scale) and core-scale (cm-scale) experiments, we show that fluctuations in pressure measured at the core-scale reflect specific fluid displacement events taking place at the pore-scale. The spectral characteristics of the pressure data depends on the flow dynamics, size of the rock sample, and heterogeneity of pore space. While high resolution imaging of large samples would be useful in assessing flow dynamics across many of the scales of interest, such an approach is currently infeasible. We suggest an alternative, pragmatic, approach examining pressure data in the time-frequency domain using wavelet transformation.

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8 Abstract

Complex flow dynamics have been observed, at the pore-scale, during multiphase through q porous rocks. These dynamics are not captured in large scale models exploring the mi-10 gration and trapping of subsurface fluids e.g., CO₂ or hydrogen. Due to limitations in 11 imaging capabilities, these dynamics cannot be observed directly at the larger, Darcy 12 scale. Instead, by using pressure data from pore-scale (mm-scale) and core-scale (cm-13 scale) experiments, we show that fluctuations in pressure measured at the core-scale re-14 flect specific fluid displacement events taking place at the pore-scale. The spectral char-15 acteristics of the pressure data depends on the flow dynamics, size of the rock sample, 16 and heterogeneity of pore space. While high resolution imaging of large samples would 17 be useful in assessing flow dynamics across many of the scales of interest, such an ap-18 proach is currently infeasible. We suggest an alternative, pragmatic, approach examin-19 ing pressure data in the time-frequency domain using wavelet transformation. 20

21 Plain Language Summary

Complex fluid dynamics have been observed in small pores within rocks. These dy-22 namics have not been accounted for in larger scale modelling efforts of CO₂ or hydro-23 gen. Limitations in imaging prevent the direct observation of these dynamic at larger 24 scales, creating uncertainty in how these dynamics manifest at larger scales. But by an-25 alyzing the pressure data, and fluctuations in pressure measurements, we can infer the 26 27 small-scale dynamics without imaging. We apply our findings to larger samples and discover that the fluctuations are dependent on the type of flow dynamics occurring, sam-28 ple size, and the composition of the sample. We present a practical approach for assess-20 ing the dynamics at the larger scale, where direct imaging is currently infeasible, by ex-30 ploring the pressure data using a technique called continuous wavelet transformation. 31

32 1 Introduction

Fluid flow in the subsurface is a complex process, controlled by the interaction of 33 multiple fluids with one another, and a heterogeneous pore space. It is central to the safe 34 storage of CO_2 in the subsurface (Rubin & De Coninck, 2005; Bachu & Adams, 2003; 35 Benson et al., 2012), and the storage and retrieval of hydrogen underground (Boon & 36 Hajibeygi, 2022; Thiyagarajan et al., 2022), as examples. Depending on the flow rate, 37 fluid viscosity, wettability of the fluids, and connectivity of the pore space, different flow 38 mechanisms will prevail (Blunt, 2017; Lenormand et al., 1983; Avraam & Payatakes, 1995; 39 Spurin et al., 2019a, 2019b; Rücker et al., 2015; Zou et al., 2018; Zhao et al., 2016). These 40 processes span many orders of magnitudes for both timescales and length scales, from 41 sub-second to hours (Berg et al., 2013; Schlüter et al., 2017; McClure et al., 2020), and 42 sub-pore to multi-pore (Berg et al., 2013; Moebius & Or, 2014; Spurin et al., 2020). Fluid 43 flow is also heavily controlled by the heterogeneity of the rock, which in itself ranges from 44 nanometers to kilometers (Ringrose & Bentley, 2016; Jackson et al., 2018). 45

This complexity makes modelling the flow and trapping of fluids in the subsurface 46 challenging, with uncertainty in which flow processes are important to characterise at 47 different spatial scales. For example, at the scale of a reservoir, many attempts to pre-48 dict CO_2 plume migration in the subsurface resulted with the CO_2 arriving earlier and 49 spreading out further than expected (Dance et al., 2019; Hosseini et al., 2013; Daley et 50 al., 2011; Ringrose et al., 2013). These analyses focused on characterising heterogene-51 ity in continuum-scale properties, like capillary pressure and relative permeability (Jackson 52 & Krevor, 2020). However, observations made at the pore-scale show dynamics that are 53 not incorporated within the framework of continuum-scale flow properties, such as in-54 termittent pathway flow and ganglion dynamics (Spurin et al., 2019a, 2019b; Rücker et 55 al., 2015; Gao et al., 2020; Avraam & Payatakes, 1995). Traditional continuum-scale mod-56 els relate flow rate linearly to an average pressure gradient across the system. They do 57

not account for any fluctuations in pressure, or a non-linear relationship between flow
rate and pressure gradient, both of which have been observed experimentally, and attributed to non-linear flow dynamics (Blunt, 2017; Muskat, 1938; Zhang et al., 2021).
These dynamics may play a role in large-scale flow properties, and will influence plume
migration (Spurin et al., 2020; Juanes et al., 2010; Zhang et al., 2021).

Micro-computed tomography (Micro-CT) experiments provide pore-scale observa-63 tions of fluid-fluid interfaces in situ at resolutions of a few microns. However, experimen-64 tal limitations including temporal resolution, expense, and management of vast quan-65 66 tities of data produced, mean it is currently infeasible to observe fluid-fluid interfaces at the centimetre to metre scale (the core-scale). Instead, medical CT scanners are used 67 to measure saturation distributions (Akin & Kovscek, 2003; Pini & Madonna, 2016; Krevor 68 et al., 2012). If observations of flow from pore-scale experiments are representative of 69 flow at larger scales they can, in principle, be used to understand results from core-scale 70 experiments. However, it is unclear if information about flow dynamics is being lost due 71 to the limited spatial and temporal scales of the pore-scale experiments, or if pore-scale 72 dynamics differ when sample size is increased. For example, viscous and gravity forces 73 may become more important at larger scales, even in capillary-dominated regimes. 74

Pore-scale and core-scale experiments have two overlapping quantities that are mea-75 sured: saturation and pressure. Saturation is important, as it can indicate the amount 76 of trapping, but without a measure of connectivity, it gives no indication of the under-77 lying dynamics. However, pressure fluctuations have been related to pore-scale dynam-78 ics and energy dissipation in the pore space through the creation and destruction of in-79 terfaces (Spurin et al., 2022; Rücker et al., 2021). In this work, we explore how pressure 80 fluctuations measured during core-scale experiments can be used to provide insight into 81 underlying flow dynamics by using continuous wavelet transforms to map the spectral 82 power of pressure data. We identify sources of spectral power as a function of time and 83 frequency. The merits of using pressure data to obtain information about multiphase flow 84 in porous media, including possible scaling relationships, are assessed. 85

86 2 Methods

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2.1 Experimental Procedure

The experiments in this work were conducted at two different scales: the pore-scale 88 and the core-scale. For the pore-scale investigation, the sample was a carbonate rock, 89 5 mm in diameter and 20 mm long. These experiments were conducted at a synchrotron 90 facility, so fluid interfaces could be resolved in real time (Spurin et al., 2020). There are 91 two experiments in the pore-scale investigation, which both explored the transition to 92 steady-state dynamics. One observes intermittent pathway flow through the co-injection 93 of gas and water, while the other observes connected pathway flow through the co-injection 94 of oil and water (Spurin et al., 2021, 2020). The capillary number, defined as $Ca = q/\sigma\lambda$ 95 where q is the flow rate, σ is the interfacial tension and λ is the mobility of the fluids 96 was 1.6×10^{-7} for the gas/water experiments and 2.2×10^{-6} for the oil/water exper-97 iments. See Spurin et al. (2020) for a full experimental description. 98

For the core-scale investigation, the sample was a carbonate rock, 5 cm in diam-99 eter and 12 cm long. The experiments were conducted in a medical CT scanner, so the 100 fluid interfaces themselves cannot be resolved, but the saturation across many pores is 101 measured (see Figure 1 for the difference in imaging resolution at the different scales). 102 Three experiments were performed to explore the transition to steady-state dynamics; 103 two explore the co-injection of gas and water. The same sample was used for both these 104 experiments, but the sample orientation was reversed between experiments, to explore 105 the role of heterogeneity of the pore space on flow dynamics. For the third experiment 106 oil and water were co-injected. Sample orientation was not reversed in this experiment 107

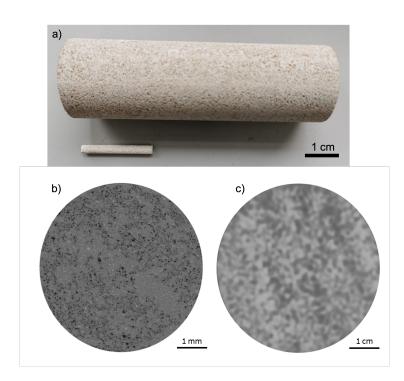


Figure 1. (a) The core-scale (large) sample shown alongside the pore-scale sample to highlight the difference in scale. (b) A CT slice through the pore-scale sample, where fluid interfaces are resolvable. (c) A CT slice through the core-scale sample, where fluid interfaces are not resolvable but grayscale values are proportion to saturation.

as oil is difficult to remove from a sample, which would have influenced the observations. The capillary number was 2.0×10^{-8} for the gas/water experiments and 5.4×10^{-7} for the oil/water experiment. With similar capillary numbers, we aimed to observe the same manifestation of the pore-scale dynamics in the pressure data as the pore-scale experiments. The full experimental procedure is provided in the Supplementary Material.

