

Characterization of turbulence in a Schwarz Diamond monolith using time-resolved MRI

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Periodic porous structures manufactured using additive manufacturing are candidates for intensified process devices. Detailed measurements of the velocity field are required to characterize the turbulent mixing generated by flow through the channel network. We used time-resolved single-shot spiral magnetic resonance velocity (MRV) imaging to noninvasively record snapshots of the velocity field for water flowing through a Schwarz Diamond monolith. The time-resolved velocity data lend themselves to more detailed analysis than is possible with conventional time-averaged magnetic resonance methods. The turbulent energy spectra measured with MRV confirmed that the pore network places an upper size limit on the eddies that could exist in the flow. The $-5/3$ power-law scaling from Kolmogorov theory was only attained at a Reynolds number around 2000, indicative of a higher transition point to a fully turbulent regime than is usually reported for random porous media. The local Reynolds stress tensor showed considerable anisotropy within the network, which affects mass transport to the channel surfaces. The time-resolved MRV method offers valuable insights into turbulence in porous media that can inform the design and operation of novel geometries.

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I. INTRODUCTION

Process intensification is the practice of designing unit operations to improve product quality while minimizing waste and financial costs. Some key principles of process intensification are (1) that each molecule should experience the same process conditions and (2) that driving forces should be applied to the maximum possible specific surface area [1]. Many processes that utilize pipes (shell and tube heat exchangers, steam methane reformers), plates (plate heat exchangers), and particles (chromatographic separations, packed bed reactors) do not satisfy these principles. Molecules in these units experience different process conditions where some travel rapidly through the widest channels, while others reside in dead ends within the void space for considerable amounts of time. Similarly, a shell and tube heat exchanger configuration does not represent a maximal specific surface area, thus heat transfer is not as effective as it theoretically could be.

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Nonetheless, these configurations are ubiquitous in the process industries because they are practical and inexpensive to manufacture.

The increased availability of additive manufacturing, also known as three-dimensional (3D) printing technology presents engineers with the opportunity to reimagine what unit operations could look like and to design novel process units that satisfy the principles of process intensification [2,3]. Of particular interest is the family of Triply Periodic Minimal Surfaces (TPMS) that exhibit good structural integrity and gradual variations in the channel morphology. Potential applications include processes such as chromatography [4], heat exchangers [5–7], gas scrubbers [8,9], and catalytic reactors [10–12].

A key advantage for 3D-printed reactors and heat exchangers is that the periodic nature of TPMS channels suppresses channeling such that flow is distributed evenly through the structure, which was confirmed by magnetic resonance imaging (MRI) experiments [13]. Furthermore, the tortuous channel geometry may promote the formation of vortex structures. However, different vortex structures can form depending on the shape of the channel. For example, particle image velocimetry (PIV) experiments of flow through zigzag microchannels [14] found that sharp, tight turns in the channel caused longitudinal vortices, i.e., recirculation zones to form, while smooth, gradual turns in the channel generated Dean vortices. This distinction is important because each vortex type affects the film heat transfer coefficient and pressure drop. An optimal channel morphology should achieve a balance of high heat transfer with low pressure drop. Channel geometry is also important for TPMS devices, where increasing the unit cell length along the flow direction enhances heat exchange performance [15].

Turbulent flow is especially relevant to process intensification because turbulence affects the extent of mixing, which in turn impacts heat transfer, reaction conversion, and selectivity [16]. Prior work for intensified heat exchangers includes the addition of porous metal foams that serve to induce turbulence at lower Reynolds numbers compared to straight pipes [17]. Metal foams help to improve the film heat transfer coefficient. However, the foam material significantly increases the pressure drop [18], rendering it unsuitable for pressure-sensitive applications such as exhaust gas recirculation. Regular pore networks may serve to reduce the pressure drop by eliminating recirculating flow and the smallest constrictions that exist for random networks. However, there is evidence to suggest that regular porous media exhibit a higher transition point to turbulence [19], where fully developed turbulence is only attained for pore Reynolds numbers above 1000, compared to a pore Reynolds number of approximately 750 for random porous media [20].

The clearest indicator of fully developed turbulence is the existence of the energy cascade, where kinetic energy is exchanged from large flow structures to progressively smaller eddies down to the Kolmogorov limit. The energy spectrum follows a $-5/3$ power law with increasing wave number. Mixing at the meso and molecular scales can be characterized in terms of the turbulent kinetic energy (TKE) dissipation rate [21]. Thus, there is considerable value in acquiring experimental measurements of the turbulent energy spectrum and the TKE dissipation rate in order to identify the extent of intensified process performance. Furthermore, such data are critical for validation of computational fluid dynamics (CFD) simulations.

The energy spectrum and dissipation rate can be measured with time-resolved techniques such as laser Doppler velocimetry and particle image velocimetry (PIV) [22], although PIV becomes challenging for porous structures due to the need to match the refractive index of the fluid with that of the structure [23]. PIV is also limited by the lack of sensitivity to flow perpendicular to the imaging plane, and the TKE dissipation rate is underestimated because the image resolution is usually too low.

MRI is a noninvasive, 3D, quantitative measurement technique that is sensitive to motion. These features make MRI unique in being able to obtain velocity fields in 3D for opaque systems without the measurement interfering with the flow. MRI is also useful for studying turbulent flows because information about the variance of the velocity distribution can be obtained using diffusion encoding [24]. MRI measurements are usually collected (and time-averaged) on the scale of minutes to hours [25] and thus cannot directly capture transient features of the system. The

total time required to perform an MRI experiment largely depends on how the spatial frequency domain, known as k -space, is sampled. In general, k -space data points are acquired at positions on a Cartesian grid. At one extreme, 3D single point imaging (SPI/SPRITE) acquires data one point at a time, potentially taking hours to acquire a complete image [26–28]. Conventional spin echo and gradient echo methods use frequency encoding to acquire k -space data line-by-line [29,30], greatly reducing the image acquisition time to the order of seconds or minutes, although for unsteady flows many repeats are required to suppress ghosting artifacts. Unsteady flow in the Schwarz Diamond TPMS geometry was studied using spin echo [31] and SPI [32] sequences. Neither of the aforementioned sampling schemes is able to resolve temporal fluctuations in unsteady flow because the experiment times are too long. Capturing transient flow features requires sampling the entirety of k -space in a single acquisition, such that the imaging time is reduced to several milliseconds.

Several single-shot rapid imaging methods exist that can be combined with velocity encoding, such as rapid acquisition with relaxation enhancement (RARE) [33,34], echo planar imaging (EPI) [35], and spiral imaging [36]. For RARE, image quality is high due to the constant refocusing of the magnetization. Velocity-encoded EPI methods [37–39] have the advantage of acquiring data on a Cartesian grid for easy image reconstruction via fast Fourier transform, but are hindered by artifacts caused by flow and off-resonance magnetization. With a single receive channel, typical RARE and EPI acquisition times are on the order of dozens of milliseconds. However, parallel imaging systems allow for notable reductions in acquisition time [40]. Additionally, multiband EPI enables systems to be imaged in 3D without significantly affecting the acquisition time compared to 2D EPI [41].