An example of the images taken during an experiment at each scale is shown in Fig-113 ure 1. It highlights the impact of the different imaging resolutions for the experiments, 114 and shows how connectivity of the fluid phases cannot be calculated from traditional med-115 ical CT imaging. Thus, due to these imaging constraints, the only parameters that are 116 constant across the experimental scales are saturation and pressure. The pressure drop 117 across the the sample with time is the parameter of interest in this work, and was recorded 118 for all experiments using a differential pressure transducer connected to the inlet line for 119 the water, and the outlet line for both phases. 120

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2.2 Spectral Analysis Using Wavelet Transformation

The spectral content of the pressure data was investigated by transforming it into 122 the frequency-time domain using a continuous wavelet transformation (CWT). This dif-123 fers from previous work using Fourier transformation of pressure data that revealed a 124 cascade of timescales for steady-state multiphase flow, with lower frequency events hav-125 ing larger amplitudes (Spurin et al., 2022). While insightful, Fourier transforms have some 126 limitations that make further analysis difficult. These include significant power spectral 127 leakage, noisy calculated power spectra, and the fact that stationary functions are un-128 likely to reflect changes in pressure as a result of flow, especially during transient flow, 129

when average pressure is a function of time. Mapping spectral power as a function of frequency and time might provide additional insight into the dynamics of the system

We use a transform that convolves a uniformly-sampled pressure data time series, 132 p_t , with a mother wavelet, ψ . Pressure time series, with constant sampling intervals of 133 either $\delta_t = 1.29$ seconds (pore-scale experiments; Figure 2) or 9.3 seconds (core-scale 134 experiments; Figures 3 & 4), were mirrored 7 times before being transformed to ame-135 liorate edge effects (Roberts et al., 2019). The Derivative-of-Gaussian (DOG) wavelet, 136 with derivative m = 6, was used as the mother wavelet in this study. It was scaled and 137 138 translated along the time series by t' to reveal variations in amplitude as a function of scale, s, and time, t. Thus, the wavelet transform, $W_t(s)$, has the form: 139

$$W_t(s) = \sum_{t'=0}^{N-1} p_t \psi^* \left[\frac{(t'-t)\delta t}{s} \right]$$
(1)

where ψ^* denotes the complex conjugate of the mother wavelet. N is the number 140 of discrete measurements of pressure. In this study, N = 385 for the gas/water core-141 scale experiments (total sampling duration ≈ 1 hour), for the oil/water core experiment 142 $N = 577 \ (\approx 1.5 \text{ hours})$. For the pore-scale experiments $N = 24,991 \ (\approx 9 \text{ hours})$ and 143 14,081 (≈ 4 hours) for gas/water and oil/water experiments, respectively. The code was 144 adapted from O'Malley and Roberts (2022), and based on the methods summarized by 145 Torrence and Compo (1998). The input signals can be recovered with errors less than 146 2.5% via the inverse transform, highlighting the fidelity of the transformations (see Sup-147 plementary Material). 148

The wavelet transform can be converted into power, ϕ , such that $\phi(t,s) = |W_t(s)|^2$. The time-averaged power spectrum is thus:

$$\phi(s) = \frac{1}{N} \sum_{t=0}^{N-1} |W_t(s)|^2.$$
(2)

Following (Liu et al., 2007), power is rectified by scale, and scales are converted into equivalent Fourier frequencies. The rectified time-averaged power spectra, $\phi_r = \phi(s)s^{-1}$, are consistent with results obtained from Fourier transformation of the time series.

Relationships between power spectral amplitudes and frequencies, f, provide insight into the scaling regimes and dynamics of many physical systems (Moura et al., 2017; Spurin et al., 2022; Rudnick & Davis, 2003; Fernandes et al., 2022; van der Schaaf et al., 2002). Many geophysical time series are characterised by:

$$\phi_r \propto f^{\alpha}.$$
 (3)

Determining the value(s) of α from the power spectra of time series can be a con-158 venient way to identify scaling regime(s). For example, $\alpha = -2$ indicates that a time 159 series can be characterized as red noise. If pressure time series are characterized by red 160 noise, it implies that the amplitudes of the pressure perturbations are proportional to 161 their duration. White noise, $\alpha = 0$, indicates that the amplitudes of pressure pertur-162 bations are roughly the same across all frequencies. A variety of other noise distributions 163 and changing patterns of spectral content can be straightforwardly identified by plot-164 ting power as a function of frequency in log-log space. For example, black, pink, and blue 165 noise have spectral slopes, α , of -3, -1 and 1, respectively. 166

¹⁶⁷ 3 Results and Discussion

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3.1 Sources of Spectral Power

There are many different potential sources for the spectral power in pressure time series during multiphase flow. The main ones identified here are (1) flow mechanisms (such as intermittent pathway flow or connected pathway flow), (2) heterogeneity of the pore space, and (3) the ratio of capillary to viscous forces.

In this research we focus on the flow mechanisms, and their representation in pres-173 sure signals in pore-scale experiments. This approach allows us to link spectral power 174 to different flow regimes. We explore if the spectral scalings obtained can be applied to 175 core-scale results to assess whether flow regimes can be deduced without pore-scale imag-176 ing. With the larger samples, we explore the role of heterogeneity on fluid flow by re-177 peating the experiment with the sample orientation reversed, so that the direction of flow 178 relative to the heterogeneity is reversed. Note that the degree and orientation of hetero-179 geneity is linked to the flow mechanisms (Spurin et al., 2019a), so it is non-trivial to iso-180 late them. With larger cores, viscous forces may also play a more important role. 181

3.2 Pore-Scale Results

The results for the pore-scale experiments are shown in Figure 2, with panels a-183 d showing the results for the gas/water experiment and panels e-h showing the results 184 for the oil/water experiment. Panels a and e in Figure 2 show the pressure drop across 185 the sample recorded during an experiment for gas/water and oil/water, respectively. The 186 shaded green strips correspond to the time intervals for the time-averaged power spec-187 tra shown in panels d and h, with a later time denoted by a darker shade. Note these 188 panels indicate ~ 1 hr intervals for the gas/water experiment, and ~ 30 min intervals 189 for the oil/water experiment because steady-state was reached quicker during the oil/water 190 experiment. Figure 2b and f show power spectra of pressure data with time for gas/water 191 and oil/water, respectively. Here, the dashed lines correspond to the shaded green strips 192 in panels a and e. Figure 2c-d and g-h show time-averaged power against frequency for 193 gas/water and oil/water, respectively. This is shown for the full recording window, and 194 the 1st and 2nd half of the pressure time series in Figure 2c and g, which can be com-195 pared to the evolution of the power spectra for shorter intervals in Figure 2d and h. 196

With the pore-scale experiments, we can relate power spectra to different flow regimes 197 observed during the experiments. For both experiments, the sample is initially saturated 198 with water. First, the non-wetting phase (the gas or oil) percolates the sample, result-199 ing in purely drainage events (gas or oil displacing the water). At approximately 20,000 s 200 for the gas/water experiment (Figure 2a) and 3,000 s for the oil/water experiment (Fig-201 ure 2e) the pressure plateaus, marking the transition to steady-state flow. For the gas/water 202 experiment this leads to intermittent pathway flow, where gas flow pathways repeatedly 203 connect and disconnect (Spurin et al., 2020). For the oil/water experiment no further 204 displacement events occur during steady-state flow; the fluids flow in their own separate 205 pathways that are connected across the pore space (Spurin et al., 2020). 206