Spiral imaging is distinct from the aforementioned time-resolved imaging methods because data are not acquired on a Cartesian grid in k -space, which makes image reconstruction more challenging. However, the spiral gradient waveforms offer a degree of flow compensation, and the constant slew rate allows for acquisition times as short as 3 ms [42]. Spiral imaging has been applied to process systems such as bubbly flow [43] and flow through random packed beds [44].

In this article, we use rapid single-shot MRI to obtain time-resolved velocity images of unsteady and turbulent flow through the Schwarz Diamond TPMS structure. From these images, we determine the turbulent energy spectrum, Reynolds stress tensor, and an estimate of the TKE dissipation rate. Section II A describes the setup of the flow system, Sec. II B covers the theory behind MR velocity imaging, and Sec. II C describes the implementation of the spiral imaging protocol. Section II D describes how the velocity images were used to calculate turbulence properties. These results are presented and interpreted in Sec. III.

II. METHODS

A. Flow loop and column fabrication

The experiments were conducted using water pumped through a 3D-printed Schwarz Diamond column. The column was seated in the MRI scanner, with the column inlet connected with polyvinyl chloride (PVC) tubing to a pump outside the magnet. Depending on the required flow rate, a Xylem FloJet reciprocating diaphragm pump (Xylem Inc., Washington, D.C.) or a Stöckert S3 peristaltic pump was used (Stöckert Instrumente GmbH, Munich, Germany). Information about the experiment conditions are provided in Table I. A pulsation dampener was included downstream of the pump to smooth out pulsations in the flow. The outlet was connected to a reservoir with PVC tubing. The reservoir was used to supply the pump with water, thus closing the flow loop. A diagram of the flow loop incorporated into the MRI scanner is shown by Fig. 1(a).

The Reynolds number of the flow was set by adjusting the pump flow rate. The volumetric flow rate, $\dot{V}$, was measured in the flow loop using a bucket and stopwatch. The pore Reynolds number was determined according to

$$\text{Re} = \frac{\rho_f D_h \dot{V}}{\epsilon A \mu_f}, \quad (1)$$

TABLE I. Flow parameters for the velocity imaging experiments.

Re	$\dot{V}$ ($\times 10^{-5} \text{ m}^3 \text{ s}^{-1}$)	$\bar{u}_z$ (m s^{-1})	g_{vel} (T m^{-1})	Pump
253	0.633	0.0252	0.600	S3
367	0.917	0.0384	0.600	S3
442	1.10	0.0425	0.450	FloJet
730	1.82	0.0706	0.300	FloJet
1030	2.58	0.100	0.257	FloJet
1540	3.85	0.147	0.173	FloJet
2180	5.46	0.219	0.108	FloJet

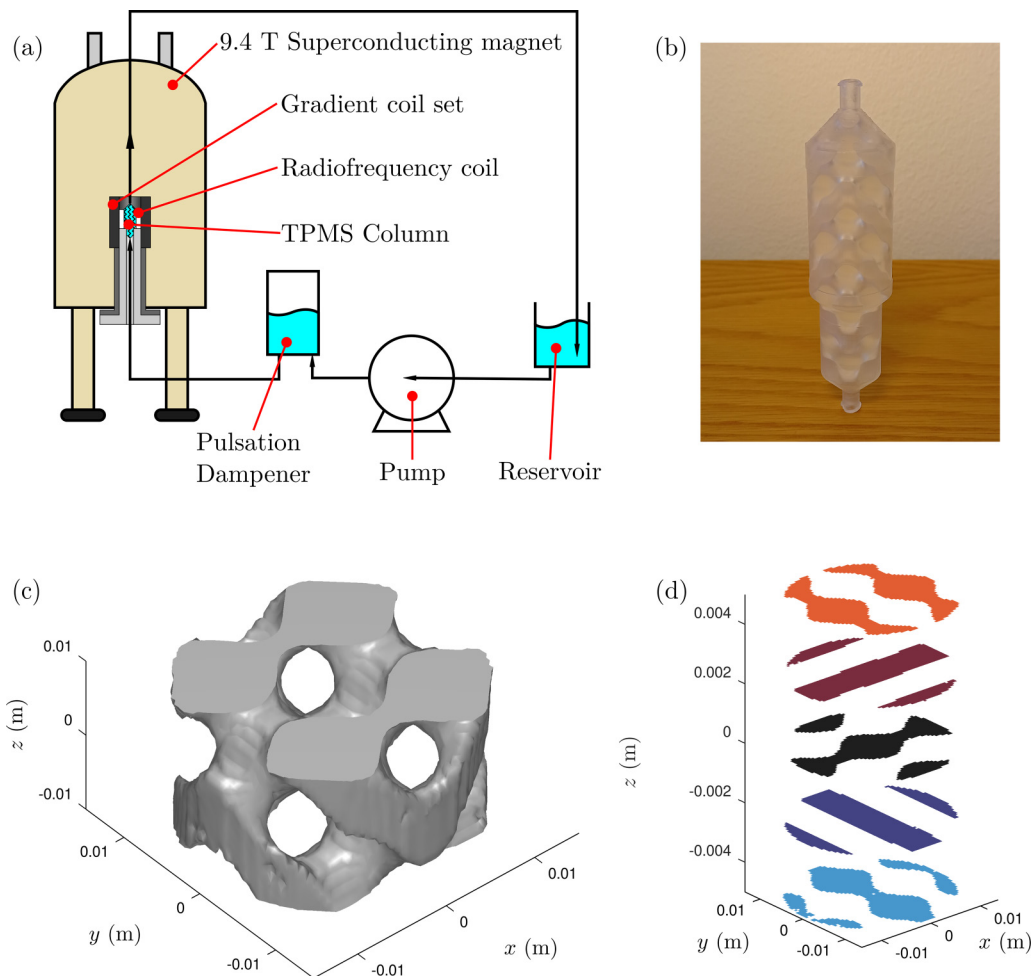


FIG. 1. Setup of MRI velocimetry experiments. (a) Diagram of flow loop. (b) Photograph of 3D-printed Schwarz Diamond column. (c) Render of 3D MR image of the central region of the column filled with water. (d) Maps of column cross-sections (slices) where time-resolved velocity images were acquired. The colors of these slices are arbitrary.

where ρ_f and μ_f are the mass density and the dynamic viscosity of the working fluid, respectively. The hydraulic diameter of the column, $D_h = 4\epsilon/a$, where $\epsilon = 0.5$ (in this case) is the voidage divided by the specific surface area a ; determined from a computer model of the Schwarz Diamond unit cell. The total cross-section area of the column is denoted by A .

The column geometry was designed with the objective of probing high Reynolds numbers without incurring a penalty on measurement accuracy caused by flow artifacts. To this end, the velocity should be kept low by increasing the hydraulic diameter. The RF coil can hold a sample 30 mm in diameter, allowing for wall thickness, the column diameter was 28 mm. The largest unit cell that can fit in the column has side lengths of 19.1 mm, corresponding to a hydraulic diameter of 9.95 mm. The Schwarz Diamond channel network was modeled using an in-house MATLAB program (Mathworks, Natick, MA). The column exterior, including Luer-type connectors, was designed using SolidWorks (Solidworks Corp., Waltham, MA). The meshes were used to create the column by taking the mesh Boolean difference between the column exterior and the channel network using Blender (Blender Foundation, Amsterdam, Netherlands).

The column was 3D-printed in clear resin using a Formlabs Form3 stereolithography 3D-printer (Formlabs Inc., Somerville, MA). The choice of material becomes important for spiral imaging due to field inhomogeneities caused by the magnetic susceptibility mismatch between the water and the resin. The Formlabs clear resin was found to have a volume magnetic susceptibility of -2.94×10^{-7} relative to water, i.e., slightly more diamagnetic [45]. The susceptibility mismatch corresponded to a field offset of ~ 120 Hz at 9.4 T, which would be prominent at the water-resin interface. The extent of field offsets that we measured were consistent with this measurement. Here, the mismatch was sufficiently low to attain images of acceptable quality, although we note that there is the opportunity to potentially improve the susceptibility match by the addition of a small amount of paramagnetic species to the resin before printing, similar to the experiments by Serial *et al.* [46].

A photograph of a printed column is shown in Fig. 1(b). When the column was filled with water, a 3D gradient echo image was acquired, from which the channel structure in the center of the column was rendered as shown in Fig. 1(c). Because the time-resolved imaging sequence is 2D, we collected images at different vertical offsets to capture various cross-sections of the column. The slices of interest are shown in Fig. 1(d). These were offset with gaps of 2.5 mm. These slices represent a subset of the pore characteristics of the Schwarz Diamond, where some pores are square-shaped in a checkerboard pattern, while others are rectangular spanning across the entire column.

B. Magnetic resonance velocity imaging: Theory

MRI operates on the principles of nuclear magnetic resonance (NMR), where the spin magnetization vector of the sample aligns along an applied magnetic field. The precession of the magnetization vector about the applied field can be detected via induced voltage in a receive coil when rotated away from the main magnetic field via a perpendicular radiofrequency pulse. Spatial localization is introduced by applying magnetic field gradients across the sample such that the precession frequency or phase of the local magnetization varies with position. The detected signal is a combination of these magnetization elements across the sample. Thus, the time-domain MRI signal data, $S(t)$ are in the spatial frequency domain (k -space), which is traversed via modulation of the magnetic field gradients. The image is reconstructed from these data using the discrete Fourier transform.

The MRI signal is given by

$$S(t) = \int \rho(\mathbf{x}) \exp[i\phi(t)] dV, \quad (2)$$

where $\rho(\mathbf{x})$ represents the spin density at position $\mathbf{x}$, and the phase angle accrued by the local magnetization, $\phi(t)$, is calculated according to

$$\phi(t) = \int_0^t \gamma \mathbf{g}(\tau) \cdot \mathbf{x}(\tau) d\tau, \quad (3)$$

where γ is the gyromagnetic ratio in units of $\text{rad s}^{-1} \text{T}^{-1}$ and $\mathbf{g}(t)$ is the magnetic field gradient. Assuming that the magnetization is advected by the flow at constant velocity, $\mathbf{u}$, such that $\mathbf{x}(\tau) = \mathbf{x}_0 + \mathbf{u}\tau$, the phase angle of the signal becomes

$$\phi(t) = \gamma \int_0^t \mathbf{g}(\tau) \cdot (\mathbf{x}_0 + \mathbf{u}\tau) d\tau. \quad (4)$$

The spatial frequency, k -space, is determined according to the zeroth moment of the gradient waveform:

$$\mathbf{k}(t) = \frac{\gamma}{2\pi} \int_0^t \mathbf{g}(\tau) d\tau. \quad (5)$$

Similarly, the first moment of a gradient waveform, representing reciprocal velocity space, $\mathbf{p}$ is defined by

$$\mathbf{p}(t) = \gamma \int_0^t \tau \mathbf{g}(\tau) d\tau. \quad (6)$$

By neglecting higher-order derivatives of the motion, we express ϕ as follows:

$$\phi(t) = 2\pi \mathbf{k}(t) \cdot \mathbf{x}_0 + \mathbf{p} \cdot \mathbf{u}(\mathbf{x}). \quad (7)$$

Equation (7) illustrates that the $\mathbf{g}(t)$ should be separated into two waveforms. First, we describe a velocity encoding waveform, $\mathbf{g}_{\text{vel}}$ comprised of a pair of opposite gradient pulses such that $\mathbf{p}$ is nonzero so that the phase angle may be only encoded for $\mathbf{u}$, but $\mathbf{k}$ is zero so that there is no contribution to image encoding. Because we encode for velocity along one coordinate axis at a time, we drop the vector notation from this point onwards. Defining the pulse amplitude as g_{vel} , the pulse time by δ , and the delay between the onset of the pulses by Δ ; it can be shown via evaluating Eq. (6) from $t = 0$ to $t = \Delta + \delta$ that the reciprocal velocity is given by

$$p = \gamma g_{\text{vel}} \delta \Delta. \quad (8)$$

The other waveform is the image encoding gradient that traverses a spiral pattern in k -space. Here, p is required to be zero to avoid extraneous velocity-encoding. The oscillatory character of spiral imaging waveforms means that they exhibit some degree of velocity and acceleration compensation, especially in the center of k -space [47,48]. Thus, we assume that the imaging gradient makes no contribution to p . We explore the robustness of this method to flow artifacts that may arise from not meeting this assumption in Sec. III A.

Substituting Eq. (7) into Eq. (2):

$$S(t, p) = \int \rho(\mathbf{x}) \exp[i(pu(\mathbf{x}) + 2\pi \mathbf{k}(t) \cdot \mathbf{x})] dV, \quad (9)$$

separating the exponentials gives

$$S(t, p) = \int \rho(\mathbf{x}) \exp[ipu(\mathbf{x})] \exp[2\pi i \mathbf{k}(t) \cdot \mathbf{x}] dV. \quad (10)$$

Instead of evaluating the velocity term as part of the volume integral, this term can be converted into a distribution in velocity space. Depending on the flow, different distributions may be used in the derivation [49]. In general, the phase shift is proportional to the mean velocity, $\bar{u}(\mathbf{x})$, and the distribution of velocity fluctuations, u' about the mean attenuates the signal magnitude. Assuming, for simplicity, that the velocity follows a Gaussian distribution with variance $\langle u'(\mathbf{x})^2 \rangle$, the velocity

space integral evaluates to

$$\begin{aligned}
 S(t, p) &= \int \rho(\mathbf{x}) \exp [2\pi i \mathbf{k}(t) \cdot \mathbf{x}] \exp [ip\bar{u}(\mathbf{x})] \int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi \langle u'(\mathbf{x})^2 \rangle}} \\
 &\quad \times \exp \left(\frac{-u'^2}{2 \langle u'(\mathbf{x})^2 \rangle} \right) \exp (ipu') du' dV \\
 &= \int \rho(\mathbf{x}) \exp [ip\bar{u}(\mathbf{x})] \exp \left(-\frac{p^2}{2} \langle u'(\mathbf{x})^2 \rangle \right) \exp [2\pi i \mathbf{k}(t) \cdot \mathbf{x}] dV.
 \end{aligned} \tag{11}$$

Fourier transform of the time-domain signal (encoded in spatial frequency domain) returns the spatial domain image $I(\mathbf{x})$, which is proportional to the spin density attenuated by the local velocity variance and with the phase angle proportional to the local mean velocity, $\bar{u}(\mathbf{x})$:

$$I(\mathbf{x}, p) = \rho(\mathbf{x}) \exp [ip\bar{u}(\mathbf{x})] \exp \left(-\frac{p^2}{2} \langle u'(\mathbf{x})^2 \rangle \right). \tag{12}$$

It is unclear if the intravoxel velocity distribution actually follows a Gaussian. For time-averaged MR velocity imaging, the velocity variance is the convolution of spatial and temporal variances, where temporal variance often tends to dominate [50]. Due to the short scan time employed here, the temporal velocity variance contribution over the course of the repetition time, T_R may be considered negligible if the velocity is approximately constant over this time scale. Hence we ignore the attenuation term, rendering the exact shape of the distribution as moot, provided that the skewness is zero. Using these assumptions, it then follows from Eq. (12) that the velocity information is encoded into the phase angle of the reconstructed image.