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3.2.1 Intermittent Pathway vs Connected Pathway Flow

For the gas/water experiment fluid rearrangement events were larger and occurred 208 even during steady-state flow, while the oil/water experiment had little to no fluid re-209 arrangement once oil had percolated the sample (Spurin et al., 2020, 2022). The differ-210 211 ent flow regimes are evident in the pressure in Figure 2a and e. First, in the oil/water experiment the pressure overshoots the stabilisation pressure (at around 3,000 s in Fig-212 ure 2e), but then relaxes to approximately 65 kPa for the rest of the experiment. In the 213 gas/water experiment, the pressure builds more gradually and then plateaus at approx-214 imately 20,000 s in Figure 2a. There are significantly more fluctuations during the gas/water 215

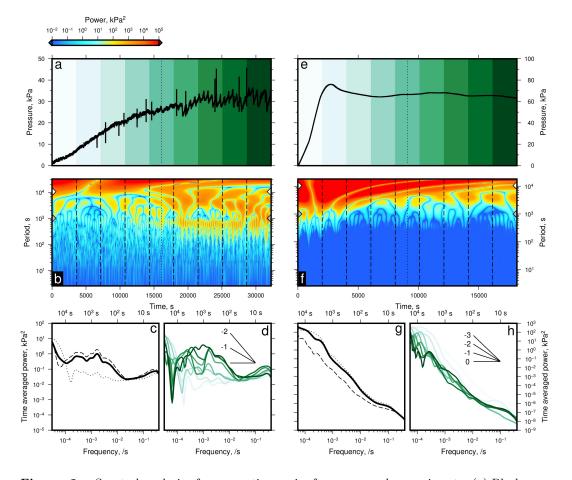


Figure 2. Spectral analysis of pressure time series from pore-scale experiments. (a) Black curve = pressure from gas/water experiment. Green strips and dotted line = time intervals indicated in panels b-d. (b) Power spectrum calculated by transforming black curve in panel a. Dashed and dotted lines correspond to time intervals indicated in panel a. Grey/white arrow heads indicate limit on low pass filters (10^3 and 10^4 s) discussed in body text. (c) Thick black curve = time-averaged, rectified, power spectrum for entire series. Dotted and dashed curves = time-averaged power for 1st and 2nd half of the time series, respectively (separated by dotted line in panels a and b). (d) Time-averaged power spectra for intervals indicated by green strips in panel a. Note graticule indicating red (-2), pink (-1) and white (0; flat) spectral slopes. (e-h) Results for the oil/water experiment.

experiment, even if the mean pressure remains constant, due to intermittent gas pathways periodically connecting and disconnecting.

Variations in the pressure time series are highlighted by the power spectra, pro-218 duced by the continuous wavelet transformation, shown in Figure 2b and f. Several re-219 sults are evident in the wavelet power spectra, which were not immediately obvious from 220 inspection of the pressure time series alone. First, pressure at the longer periods/lower 221 frequencies increases in power as the system transitions to steady-state for the exper-222 iment with intermittency (shown by the increase in power at periods of approximately 223 224 10^3 s in Figure 2b). This observation corresponds to the approximately 10 minute cycles observed in the pressure data in Figure 2a). These cycles were linked to disconnec-225 tion and re-connection events in a key location, controlling flow across the sample (Spurin 226 et al., 2020). 227

Secondly, pressure at shorter periods/higher frequencies contributes less to total power than the longer period fluctuations. Consider that inverse wavelet transforms produced including periods $> 10^3$ s have a mean error, which we define here as

$$\frac{1}{N\bar{p_t}} \sum_{t=1}^{N} \left[\left(p_t - p_{tf} \right)^2 \right]^{1/2} \times 100(\%), \tag{4}$$

where p_{tf} are pressures in the filtered series and \bar{p}_t is mean pressure of the unfil-231 tered series, of only 3% for the gas/water experiment (Figure 2b: grey arrow heads). The 232 mean error is even less (2%) for the oil/water experiment (Figure 2f: grey arrow heads). 233 Inverse transforms with only periods $> 10^4$ s included yield a mean error of 5% for the 234 gas/water experiment, and 8% for the oil/water experiment (white arrow heads in Fig-235 ure 2b and f, respectively). Shorter period variations in pressure ($< 10^3$ s), whilst still 236 providing relatively little overall power, account for a greater proportion of the total power 237 in the gas/water experiments compared to the oil/water experiment. In contrast, longer 238 periods $(> 10^4 \text{ s})$ contribute a relatively larger proportion of the total power for the oil/water 239 experiment. In summary, pressure fluctuations at short periods play a greater role in larger 240 scale flow properties in the gas/water experiment compared to in the oil/water exper-241 iment. This observation is indicative of the role that the pore-scale intermittency has 242 in enabling flow at relatively little energy cost (Spurin et al., 2021). 243

Finally, time-averaged power spectra (Figure 2c-d and g-h) show that, for the gas/water 244 experiment, different spectral slopes exist as the system evolves to steady-state conditions, transitioning from a slope of -1 (pink noise) to -2 (red noise) in Figure 2d, whilst 246 a roughly constant spectral slope exists at all times and across all timescales during the 247 oil/water experiment. This observation highlights the complexity of intermittent path-248 way flow, with events occurring over a wide range of frequencies, length-scales, and be-249 ing non-local in nature (Spurin et al., 2020). At steady-state, intermittent pathway flow 250 manifests as red noise (a spectral slope of -2) and connected pathway flow manifests as 251 a spectral slope of -3. A slope of -2 agrees with observations made using Fourier trans-252 formation on steady-state pressure data (Spurin et al., 2022). A slope of -3 is typical 253 for pseudo-turbulent flows (Mercado et al., 2010; Roghair et al., 2011; Mendez-Diaz et 254 al., 2013). These are flows that appear turbulent but are in fact the result of the com-255 plex interaction of fluids with the surrounding space (other fluids, and in this case, po-256 tentially the rock grains) instead of inertial forces (Mercado et al., 2010). Further research, 257 including velocity measurements are required to determine if pseudo-turbulence is oc-258 curring in multi-phase flow through porous media. 259

260 3.2.2 Possibility of Upscaling

For the oil/water experiment, where both fluids flowed in continuously connected pathways (as assumed in the multiphase extension of Darcy's law), a broadly constant spectral slope of -3 exists for all frequencies during transient and steady-state flow (Figure 2h). These observations imply that there is limited temporal evolution during connected pathway flow, creating less uncertainty in predictions made for periods outside the experimental observation window.

For the gas/water experiment, spectral slopes depend on frequency and time, which implies a change in dynamics at different periods and times (Figure 2d). Spectral slopes steepen at long $(> 10^4 \text{ s})$ and short $(< 10^3 \text{ s})$ periods as the system transitions to 'steadystate' flow. This higlights the presence of non-linear dynamics not included in the multiphase extension of Darcy's law. The presence of multiple spectral slopes makes simple upscaling of predictions challenging. Thus, the success of upscaling efforts depends on how the dynamics present manifest in larger samples.

3.3 Core-Scale Results

The core-scale experiments follow the same procedure as the pore-scale experiments, with the same fluid pairings. This allows us to establish if the pore-scale observations can be upscaled to the core-scale experiments typically used for subsurface characterization (Pini & Benson, 2013; Perrin et al., 2009; Ruprecht et al., 2014). Figure 3a-d shows the results for the gas/water experiment, e-h shows the results for the oil/water experiment and i-l shows the results for the gas/water experiment in which sample orientation was reversed.

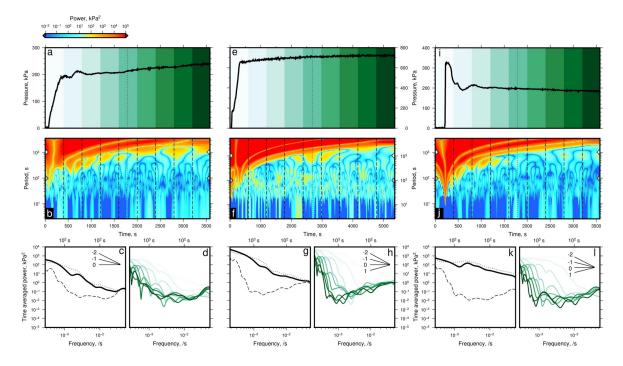


Figure 3. Spectral analysis of pressure time series from core-scale experiments. (a-d) Gas/water experiment initial sample orientation. (e-h) Oil/water experiment. (i-l) Gas/water experiment reversed sample orientation. Annotation is the same as Figure 2.