C. Magnetic resonance velocity imaging: Implementation

The experiments were conducted using an MRI scanner that comprised of a 9.4 T (400 MHz) superconducting magnet, a Bruker Avance I NMR spectrometer, and a Bruker Micro2.5 microimaging three-axis gradient coil (Bruker Biospin GmbH & Co KG, Ettlingen, Germany). The pulse sequences outlined here were implemented into TopSpin 2.1.

1. Spiral design

We first design the trajectories for the k -space vector. As a starting point, we implemented the Archimedian spiral formulation outlined by Glover [51]. In 2D, k -space can be considered as a vector, $\mathbf{k}' = k_x \hat{\mathbf{i}} + k_y \hat{\mathbf{j}}$, or similarly, as a complex number, $k' = k_x + ik_y$, where i is the imaginary unit,

$$k' = \lambda \theta \exp (i\theta), \tag{13}$$

where λ is the reciprocal of the FOV. Reference [51] derives how θ depends on time in order to address hardware limitations. For our purposes, the important detail is the value of θ at the end of the spiral, $\theta_{\text{End}} = \pi N$, where N is the number of turns in the spiral.

An Archimedian spiral can be said to fully sample k -space, where the number of turns in the spiral is half the image raster and the reciprocal of the k gap between successive turns gives the FOV. Speeding up the acquisition by reducing the number of turns subsequently reduces the image resolution. To counteract this loss of resolution, variable density spirals [52] were formulated where the spacing between the turns was narrower in the center to attain the desired FOV and further apart further away to attain the desired resolution. In this case, turn density scales via a power law, which, in our experience, tends to make the center excessively dense and the outer too sparse. Similar to the approach from Pipe and Zwart [53], we utilized a compound spiral with the standard density in the center and then decreasing linearly starting from halfway out such that a greater extent of k -space is reached with fewer turns. For example, for a 64×64 image raster, instead of the fully

sampled $N' = 32$ turns, we used $N = 16$ turns, calculated the Archimedian spiral trajectory, k' and then rescaled it by $k = k'f(\theta)$, where

$$f(\theta) = \begin{cases} 1 & \text{if } \theta \leq \theta_f \\ 1 + \left(\frac{N'}{N} - 1\right) \frac{(\theta - \theta_f)}{(\theta_{\text{End}} - \theta_f)} & \text{if } \theta > \theta_f \end{cases}, \quad (14)$$

where θ_f is the angular displacement at which the spiral starts to be rescaled, in our case it was set to $0.6\theta_{\text{End}}$. Equation (14) is smoothed using Savitzky-Golay filtering so that the numerical derivatives become continuous. We now obtain the gradient and time scale from the modified spiral by setting the slew rate, s . The fastest traversal of k -space is when $s = s_{\text{max}}$, where s_{max} represents the maximum slew rate. While the gradient rise time gives an indication of a theoretical maximum for s_{max} , in our experience, a much lower slew rate was required in order to avoid mechanical vibrations from Lorentz forces that caused unwanted signal spikes. For the Micro2.5 gradient coil set, we found that an s_{max} of $\sim 3500 \text{ T m}^{-1} \text{ s}^{-1}$ was suitable for this study, well short of the theoretical maximum of $\sim 1 \times 10^4 \text{ T m}^{-1} \text{ s}^{-1}$.

The slew rate is proportional to the second derivative of k with respect to time. Representing k as a series of discrete time points, the time interval Δt_i between successive points with indices $i - 1$ and i was calculated for a specified s_{max} according to the finite-difference approximation of the second derivative:

$$\Delta t_i = \frac{\gamma}{s_{\text{max}}} |\sqrt{k_{i+1} - 2k_i + k_{i-1}}|. \quad (15)$$

The finite differencing formula assumes that Δt is constant for all steps. However, Δt_i is not constant because the difference between k_i and its neighbors changes along the spiral trajectory. We keep the error small by using a high number of points ($> 10^4$) such that Δt_i changes gradually over the trajectory to be considered locally constant. The time coordinates were obtained by cumulative summation of Δt . The gradients for the compound spiral were found by the finite-difference approximation of the first derivative of k with respect to t . The waveform was then downsampled so that it was suitable to be read by the spectrometer.

There is a trade-off between the image frame rate and the image resolution. Increasing the image resolution requires more turns of the spiral, which results in a longer image encoding (acquisition) time, t_{Acq} thereby increasing the repetition time. Furthermore, a longer image encoding time increases the extent of flow artifacts in the image. For these reasons, we collect images at two different rasters, $32 \times 32 \text{ px}$, $t_{\text{Acq}} = 2.06 \text{ ms}$, and $64 \times 64 \text{ px}$, $t_{\text{Acq}} = 5.26 \text{ ms}$.

2. Spiral imaging

The velocity encoded spiral imaging sequence is shown in Fig. 2(a). The sample is excited using a soft RF pulse, with a velocity-compensated slice gradient. The RF pulse shape was a sinc convolved with a Gaussian with a pulse time of $256 \mu\text{s}$. RF spoiling was implemented by setting the phase of the RF pulse according to a quadratic sequence in order to spoil residual transverse magnetization from prior scans [54]. The slice thickness was 1.15 mm for the $32 \times 32 \text{ px}$ images and 0.75 mm for the $64 \times 64 \text{ px}$ images. The short repetition time of the low-resolution images resulted in an increase in the gradient duty cycle. In order to maintain a reasonable duty cycle on the gradient set and prevent excess heat generation, the slice gradient strength had to be reduced.