282 3.3.1 Gas/water vs Oil/water

The pressure response for the gas/water experiment and the oil/water experiment shown in Figure 3a and e appear similar in nature; in the first 500 s there is a steep increase in pressure as the gas or oil percolates the sample, then there is a gradual increase

in pressure with time, with pressure fluctuations of a similar magnitude (around 5 kPa). 286 The spectral power provides additional insight into evolution of pressure, and reveals sub-287 the differences between the experiments. First, power at longer periods (> 10^3 s) decreases 288 with time, as shown by the reduction in red colours with increasing time in Figure 3b 289 and f. Pressure at periods $> 10^3$ s contributes a similar proportion of power (mean er-290 ror $\sim 2\%$) in both the gas/water and oil/water experiments (white arrow heads in Fig-291 ure 3b and f). This implies that pressure at periods $> 10^3$ s contributes $\sim 98\%$ of to-292 tal power. 293

Power at frequencies of 3×10^{-2} to 2×10^{-3} Hz also decrease with time (Figure 3d & h). Spectral slopes at these frequencies are observed to flatten (i.e. whiten). While longer periods contain less power at later experimental times, they continue to contribute significantly to the total power, shown by the spectral slope steepening as the system evolves in time. Pressure at shorter periods (< 10³ s) contributes far less (~ 2%) to total power in both the oil/water and gas/water experiments.

When averaged over the entire experimental run time, as shown in Figure 3c and g, spectral power can be described by a single spectral slope of -2 i.e. red noise for both experiments. However, at the end of both experiments (the darkest green lines in Figure 3d and h) the time-averaged power requires two spectral slopes, and differ for gas/water and oil/water, with a steeper spectral slope for frequencies $> 10^{-3}$ Hz for the latter.

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3.3.2 The Role of Heterogeneity

The role of heterogeneity is evident in Figures 3a-d and i-l. In the experiment where the section of the sample with a lower porosity was closest to the inlet, there is an overshoot in pressure prior to stabilisation (seen in Figure 3i) at approximately 200 s. Whereas, when flow direction is reversed, there is a gradual increase in pressure, and no marked overshoot, as shown in Figure 3a. Both experiments reach the same differential pressure of approximately 200 kPa within 15 minutes of injection.

For both experiments, pressure at lower frequencies decreases in power with time 312 (shown in Figure 3d and l). Power is increasingly concentrated at lower frequencies dur-313 ing an experiment, regardless of the orientation of the sample. During steady-state flow, 314 the spectral slope for higher frequencies ($< 10^{-3}$ Hz) resembles white noise i.e. the am-315 plitude is independent of frequency, while it is steeper for the lower frequencies. The spec-316 tral slope is -2 (red noise) when averaged over the whole time series. However, the evo-317 lution of the flow properties is dependent on the heterogeneity of the pore space and its 318 orientation with respect to flow. This means that previous heterogeneity classifications, 319 that rely on average porosity and permeability values for the whole core, miss the im-320 portance of the orientation of heterogeneity with respect to the flow direction (Li & Ben-321 son, 2015; Ni et al., 2019). 322

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3.4 Core-scale vs Pore-scale Results

The magnitude of pressure fluctuations are similar for the core-scale and pore-scale experiments (they are on the order of 1–5 kPa). The total pressure drop is significantly higher for the core-scale experiments, meaning the pressure fluctuations are much smaller relative to the total pressure drop across the sample.

While the magnitude of pressure fluctuations are similar, spectral analysis reveals differences between the pore-scale and core-scale experiments. First, the single spectral slope observed in the pore-scale oil/water experiment is not observed in the core-scale oil/water experiment. While the spectral slope is approximately -3 for frequencies < 10^{-3} Hz during steady-state flow at the core-scale, it flattens to 0 (white noise), for the higher frequencies/shorter periods (Figure 3h). This suggests that the shorter periods are less significant in the core-scale experiments during steady-state flow. Secondly, at

periods 10^2 to 10^4 s, power increases in the pore-scale gas/water experiment, while the 335 shorter periods (down to ~ 10 s) remain approximately constant (Figure 2d). For the 336 core-scale gas/water experiments, power decreases across almost all periods (down to \sim 337 10 s), albeit at a slower rate. Thus the evolution of the time-averaged power spectra is 338 dependent on scale. While similar spectral slopes are observable (between -1 and -3), 339 these slopes are dependent on frequency and time. In all cases, the shorter timescales 340 play a more significant role during transient flow, but this significance decreases markedly 341 for the core-scale experiments, possibly due to viscous dampening. 342

For the larger, core-scale experiments, a single spectral slope (attributed to the flow regime assumed in the multiphase extension of Darcy's law) is not observed under any of the experimental conditions explored in this work. This result suggests that the onset of non-linear flow may occur at lower capillary numbers in larger samples. This assertion agrees with dynamic pore network modelling observations showing that the onset of non-linear flow regimes start at lower flow rates as system size increases (Hansen et al., 2023; Pedersen & Hansen, 2023).

350 4 Conclusions

In this work we used continuous wavelet transformation to investigate sources of 351 spectral power in the pressure time series of multiphase flow experiments. We showed 352 that spectral power is dependent on frequency, sample size, and the heterogeneity present. 353 Since pressure series spectral slopes are dependent on frequency and time, it is challeng-354 ing to extrapolate flow dynamics to larger spatial scales and longer temporal scales in 355 a straightforward way. However, we can relate spectral signals to dynamics in the pore 356 space. This shows how pressure time series can provide useful information about the un-357 derlying pore-scale dynamics at larger scales. Thus, an analysis of the pressure fluctu-358 ations is an important step to understanding larger scale flow processes. We showed it 359 is possible to gain new insights into the underlying flow regimes without recourse to novel 360 experimental techniques, or increased imaging capabilities. 361

Further work is needed to fully characterize the impact of heterogeneity, e.g. layering or different lithologies, on spectral power and associated scaling regimes. Experiments should be conducted at lower flow rates, and different fractional flows to ascertain if the connected pathway flow signal observed for the pore-scale results is possible in core-scale experiments. Further work is also needed to create analytical spectra from physical models, to increase the applicability of the findings made in this work to other flow regimes and different samples.

- ³⁶⁹ 5 Open Research
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The pressure data shown in this work is attached in the Supporting Information.

The continuous wavelet transformation analysis is available on Github: https:// github.com/Malley1/Wavelets-pycwt-wrapper.

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Pore-scale fluid dynamics resolved in pressure fluctuations at the Darcy scale

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5 6 7 Catherine Spurin¹, Gareth G. Roberts², Conor P. B. O'Malley², Takeshi Kurotori^{1,3}, Samuel Krevor², Martin J. Blunt², and Hamdi Tchelepi¹

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8 Abstract

Complex flow dynamics have been observed, at the pore-scale, during multiphase through q porous rocks. These dynamics are not captured in large scale models exploring the mi-10 gration and trapping of subsurface fluids e.g., CO_2 or hydrogen. Due to limitations in 11 imaging capabilities, these dynamics cannot be observed directly at the larger, Darcy 12 scale. Instead, by using pressure data from pore-scale (mm-scale) and core-scale (cm-13 scale) experiments, we show that fluctuations in pressure measured at the core-scale re-14 flect specific fluid displacement events taking place at the pore-scale. The spectral char-15 acteristics of the pressure data depends on the flow dynamics, size of the rock sample, 16 and heterogeneity of pore space. While high resolution imaging of large samples would 17 be useful in assessing flow dynamics across many of the scales of interest, such an ap-18 proach is currently infeasible. We suggest an alternative, pragmatic, approach examin-19 ing pressure data in the time-frequency domain using wavelet transformation. 20

21 Plain Language Summary

Complex fluid dynamics have been observed in small pores within rocks. These dy-22 namics have not been accounted for in larger scale modelling efforts of CO₂ or hydro-23 gen. Limitations in imaging prevent the direct observation of these dynamic at larger 24 scales, creating uncertainty in how these dynamics manifest at larger scales. But by an-25 alyzing the pressure data, and fluctuations in pressure measurements, we can infer the 26 27 small-scale dynamics without imaging. We apply our findings to larger samples and discover that the fluctuations are dependent on the type of flow dynamics occurring, sam-28 ple size, and the composition of the sample. We present a practical approach for assess-20 ing the dynamics at the larger scale, where direct imaging is currently infeasible, by ex-30 ploring the pressure data using a technique called continuous wavelet transformation. 31

32 1 Introduction

Fluid flow in the subsurface is a complex process, controlled by the interaction of 33 multiple fluids with one another, and a heterogeneous pore space. It is central to the safe 34 storage of CO_2 in the subsurface (Rubin & De Coninck, 2005; Bachu & Adams, 2003; 35 Benson et al., 2012), and the storage and retrieval of hydrogen underground (Boon & 36 Hajibeygi, 2022; Thiyagarajan et al., 2022), as examples. Depending on the flow rate, 37 fluid viscosity, wettability of the fluids, and connectivity of the pore space, different flow 38 mechanisms will prevail (Blunt, 2017; Lenormand et al., 1983; Avraam & Payatakes, 1995; 39 Spurin et al., 2019a, 2019b; Rücker et al., 2015; Zou et al., 2018; Zhao et al., 2016). These 40 processes span many orders of magnitudes for both timescales and length scales, from 41 sub-second to hours (Berg et al., 2013; Schlüter et al., 2017; McClure et al., 2020), and 42 sub-pore to multi-pore (Berg et al., 2013; Moebius & Or, 2014; Spurin et al., 2020). Fluid 43 flow is also heavily controlled by the heterogeneity of the rock, which in itself ranges from 44 nanometers to kilometers (Ringrose & Bentley, 2016; Jackson et al., 2018). 45

This complexity makes modelling the flow and trapping of fluids in the subsurface 46 challenging, with uncertainty in which flow processes are important to characterise at 47 different spatial scales. For example, at the scale of a reservoir, many attempts to pre-48 dict CO_2 plume migration in the subsurface resulted with the CO_2 arriving earlier and 49 spreading out further than expected (Dance et al., 2019; Hosseini et al., 2013; Daley et 50 al., 2011; Ringrose et al., 2013). These analyses focused on characterising heterogene-51 ity in continuum-scale properties, like capillary pressure and relative permeability (Jackson 52 & Krevor, 2020). However, observations made at the pore-scale show dynamics that are 53 not incorporated within the framework of continuum-scale flow properties, such as in-54 termittent pathway flow and ganglion dynamics (Spurin et al., 2019a, 2019b; Rücker et 55 al., 2015; Gao et al., 2020; Avraam & Payatakes, 1995). Traditional continuum-scale mod-56 els relate flow rate linearly to an average pressure gradient across the system. They do 57

not account for any fluctuations in pressure, or a non-linear relationship between flow
rate and pressure gradient, both of which have been observed experimentally, and attributed to non-linear flow dynamics (Blunt, 2017; Muskat, 1938; Zhang et al., 2021).
These dynamics may play a role in large-scale flow properties, and will influence plume
migration (Spurin et al., 2020; Juanes et al., 2010; Zhang et al., 2021).

Micro-computed tomography (Micro-CT) experiments provide pore-scale observa-63 tions of fluid-fluid interfaces in situ at resolutions of a few microns. However, experimen-64 tal limitations including temporal resolution, expense, and management of vast quan-65 66 tities of data produced, mean it is currently infeasible to observe fluid-fluid interfaces at the centimetre to metre scale (the core-scale). Instead, medical CT scanners are used 67 to measure saturation distributions (Akin & Kovscek, 2003; Pini & Madonna, 2016; Krevor 68 et al., 2012). If observations of flow from pore-scale experiments are representative of 69 flow at larger scales they can, in principle, be used to understand results from core-scale 70 experiments. However, it is unclear if information about flow dynamics is being lost due 71 to the limited spatial and temporal scales of the pore-scale experiments, or if pore-scale 72 dynamics differ when sample size is increased. For example, viscous and gravity forces 73 may become more important at larger scales, even in capillary-dominated regimes. 74

Pore-scale and core-scale experiments have two overlapping quantities that are mea-75 sured: saturation and pressure. Saturation is important, as it can indicate the amount 76 of trapping, but without a measure of connectivity, it gives no indication of the under-77 lying dynamics. However, pressure fluctuations have been related to pore-scale dynam-78 ics and energy dissipation in the pore space through the creation and destruction of in-79 terfaces (Spurin et al., 2022; Rücker et al., 2021). In this work, we explore how pressure 80 fluctuations measured during core-scale experiments can be used to provide insight into 81 underlying flow dynamics by using continuous wavelet transforms to map the spectral 82 power of pressure data. We identify sources of spectral power as a function of time and 83 frequency. The merits of using pressure data to obtain information about multiphase flow 84 in porous media, including possible scaling relationships, are assessed. 85

86 2 Methods

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2.1 Experimental Procedure

The experiments in this work were conducted at two different scales: the pore-scale 88 and the core-scale. For the pore-scale investigation, the sample was a carbonate rock, 89 5 mm in diameter and 20 mm long. These experiments were conducted at a synchrotron 90 facility, so fluid interfaces could be resolved in real time (Spurin et al., 2020). There are 91 two experiments in the pore-scale investigation, which both explored the transition to 92 steady-state dynamics. One observes intermittent pathway flow through the co-injection 93 of gas and water, while the other observes connected pathway flow through the co-injection 94 of oil and water (Spurin et al., 2021, 2020). The capillary number, defined as $Ca = q/\sigma\lambda$ 95 where q is the flow rate, σ is the interfacial tension and λ is the mobility of the fluids 96 was 1.6×10^{-7} for the gas/water experiments and 2.2×10^{-6} for the oil/water exper-97 iments. See Spurin et al. (2020) for a full experimental description. 98

For the core-scale investigation, the sample was a carbonate rock, 5 cm in diam-99 eter and 12 cm long. The experiments were conducted in a medical CT scanner, so the 100 fluid interfaces themselves cannot be resolved, but the saturation across many pores is 101 measured (see Figure 1 for the difference in imaging resolution at the different scales). 102 Three experiments were performed to explore the transition to steady-state dynamics; 103 two explore the co-injection of gas and water. The same sample was used for both these 104 experiments, but the sample orientation was reversed between experiments, to explore 105 the role of heterogeneity of the pore space on flow dynamics. For the third experiment 106 oil and water were co-injected. Sample orientation was not reversed in this experiment 107

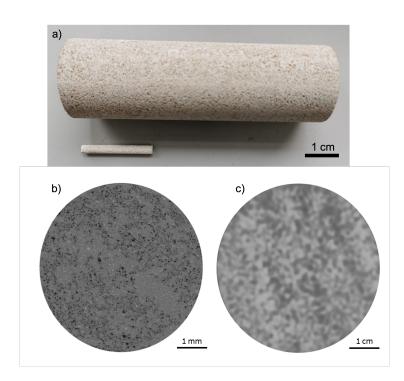


Figure 1. (a) The core-scale (large) sample shown alongside the pore-scale sample to highlight the difference in scale. (b) A CT slice through the pore-scale sample, where fluid interfaces are resolvable. (c) A CT slice through the core-scale sample, where fluid interfaces are not resolvable but grayscale values are proportion to saturation.

as oil is difficult to remove from a sample, which would have influenced the observations. The capillary number was 2.0×10^{-8} for the gas/water experiments and 5.4×10^{-7} for the oil/water experiment. With similar capillary numbers, we aimed to observe the same manifestation of the pore-scale dynamics in the pressure data as the pore-scale experiments. The full experimental procedure is provided in the Supplementary Material.