The velocity compensation takes the form of two pulses run after the slice selective portion of the waveform such that the zeroth and first moments of the waveform are zero. The velocity encoding pulses were run concurrently with these compensating pulses to keep the repetition time as short as possible. The effective length of these pulses was $285 \mu\text{s}$. This short pulse length and the minimal delay between pulses serves to keep unwanted acceleration encoding to a minimum. Depending on the volumetric flow rate, the velocity encoding gradient amplitude, g_{vel} was set to one of the values given in Table I. The same g_{vel} value was used to encode u_x , u_y , and u_z . The repetition time when using the $32 \times 32 \text{ px}$ spiral was 3.84 ms and 7.04 ms for the $64 \times 64 \text{ px}$ spiral. Only

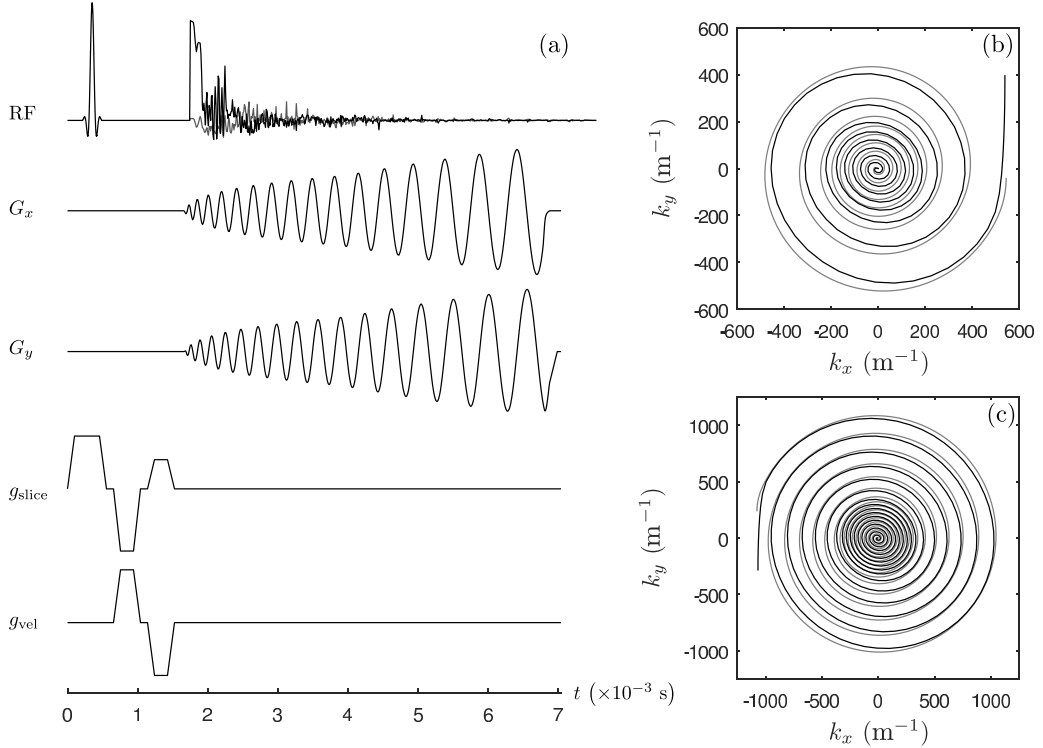


FIG. 2. (a) Pulse diagram for the 64×64 px velocity encoded spiral imaging sequence. Plots of the 2D k -space trajectory are shown in (b) for 32×32 px and (c) for 64×64 px image rasters. Designed trajectories are shown by the gray lines. Measured trajectories are shown by the black lines.

one velocity encoding step in reciprocal velocity space was performed in order to maximize the temporal resolution. Background phase was removed by running the experiment with the pump on (flow) and again with the pump off (static) and subtracting the static phase image from the phase image with flow.

We performed single velocity component measurements, which we abbreviate to 2D1C, short for two-dimensional imaging with one velocity component. In this case, the same velocity component was encoded in successive scans, which allowed us to capture fluctuations at the shortest time scale. We also collected 2D images with three velocity components (2D3C) to capture out-of-plane and in-plane flow by switching the velocity encodes in the looping sequence u_x , u_y , u_z , u_x , ... The frame rate for these experiments ranged from 47 Hz for the 2D3C 64×64 px raster to 260 Hz for the 2D1C 32×32 px raster. To allow for the shortest possible repetition times, scan data were stored in memory and written at the end of the experiment to avoid slow processes such as transferring to buffers or writing to disk. The number of frames, and hence the total duration of the experiment was determined by the available memory. For the 32×32 px raster, 512 images were collected for a total experiment time of 1.97 s. For the 64×64 px raster, 224 images were collected for a total experiment time of 1.58 s.

3. k -space trajectory mapping

The true k -space trajectory will not agree with the designed trajectory due to factors such as coil inductance and eddy currents. This mismatch in true versus designed k -space causes distortions in the reconstructed image. Therefore, the k -space trajectory needs to be measured directly in the MRI scanner.

Image-based methods can be used to measure the trajectory by encoding k in the signal. One of two main approaches is the offset slice method, where a slice offset from the gradient center is excited, followed by running the test waveform. The phase angle of the detected signal is proportional to the slice offset distance multiplied by k [55]. The efficacy of this method depends on being able to generate a narrow, rectangular slice profile, which runs into signal-to-noise limitations. The alternative method—which we use here—is the self-encoding method. This method performs 1D phase encoding, denoted by k_{self} , prior to the test waveform. Via the Fourier shift theorem, the phase encoding may be separated from the phase shift caused by the test waveform [56],

$$S(k_{\text{self}}, t) = \int \rho(x) \exp(2\pi i[k_{\text{self}} + k(t)]x) dV, \quad (16)$$

such that image reconstruction returns a 1D image, I , that evolves through time, where the spatial gradient of the phase of the image is equal to k :

$$I(x, t) = \rho(x) \exp[2\pi i k(t)x]. \quad (17)$$

The disadvantage of this approach is that the image resolution needs to be high enough to prevent the test waveform from phase wrapping between voxels [57], which can increase the total experiment run time. However, the experiment only needs to be run once for each waveform and exhibits far better signal-to-noise than the offset slice method. Here, we performed 32 repeat scans for each acquired image to ensure that the signal-to-noise ratio (SNR) would be adequate for all data points in the test waveform. This measurement was performed separately along the x axis using the x gradient coil running the $g_x(t)$ waveform and along the y axis using the y gradient coil with $g_y(t)$. We ran this experiment using a square-shaped hole phantom filled with water. The square shape ensured that the spin density was uniform along the imaging axis so that we could obtain an accurate linear fit to the phase and position data over the entire water-filled region where all voxels had high SNR. The designed and measured k -space trajectories for the spirals employed for the two image rasters explored in this study are shown in Figs. 2(b) and 2(c). Deviations from the designed trajectories developed in Sec. II C 1 are clearly apparent, illustrating the necessity of this procedure for accurate image reconstruction.

4. Static field homogeneity

A critical concern with spiral imaging is that off-resonance magnetization has a strong blurring effect due to considerable phase dispersion that can occur during the comparatively long image encoding time. Off-resonance artifacts are minimized by setting the image encoding time as short as possible and setting the static magnetic field as homogeneous as possible. The latter is done via manipulation of the currents in the shim coils to reach an optimal configuration. In the absence of automated shimming on the spectrometer, and considering the difficulty of finding the optimum by hand, we utilized the mathematical method developed by [58].

The frequency offset in an image voxel is given by the product of the current through the shim coil (in terms of the numerical settings on the shim unit), and the shim coil profile, which describes how much the local frequency changes with respect to the shim current for a given point in space. These were measured by collecting 3D gradient echo images of a tube of doped water with a series of echo times for several different shim settings for each shim coil.

To find the optimal shim currents for the sample, a 3D field map, Δf was collected using the same 3D gradient echo sequence as above for a baseline set of shim currents. The shim profiles were interpolated onto the field map raster. For a field map with M voxels and a scanner with N shim coils, the shim profiles were concatenated to give an $M \times N$ matrix, F_{shim} . We can express the problem as a system of linear equations, where we want to find the vector of shim currents i_{shim} (relative to the baseline shim settings used to acquire the field map) that minimizes Δf using least-squares:

$$\min_{i_{\text{shim}}} |F_{\text{shim}} i_{\text{shim}} - \Delta f|^2. \quad (18)$$

Having found the optimal set of shim currents for the sample, these were loaded onto the shim unit using python scripting in TopSpin. The root-mean-squared deviation in the frequency spectrum for the region of interest was around 30 Hz, which was sufficiently homogeneous to reconstruct images without applying additional off-resonance correction [59].