An example of the images taken during an experiment at each scale is shown in Fig-113 ure 1. It highlights the impact of the different imaging resolutions for the experiments, 114 and shows how connectivity of the fluid phases cannot be calculated from traditional med-115 ical CT imaging. Thus, due to these imaging constraints, the only parameters that are 116 constant across the experimental scales are saturation and pressure. The pressure drop 117 across the the sample with time is the parameter of interest in this work, and was recorded 118 for all experiments using a differential pressure transducer connected to the inlet line for 119 the water, and the outlet line for both phases. 120

121

2.2 Spectral Analysis Using Wavelet Transformation

The spectral content of the pressure data was investigated by transforming it into 122 the frequency-time domain using a continuous wavelet transformation (CWT). This dif-123 fers from previous work using Fourier transformation of pressure data that revealed a 124 cascade of timescales for steady-state multiphase flow, with lower frequency events hav-125 ing larger amplitudes (Spurin et al., 2022). While insightful, Fourier transforms have some 126 limitations that make further analysis difficult. These include significant power spectral 127 leakage, noisy calculated power spectra, and the fact that stationary functions are un-128 likely to reflect changes in pressure as a result of flow, especially during transient flow, 129

when average pressure is a function of time. Mapping spectral power as a function of frequency and time might provide additional insight into the dynamics of the system

We use a transform that convolves a uniformly-sampled pressure data time series, 132 p_t , with a mother wavelet, ψ . Pressure time series, with constant sampling intervals of 133 either $\delta_t = 1.29$ seconds (pore-scale experiments; Figure 2) or 9.3 seconds (core-scale 134 experiments; Figures 3 & 4), were mirrored 7 times before being transformed to ame-135 liorate edge effects (Roberts et al., 2019). The Derivative-of-Gaussian (DOG) wavelet, 136 with derivative m = 6, was used as the mother wavelet in this study. It was scaled and 137 138 translated along the time series by t' to reveal variations in amplitude as a function of scale, s, and time, t. Thus, the wavelet transform, $W_t(s)$, has the form: 139

$$W_t(s) = \sum_{t'=0}^{N-1} p_t \psi^* \left[\frac{(t'-t)\delta t}{s} \right]$$
(1)

where ψ^* denotes the complex conjugate of the mother wavelet. N is the number 140 of discrete measurements of pressure. In this study, N = 385 for the gas/water core-141 scale experiments (total sampling duration ≈ 1 hour), for the oil/water core experiment 142 $N = 577 \ (\approx 1.5 \text{ hours})$. For the pore-scale experiments $N = 24,991 \ (\approx 9 \text{ hours})$ and 143 14,081 (≈ 4 hours) for gas/water and oil/water experiments, respectively. The code was 144 adapted from O'Malley and Roberts (2022), and based on the methods summarized by 145 Torrence and Compo (1998). The input signals can be recovered with errors less than 146 2.5% via the inverse transform, highlighting the fidelity of the transformations (see Sup-147 plementary Material). 148

The wavelet transform can be converted into power, ϕ , such that $\phi(t,s) = |W_t(s)|^2$. The time-averaged power spectrum is thus:

$$\phi(s) = \frac{1}{N} \sum_{t=0}^{N-1} |W_t(s)|^2.$$
(2)

Following (Liu et al., 2007), power is rectified by scale, and scales are converted into equivalent Fourier frequencies. The rectified time-averaged power spectra, $\phi_r = \phi(s)s^{-1}$, are consistent with results obtained from Fourier transformation of the time series.

Relationships between power spectral amplitudes and frequencies, f, provide insight into the scaling regimes and dynamics of many physical systems (Moura et al., 2017; Spurin et al., 2022; Rudnick & Davis, 2003; Fernandes et al., 2022; van der Schaaf et al., 2002). Many geophysical time series are characterised by:

$$\phi_r \propto f^{\alpha}.$$
 (3)

Determining the value(s) of α from the power spectra of time series can be a con-158 venient way to identify scaling regime(s). For example, $\alpha = -2$ indicates that a time 159 series can be characterized as red noise. If pressure time series are characterized by red 160 noise, it implies that the amplitudes of the pressure perturbations are proportional to 161 their duration. White noise, $\alpha = 0$, indicates that the amplitudes of pressure pertur-162 bations are roughly the same across all frequencies. A variety of other noise distributions 163 and changing patterns of spectral content can be straightforwardly identified by plot-164 ting power as a function of frequency in log-log space. For example, black, pink, and blue 165 noise have spectral slopes, α , of -3, -1 and 1, respectively. 166

¹⁶⁷ 3 Results and Discussion

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3.1 Sources of Spectral Power

There are many different potential sources for the spectral power in pressure time series during multiphase flow. The main ones identified here are (1) flow mechanisms (such as intermittent pathway flow or connected pathway flow), (2) heterogeneity of the pore space, and (3) the ratio of capillary to viscous forces.

In this research we focus on the flow mechanisms, and their representation in pres-173 sure signals in pore-scale experiments. This approach allows us to link spectral power 174 to different flow regimes. We explore if the spectral scalings obtained can be applied to 175 core-scale results to assess whether flow regimes can be deduced without pore-scale imag-176 ing. With the larger samples, we explore the role of heterogeneity on fluid flow by re-177 peating the experiment with the sample orientation reversed, so that the direction of flow 178 relative to the heterogeneity is reversed. Note that the degree and orientation of hetero-179 geneity is linked to the flow mechanisms (Spurin et al., 2019a), so it is non-trivial to iso-180 late them. With larger cores, viscous forces may also play a more important role. 181

3.2 Pore-Scale Results

The results for the pore-scale experiments are shown in Figure 2, with panels a-183 d showing the results for the gas/water experiment and panels e-h showing the results 184 for the oil/water experiment. Panels a and e in Figure 2 show the pressure drop across 185 the sample recorded during an experiment for gas/water and oil/water, respectively. The 186 shaded green strips correspond to the time intervals for the time-averaged power spec-187 tra shown in panels d and h, with a later time denoted by a darker shade. Note these 188 panels indicate ~ 1 hr intervals for the gas/water experiment, and ~ 30 min intervals 189 for the oil/water experiment because steady-state was reached quicker during the oil/water 190 experiment. Figure 2b and f show power spectra of pressure data with time for gas/water 191 and oil/water, respectively. Here, the dashed lines correspond to the shaded green strips 192 in panels a and e. Figure 2c-d and g-h show time-averaged power against frequency for 193 gas/water and oil/water, respectively. This is shown for the full recording window, and 194 the 1st and 2nd half of the pressure time series in Figure 2c and g, which can be com-195 pared to the evolution of the power spectra for shorter intervals in Figure 2d and h. 196

With the pore-scale experiments, we can relate power spectra to different flow regimes 197 observed during the experiments. For both experiments, the sample is initially saturated 198 with water. First, the non-wetting phase (the gas or oil) percolates the sample, result-199 ing in purely drainage events (gas or oil displacing the water). At approximately 20,000 s 200 for the gas/water experiment (Figure 2a) and 3,000 s for the oil/water experiment (Fig-201 ure 2e) the pressure plateaus, marking the transition to steady-state flow. For the gas/water 202 experiment this leads to intermittent pathway flow, where gas flow pathways repeatedly 203 connect and disconnect (Spurin et al., 2020). For the oil/water experiment no further 204 displacement events occur during steady-state flow; the fluids flow in their own separate 205 pathways that are connected across the pore space (Spurin et al., 2020). 206

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3.2.1 Intermittent Pathway vs Connected Pathway Flow

For the gas/water experiment fluid rearrangement events were larger and occurred 208 even during steady-state flow, while the oil/water experiment had little to no fluid re-209 arrangement once oil had percolated the sample (Spurin et al., 2020, 2022). The differ-210 211 ent flow regimes are evident in the pressure in Figure 2a and e. First, in the oil/water experiment the pressure overshoots the stabilisation pressure (at around 3,000 s in Fig-212 ure 2e), but then relaxes to approximately 65 kPa for the rest of the experiment. In the 213 gas/water experiment, the pressure builds more gradually and then plateaus at approx-214 imately 20,000 s in Figure 2a. There are significantly more fluctuations during the gas/water 215

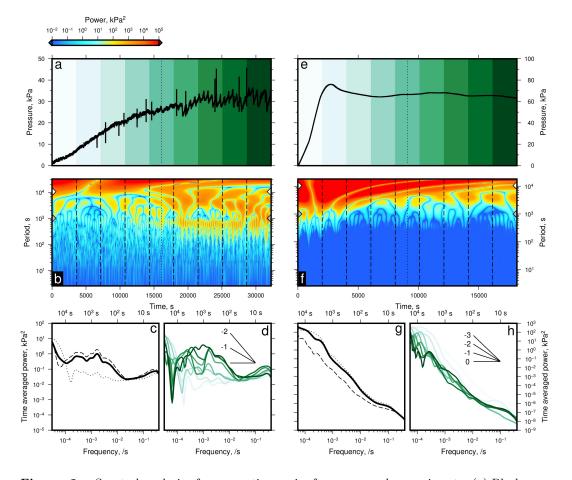


Figure 2. Spectral analysis of pressure time series from pore-scale experiments. (a) Black curve = pressure from gas/water experiment. Green strips and dotted line = time intervals indicated in panels b-d. (b) Power spectrum calculated by transforming black curve in panel a. Dashed and dotted lines correspond to time intervals indicated in panel a. Grey/white arrow heads indicate limit on low pass filters (10^3 and 10^4 s) discussed in body text. (c) Thick black curve = time-averaged, rectified, power spectrum for entire series. Dotted and dashed curves = time-averaged power for 1st and 2nd half of the time series, respectively (separated by dotted line in panels a and b). (d) Time-averaged power spectra for intervals indicated by green strips in panel a. Note graticule indicating red (-2), pink (-1) and white (0; flat) spectral slopes. (e-h) Results for the oil/water experiment.

experiment, even if the mean pressure remains constant, due to intermittent gas pathways periodically connecting and disconnecting.