5. Image reconstruction

Conventional MR image reconstruction involves fast Fourier transform (FFT) of k -space data sampled on a Cartesian grid. Because the spiral trajectory is non-Cartesian, a nonuniform FFT algorithm must be used. Due to the variable density of the spiral and the comparatively low sampling frequency of 100 kHz, the k -space data were considered to be undersampled. Compressed sensing methods have been developed to reconstruct images from undersampled data acquisitions and are especially well-suited for non-Cartesian k -space trajectories such as spirals [60]. For this reason, we utilize the Parallel Imaging Compressed Sensing (PICS) program from the Berkeley Advanced Reconstruction Toolbox (BART) [61] to reconstruct the images. Because we only have a single receive channel, the coil sensitivity is set to unity. The PICS program computes the image I that satisfies the constrained optimization problem:

$$\min_x ||\mathcal{F}I - y||_2^2 + ||\alpha\mathcal{T}(I)||_1, \quad (19)$$

where $\mathcal{F}$ is the undersampled non-Cartesian Fourier transform operator, y is the measured data, and $\mathcal{T}$ is a sparse transform operator. The first term describes the congruence of the solution with the measured data (l_2). The second term describes the 1-norm of the transform sparsity (l_1). Here we use the total variation as the sparsity transform operator,

$$\mathcal{T}_{TV}(x) = \sqrt{\sum_{i=1}^D |\nabla_i x|^2}, \quad (20)$$

where ∇_i denotes the gradient along dimension i for an image with D dimensions.

Compressed sensing exploits the phenomenon where transformed images are sparse in the transform domain. This sparsity constraint serves to regularize the problem and obtain a stable solution to Eq. (19) that is not overly sensitive to noise patterns. The free parameter α controls the strength of the regularization term. As α is varied, the l_1 and l_2 terms can be plotted to construct an “L” curve, where the optimal α corresponds to the crook of the “L,” which represents minimal l_1 (transform sparsity) and minimal l_2 (high data fidelity). Figure 3 shows a plot of l_1 against l_2 of reconstructed images for varying α . There is an optimum where both l_1 and l_2 are near minimum, at $\alpha \approx 10$, thus we set α to 10 for the images reconstructed in this study.

D. Estimation of turbulence statistics

Direct simulation of turbulence using the Navier-Stokes equations is infeasible for many systems due to the smallest eddies, and thus the grid resolution, being many orders of magnitude smaller than the overall length of the system. The required level of grid resolution leads to prohibitively expensive computational overheads. The Reynolds-averaged Navier-Stokes approach (RANS) decomposes the velocity into mean and fluctuating components $u_i = \bar{u} + u'_i$. The products of the fluctuating component u'_i are time-averaged, leading to the appearance of Reynolds stresses $\overline{u'_i u'_j}$. To close the system of equations, the Reynolds stresses need to be defined in terms of the mean velocity. This can be achieved by describing the Reynolds stresses as an additional effective viscous stress term characterized by its own eddy viscosity, distinct from the molecular viscosity. The eddy viscosity is defined in terms of turbulence properties that themselves require additional transport equations. These properties describe the strength of turbulent fluctuations via the TKE:

$$\kappa = \frac{1}{2} \overline{u'_i u'_i}, \quad (21)$$

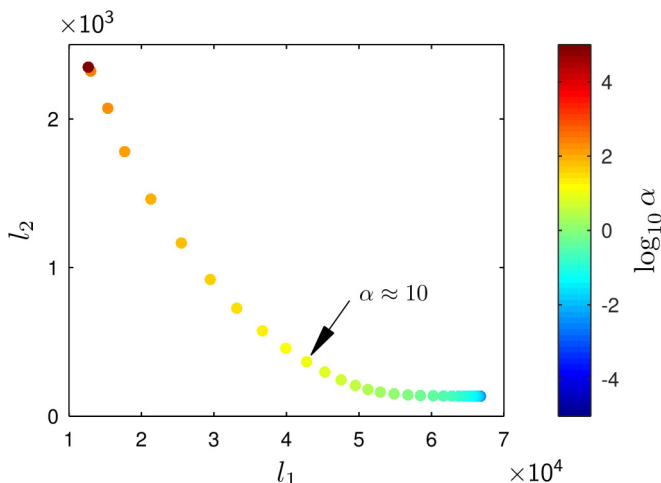


FIG. 3. Plot of the transform sparsity, l_1 , against the solution error, l_2 , for various regularization strengths. For this system, the optimal α is shown by the arrow.

and the TKE dissipation rate [62]:

$$\varepsilon = \nu \overline{\left(\frac{\partial u'_i}{\partial x_j} + \frac{\partial u'_j}{\partial x_i} \right)^2}. \quad (22)$$

The TKE and its dissipation rate can be related to the microscale behavior of the flow via the turbulent energy spectrum, $E(k_E)$, which represents the energy contributed by fluctuations in terms of their wavenumber, k_E . As such, the TKE is given by the integral of $E(k_E)$ over wave-number space, while the dissipation rate is given by the second moment of the energy spectrum. Three distinct regions exist in the energy spectrum; (1) at low wavenumbers, coherent flow features at the system (or pore) length scale have most of the fluctuating kinetic energy in the flow. (2) TKE dissipation only occurs at a sufficiently high wave number (small-scale eddies) where velocity fluctuations are damped by the kinematic viscosity ν of the fluid. This limit is associated with the Kolmogorov microscales, which are relevant to simulation and experimentation because they specify the requisite space and time resolution as well as the sensitivity to velocity in order to measure the entire energy spectrum. (3) For intermediate wavenumbers between these extremes, fluctuating kinetic energy is transported from the large scale flow structures to the dissipation scale via a cascade of progressively smaller eddies. Assuming that the distribution of velocity fluctuations depends only on the TKE dissipation rate, Kolmogorov's $-5/3$ scaling law for this inertial subrange can be derived by dimensional analysis:

$$E(k_E) = \text{Const.} \, \varepsilon^{\frac{2}{3}} k_E^{-\frac{5}{3}}. \quad (23)$$

Energy spectra are typically measured by experiments operating in an Eulerian reference frame. Although MRI experiments are, strictly speaking, in a Lagrangian frame, we can approximate the velocity images as being in an Eulerian frame owing to the short timescales over which each image is acquired. Therefore, we apply the analysis procedures employed previously in optical imaging experiments. The energy spectrum can be computed from the Fourier transform of the two-point correlation of the velocity. According to the Wiener-Khinchin theorem, the power spectral density of the velocity in the spatial frequency, i.e., wave-number domain is equivalent. Because the latter is easier to compute, we use the approach from [63], which is explained well by [64]. First, the time domain fluctuation velocity maps, $u'_i(\mathbf{x}, t)$ were converted into the spatial frequency domain,

$\hat{u}'_i(\mathbf{k}_E, t)$ via 2D-FFT, where $\mathbf{k}_E$ is the spatial wave vector, not to be confused with k -space used in image reconstruction. The power spectral density (PSD) is:

$$E(\mathbf{k}_E, t) = \hat{u}'_i(\mathbf{k}_E, t) \hat{u}'_i{}^*(\mathbf{k}_E, t), \quad (24)$$

where the asterisk denotes the complex conjugate. Next, the PSDs were time-averaged. This 2D energy spectrum was converted into a 1D energy spectrum by binning the PSD values according to the magnitude of the 2D wave vector, $||\mathbf{k}_E|| = k_E$. To account for the increased degeneracy with respect to higher wave number, the radially binned PSD was multiplied by the circumference of the annulus with radius k_E .

The integral of $E(k_E)$ with respect to k_E yields the TKE, which should be consistent with the TKE computed from the variance of velocity over the entire image set as defined by Eq. (21). In this case, it is important to consider the contribution of voxels that do not contain fluid. From the perspective of spectral analysis of the images, these are treated as zero flow, which decreases the mean and variance of the velocity. Meanwhile, statistical analysis ignores voxels that do not contain fluid. Thus, it was necessary to rescale the velocity spectra by dividing by the occupancy fraction, i.e., the fraction of voxels in the image which contained fluid. With this correction, we found that the TKE estimates from the spectrum were within about 1% of the statistical TKE calculated from the variance of velocity for the 64×64 images, indicating that the fluctuating energy was well-preserved by the processing routine.

Determining the TKE dissipation rate requires computation of the velocity gradient terms. Expressing Eq. (22) in full as the sum of individual terms gives

$$\begin{aligned} \varepsilon = \nu & \left[2 \overline{\left(\frac{\partial u'_x}{\partial x} \right)^2} + 2 \overline{\left(\frac{\partial u'_y}{\partial y} \right)^2} + 2 \overline{\left(\frac{\partial u'_z}{\partial z} \right)^2} \right. \\ & + \overline{\left(\frac{\partial u'_x}{\partial y} \right)^2} + \overline{\left(\frac{\partial u'_x}{\partial z} \right)^2} + \overline{\left(\frac{\partial u'_y}{\partial x} \right)^2} + \overline{\left(\frac{\partial u'_y}{\partial z} \right)^2} + \overline{\left(\frac{\partial u'_z}{\partial x} \right)^2} + \overline{\left(\frac{\partial u'_z}{\partial y} \right)^2} \\ & \left. + 2 \overline{\frac{\partial u'_x}{\partial y} \frac{\partial u'_y}{\partial x}} + 2 \overline{\frac{\partial u'_x}{\partial z} \frac{\partial u'_z}{\partial x}} + 2 \overline{\frac{\partial u'_y}{\partial z} \frac{\partial u'_z}{\partial y}} \right]. \quad (25) \end{aligned}$$

MRI methods have not been used to measure ε owing to the limited temporal resolution which makes it impossible to calculate the instantaneous velocity fluctuation gradients required for mean-squared values. 2D optical methods such as PIV estimate ε by approximating seven unknown gradient terms as an average of five known terms $\frac{\partial u'_x}{\partial x}$, $\frac{\partial u'_x}{\partial y}$, $\frac{\partial u'_y}{\partial x}$, $\frac{\partial u'_y}{\partial y}$, and $\frac{\partial u'_x}{\partial y} \frac{\partial u'_y}{\partial x}$ via the assumption of statistically isotropic turbulence [65]. Because we measure out of plane motion with MRI, thereby attaining $\frac{\partial u'_x}{\partial x}$, $\frac{\partial u'_z}{\partial y}$, and because our experiments use liquid water, we may determine $\frac{\partial u'_z}{\partial z}$ from the other diagonal velocity gradient terms according to the incompressible continuity equation [66]. Therefore the number of known terms increases to eight. The unknown square terms were set to the mean of the known square terms, and the unknown cross product terms were assumed to be equal to the known cross term [67]. These approximations may cause the TKE dissipation rate to be underestimated.

Energy dissipation caused by eddies smaller than the image voxels does not contribute to the measurement. Although MRI is sensitive to intravoxel velocity distributions, no clear relation to the mean squared velocity gradients is apparent. Thus, similar to optical methods, these MRI measurements will underestimate ε due to the image resolution being above the Kolmogorov microscale. Furthermore, the SNR of these images was rather low due to being single scans with a low RF tip angle. For these reasons, we do not attempt to approximate subvoxel dissipation in this study.

The eddy viscosity also emerges from the large eddy simulation (LES) framework, where the Navier-Stokes Equations are filtered to remove small-scale flow information from direct simulation.

The eddy viscosity term is used to close the equations such that the energy dissipated by sub-grid fluctuations is properly accounted for. In principle, the eddy viscosity could be estimated directly using the MRI velocity images via computation of the viscous stress tensor and the strain rate tensor. However, this is an emerging area of research that has so far proved challenging using optical velocimetry techniques [68] due to artifacts that emerge from computation of the velocity gradients. For this reason, we consider direct estimates of eddy viscosity beyond the scope of the present study.

III. RESULTS AND DISCUSSION

All experiment data and postprocessing scripts are freely available in the Zenodo repository [69]. This dataset also includes animations of the time-varying velocity fields for the 2D3C experiments at different slice positions and Reynolds numbers.

To briefly illustrate the time variation of the flow, a series of 2D3C images for each slice are shown in Fig. 4. To accentuate the changes in the velocity field over time, we have shown images five frames (105 ms) apart. The color map represents the axial velocity along the z direction, which uses the ColorBrewer 2.0 *Divergent Spectral* color scale [70]. The 3D quivers, generated using the MATLAB functions by [71], represent the velocity vectors. The maps and quiver plots are superimposed over sections of the 3D rendering from Fig. 1(c) to indicate the slice location in the column and to provide an illustration of the pore connectivity between the slices.

From Fig. 4, the velocity field can be seen to change from frame to frame with respect to time. To draw the reader's eye to some of these intermittent features, we have added outlines around some regions of high, low, and swirling flow. The maps indicate that the streams were diverted into different downstream pores during the course of the experiment. This observation runs contrary to the idea of spatially uniform flow distribution observed under steady flow. Under unsteady conditions, the flow distribution is only uniform once time-averaged. The flow pattern is more stable in the outer region of the column, where the pores are truncated by the column wall. The insights that can be drawn from visual analysis are qualitative and imprecise, hence we conduct quantitative analysis of the images in the following sections.

A. Method verification

Sources of quantitative error in the velocity fields include additional velocity encoding caused by the spiral gradients, and image distortion caused by non-negligible motion of fluid elements during a scan. We check for velocity encoding error by estimating the overall mean velocity, $\bar{u}_z$, from the axial velocity images and compare it to an independent measurement. Here, we used a bucket and stopwatch to measure the volumetric flow rate. Knowing the cross-sectional area and voidage of the column, we can convert the volumetric flow rate into a mean interstitial velocity. The mean velocity from the MRI experiments is plotted against the mean velocity determined from bucket and stopwatch measurements in Fig. 5(a). Horizontal error bars represent the uncertainty in the bucket and stopwatch experiments. These uncertainties were negligible except for the fastest flow rate due to short measurement times to avoid emptying the reservoir. We estimated the uncertainty in the overall mean velocity from the MRI experiments using the temporal standard deviation of the spatially averaged velocity in each image. This uncertainty was sufficiently small ($<1.5\%$) such that the vertical error bars were too small to be distinguishable in Fig. 5(a). The uncertainty estimates only apply to random measurement errors and not systematic errors that may arise from features of the experimental setup.