Variations in the pressure time series are highlighted by the power spectra, pro-218 duced by the continuous wavelet transformation, shown in Figure 2b and f. Several re-219 sults are evident in the wavelet power spectra, which were not immediately obvious from 220 inspection of the pressure time series alone. First, pressure at the longer periods/lower 221 frequencies increases in power as the system transitions to steady-state for the exper-222 iment with intermittency (shown by the increase in power at periods of approximately 223 224 10^3 s in Figure 2b). This observation corresponds to the approximately 10 minute cycles observed in the pressure data in Figure 2a). These cycles were linked to disconnec-225 tion and re-connection events in a key location, controlling flow across the sample (Spurin 226 et al., 2020). 227

Secondly, pressure at shorter periods/higher frequencies contributes less to total power than the longer period fluctuations. Consider that inverse wavelet transforms produced including periods $> 10^3$ s have a mean error, which we define here as

$$\frac{1}{N\bar{p_t}} \sum_{t=1}^{N} \left[\left(p_t - p_{tf} \right)^2 \right]^{1/2} \times 100(\%), \tag{4}$$

where p_{tf} are pressures in the filtered series and \bar{p}_t is mean pressure of the unfil-231 tered series, of only 3% for the gas/water experiment (Figure 2b: grey arrow heads). The 232 mean error is even less (2%) for the oil/water experiment (Figure 2f: grey arrow heads). 233 Inverse transforms with only periods $> 10^4$ s included yield a mean error of 5% for the 234 gas/water experiment, and 8% for the oil/water experiment (white arrow heads in Fig-235 ure 2b and f, respectively). Shorter period variations in pressure ($< 10^3$ s), whilst still 236 providing relatively little overall power, account for a greater proportion of the total power 237 in the gas/water experiments compared to the oil/water experiment. In contrast, longer 238 periods $(> 10^4 \text{ s})$ contribute a relatively larger proportion of the total power for the oil/water 239 experiment. In summary, pressure fluctuations at short periods play a greater role in larger 240 scale flow properties in the gas/water experiment compared to in the oil/water exper-241 iment. This observation is indicative of the role that the pore-scale intermittency has 242 in enabling flow at relatively little energy cost (Spurin et al., 2021). 243

Finally, time-averaged power spectra (Figure 2c-d and g-h) show that, for the gas/water 244 experiment, different spectral slopes exist as the system evolves to steady-state conditions, transitioning from a slope of -1 (pink noise) to -2 (red noise) in Figure 2d, whilst 246 a roughly constant spectral slope exists at all times and across all timescales during the 247 oil/water experiment. This observation highlights the complexity of intermittent path-248 way flow, with events occurring over a wide range of frequencies, length-scales, and be-249 ing non-local in nature (Spurin et al., 2020). At steady-state, intermittent pathway flow 250 manifests as red noise (a spectral slope of -2) and connected pathway flow manifests as 251 a spectral slope of -3. A slope of -2 agrees with observations made using Fourier trans-252 formation on steady-state pressure data (Spurin et al., 2022). A slope of -3 is typical 253 for pseudo-turbulent flows (Mercado et al., 2010; Roghair et al., 2011; Mendez-Diaz et 254 al., 2013). These are flows that appear turbulent but are in fact the result of the com-255 plex interaction of fluids with the surrounding space (other fluids, and in this case, po-256 tentially the rock grains) instead of inertial forces (Mercado et al., 2010). Further research, 257 including velocity measurements are required to determine if pseudo-turbulence is oc-258 curring in multi-phase flow through porous media. 259

260 3.2.2 Possibility of Upscaling

For the oil/water experiment, where both fluids flowed in continuously connected pathways (as assumed in the multiphase extension of Darcy's law), a broadly constant spectral slope of -3 exists for all frequencies during transient and steady-state flow (Figure 2h). These observations imply that there is limited temporal evolution during connected pathway flow, creating less uncertainty in predictions made for periods outside the experimental observation window.

For the gas/water experiment, spectral slopes depend on frequency and time, which implies a change in dynamics at different periods and times (Figure 2d). Spectral slopes steepen at long $(> 10^4 \text{ s})$ and short $(< 10^3 \text{ s})$ periods as the system transitions to 'steadystate' flow. This higlights the presence of non-linear dynamics not included in the multiphase extension of Darcy's law. The presence of multiple spectral slopes makes simple upscaling of predictions challenging. Thus, the success of upscaling efforts depends on how the dynamics present manifest in larger samples.

3.3 Core-Scale Results

The core-scale experiments follow the same procedure as the pore-scale experiments, with the same fluid pairings. This allows us to establish if the pore-scale observations can be upscaled to the core-scale experiments typically used for subsurface characterization (Pini & Benson, 2013; Perrin et al., 2009; Ruprecht et al., 2014). Figure 3a-d shows the results for the gas/water experiment, e-h shows the results for the oil/water experiment and i-l shows the results for the gas/water experiment in which sample orientation was reversed.

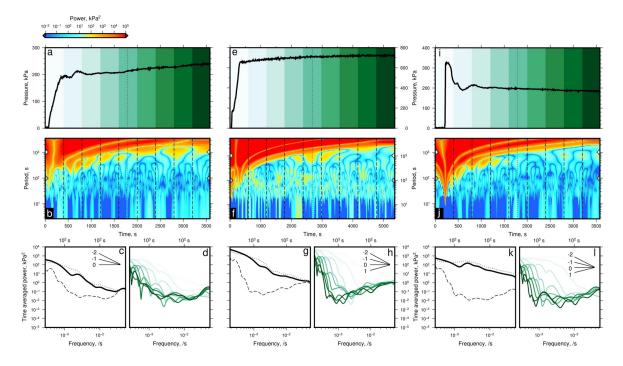


Figure 3. Spectral analysis of pressure time series from core-scale experiments. (a-d) Gas/water experiment initial sample orientation. (e-h) Oil/water experiment. (i-l) Gas/water experiment reversed sample orientation. Annotation is the same as Figure 2.

282 3.3.1 Gas/water vs Oil/water

The pressure response for the gas/water experiment and the oil/water experiment shown in Figure 3a and e appear similar in nature; in the first 500 s there is a steep increase in pressure as the gas or oil percolates the sample, then there is a gradual increase

in pressure with time, with pressure fluctuations of a similar magnitude (around 5 kPa). 286 The spectral power provides additional insight into evolution of pressure, and reveals sub-287 the differences between the experiments. First, power at longer periods (> 10^3 s) decreases 288 with time, as shown by the reduction in red colours with increasing time in Figure 3b 289 and f. Pressure at periods $> 10^3$ s contributes a similar proportion of power (mean er-290 ror $\sim 2\%$) in both the gas/water and oil/water experiments (white arrow heads in Fig-291 ure 3b and f). This implies that pressure at periods $> 10^3$ s contributes $\sim 98\%$ of to-292 tal power. 293

Power at frequencies of 3×10^{-2} to 2×10^{-3} Hz also decrease with time (Figure 3d & h). Spectral slopes at these frequencies are observed to flatten (i.e. whiten). While longer periods contain less power at later experimental times, they continue to contribute significantly to the total power, shown by the spectral slope steepening as the system evolves in time. Pressure at shorter periods (< 10³ s) contributes far less (~ 2%) to total power in both the oil/water and gas/water experiments.

When averaged over the entire experimental run time, as shown in Figure 3c and g, spectral power can be described by a single spectral slope of -2 i.e. red noise for both experiments. However, at the end of both experiments (the darkest green lines in Figure 3d and h) the time-averaged power requires two spectral slopes, and differ for gas/water and oil/water, with a steeper spectral slope for frequencies $> 10^{-3}$ Hz for the latter.

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3.3.2 The Role of Heterogeneity

The role of heterogeneity is evident in Figures 3a-d and i-l. In the experiment where the section of the sample with a lower porosity was closest to the inlet, there is an overshoot in pressure prior to stabilisation (seen in Figure 3i) at approximately 200 s. Whereas, when flow direction is reversed, there is a gradual increase in pressure, and no marked overshoot, as shown in Figure 3a. Both experiments reach the same differential pressure of approximately 200 kPa within 15 minutes of injection.

For both experiments, pressure at lower frequencies decreases in power with time 312 (shown in Figure 3d and l). Power is increasingly concentrated at lower frequencies dur-313 ing an experiment, regardless of the orientation of the sample. During steady-state flow, 314 the spectral slope for higher frequencies ($< 10^{-3}$ Hz) resembles white noise i.e. the am-315 plitude is independent of frequency, while it is steeper for the lower frequencies. The spec-316 tral slope is -2 (red noise) when averaged over the whole time series. However, the evo-317 lution of the flow properties is dependent on the heterogeneity of the pore space and its 318 orientation with respect to flow. This means that previous heterogeneity classifications, 319 that rely on average porosity and permeability values for the whole core, miss the im-320 portance of the orientation of heterogeneity with respect to the flow direction (Li & Ben-321 son, 2015; Ni et al., 2019). 322

323

3.4 Core-scale vs Pore-scale Results

The magnitude of pressure fluctuations are similar for the core-scale and pore-scale experiments (they are on the order of 1–5 kPa). The total pressure drop is significantly higher for the core-scale experiments, meaning the pressure fluctuations are much smaller relative to the total pressure drop across the sample.

While the magnitude of pressure fluctuations are similar, spectral analysis reveals differences between the pore-scale and core-scale experiments. First, the single spectral slope observed in the pore-scale oil/water experiment is not observed in the core-scale oil/water experiment. While the spectral slope is approximately -3 for frequencies < 10^{-3} Hz during steady-state flow at the core-scale, it flattens to 0 (white noise), for the higher frequencies/shorter periods (Figure 3h). This suggests that the shorter periods are less significant in the core-scale experiments during steady-state flow. Secondly, at

periods 10^2 to 10^4 s, power increases in the pore-scale gas/water experiment, while the 335 shorter periods (down to ~ 10 s) remain approximately constant (Figure 2d). For the 336 core-scale gas/water experiments, power decreases across almost all periods (down to \sim 337 10 s), albeit at a slower rate. Thus the evolution of the time-averaged power spectra is 338 dependent on scale. While similar spectral slopes are observable (between -1 and -3), 339 these slopes are dependent on frequency and time. In all cases, the shorter timescales 340 play a more significant role during transient flow, but this significance decreases markedly 341 for the core-scale experiments, possibly due to viscous dampening. 342

For the larger, core-scale experiments, a single spectral slope (attributed to the flow regime assumed in the multiphase extension of Darcy's law) is not observed under any of the experimental conditions explored in this work. This result suggests that the onset of non-linear flow may occur at lower capillary numbers in larger samples. This assertion agrees with dynamic pore network modelling observations showing that the onset of non-linear flow regimes start at lower flow rates as system size increases (Hansen et al., 2023; Pedersen & Hansen, 2023).

350 4 Conclusions

In this work we used continuous wavelet transformation to investigate sources of 351 spectral power in the pressure time series of multiphase flow experiments. We showed 352 that spectral power is dependent on frequency, sample size, and the heterogeneity present. 353 Since pressure series spectral slopes are dependent on frequency and time, it is challeng-354 ing to extrapolate flow dynamics to larger spatial scales and longer temporal scales in 355 a straightforward way. However, we can relate spectral signals to dynamics in the pore 356 space. This shows how pressure time series can provide useful information about the un-357 derlying pore-scale dynamics at larger scales. Thus, an analysis of the pressure fluctu-358 ations is an important step to understanding larger scale flow processes. We showed it 359 is possible to gain new insights into the underlying flow regimes without recourse to novel 360 experimental techniques, or increased imaging capabilities. 361

Further work is needed to fully characterize the impact of heterogeneity, e.g. layering or different lithologies, on spectral power and associated scaling regimes. Experiments should be conducted at lower flow rates, and different fractional flows to ascertain if the connected pathway flow signal observed for the pore-scale results is possible in core-scale experiments. Further work is also needed to create analytical spectra from physical models, to increase the applicability of the findings made in this work to other flow regimes and different samples.

- ³⁶⁹ 5 Open Research
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The pressure data shown in this work is attached in the Supporting Information.

The continuous wavelet transformation analysis is available on Github: https:// github.com/Malley1/Wavelets-pycwt-wrapper.

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Supplementary Material for: Pore-scale fluid dynamics resolved
 in pressure fluctuations at the Darcy scale
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Experiment	Flow rate NWP (ml/min)	Flow rate water (ml/min)	Total flow rate (ml/min)	Ca number
gas/water pore	0.015	0.085	0.1	1.6×10^{-7}
oil/water pore	0.05	0.05	0.1	2.2×10^{-6}
gas/water core	5	1	6	2.0×10^{-8}
oil/water core	3	0.6	3.6	5.4×10^{-7}

TABLE I: Flow rates and Capillary numbers for the experiments used in this work. The Capillary number $Ca = \frac{q}{\sigma\lambda}$, as defined by [1].

5 I. CORE-SCALE EXPERIMENTAL METHODOLOGY

Prior to any injection, the sample is loaded into a core holder that allows pressurisation
of the fluids, and placed in the medical CT scanner used to image the core and the fluid
distributions within. A confining pressure is applied that is always 2 MPa above the pressure
in the core. Before an experiment begins the sample is filled with water and pressurised to
8 MPa (the outlet of the core is held constant at 8 MPa during an experiment).

For the co-injection experiments, a drainage sequence is performed with both fluids (gas or oil, and water) injected at a constant flow rate, and fractional flow, for 60 minutes. The flow rates chosen are given in Table I. The flow rates were chosen to observe flow in the capillary dominated regime, and to have similar capillary numbers for the different fluid pairings. CT imaging occurred during flow, and at the end of both the drainage and imbibition cycles.

Between the gas/water experiments the core is depressurised and flushed with water, before the system is re-pressurised. This removes gas from the system, as confirmed by the wet scans prior to next experiment. For the oil/water experiment, the sample orientation was not reversed as it is difficult to completely remove oil from the sample.

20 II. CONTINUOUS WAVELET TRANSFORMATION (CWT) FIDELITY

To test the fidelity of the transformations, we calculated inverse wavelet transforms using all 21 frequencies and using only low frequencies (a form of low-pass filtering). Error was < 2.5%22 for full inverse transforms (i.e. including all frequencies; see Equation 4). Filtered inverse 23 wavelet transforms that only include low frequency contributions to the signals are shown 24 in Figure 1 and discussed in the body text of the main manuscript. These results indicate 25 that continuous wavelet transforms can yield fair representations of pressure series in the 26 time-frequency domain, and that nearly all signal power is concentrated at low frequencies 27 (see caption to Figure 1 and Figures 2–4). 28

[1] Spurin, C., T. Bultreys, B. Bijeljic, M. J. Blunt, and S. Krevor, Intermittent fluid connectivity during two-phase flow in a heterogeneous carbonate rock, *Physical Review E*, 100(4), 043,103, 2019.

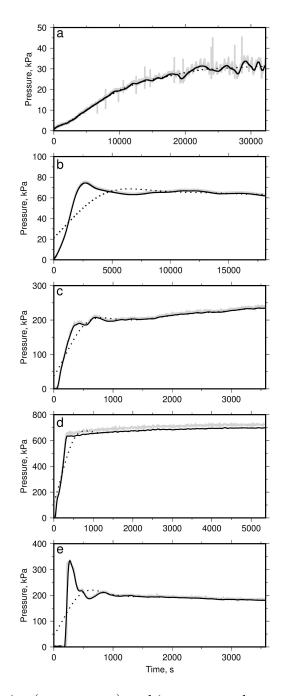


FIG. 1: Pressure time series (gray curves) and inverse wavelet transforms. Black solid and dotted curves = inverse wavelet transforms for periods > 10³ s and > 10⁴ s (panels a-b), respectively, or > 10² s and > 10³ s (panels c-e). (a) Pore-scale gas/water experiment (see Figure 2a-d). (b) Pore-scale oil/water experiment (Figure 2e-h). (c) Core-scale gas/water experiment (Figure 3a-d). (d) Core-scale oil/water experiment (Figure 3e-h). (e)

Core-scale gas/water experiment with the sample reversed (Figure 4).