The MRI experiments tend to overestimate the mean velocity compared to the bucket and stopwatch, although parity is generally within experimental uncertainty. The 32×32 raster follows parity more closely than the 64×64 raster due to reduced velocity encoding from the shorter and weaker imaging gradients. For the fastest flow case, MRI overestimated the mean velocity by about 12% in the 64×64 raster, which may be tolerable for semiquantitative analysis. However, it likely represents an upper limit for what can be accurately imaged with the experimental setup used here.

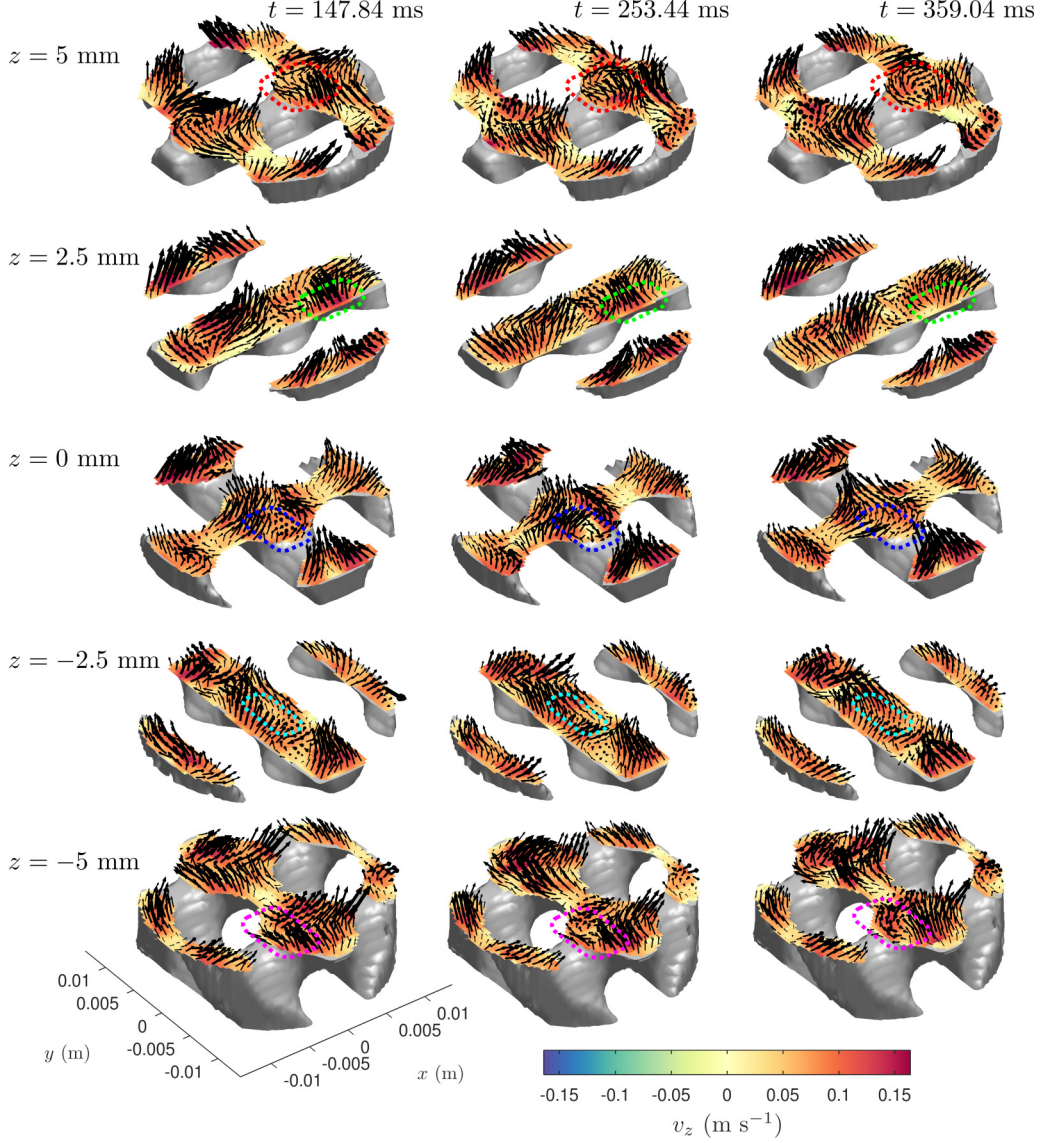


FIG. 4. Snapshots of velocity field in the Schwarz Diamond column obtained using spiral MRI velocity imaging using the 2D3C encoding scheme and a 64×64 px image raster for $Re = 730$. Color map represents axial velocity, quivers represent velocity vectors. Each row represents a different slice position, and each column represents a different snapshot in time. Note that the different slice positions were acquired in sequential experiments and there is no correlation between the velocity fields at different slices for the same given time. Intermittent flow features of interest are highlighted by the dashed outlines.

To quantify image distortion, we calculated the relative mean-squared deviation (relative MSD) in the image intensity between a time-averaged image acquired under flow, I_{Flow} , compared to a time-averaged image acquired with the flow off, I_{Static} , according to

$$I_{\text{MSD}} = \frac{\sum^{N_v} (I_{\text{Flow}} - I_{\text{Static}})^2}{\sum^{N_v} I_{\text{Static}}^2}, \quad (26)$$

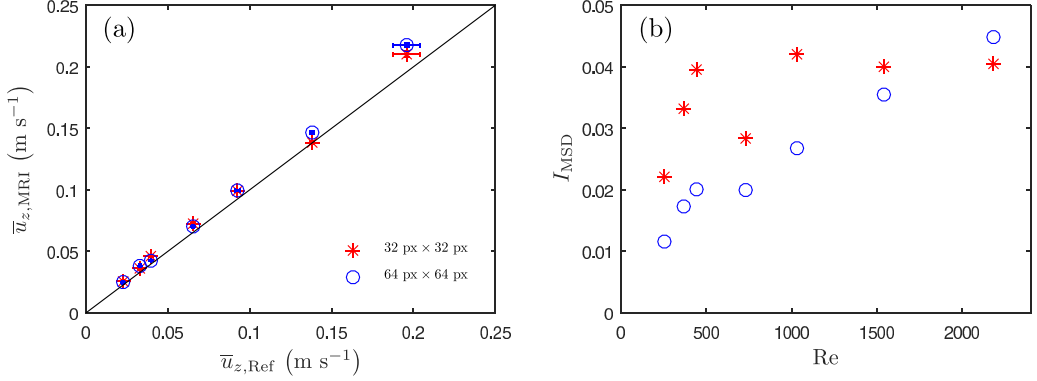


FIG. 5. (a) Mean velocity measured using MRI vs the mean velocity estimated from bucket and stopwatch measurements of the volumetric flow rate. The solid line represents parity. (b) Relative mean-square deviation between intensity of an image acquired under flow and an image with the flow off.

where the summation is done over the number of voxels, N_v . The plot of the relative MSD intensity against Reynolds numbers for both image rasters is shown in Fig. 5(b). In all the cases investigated here, the relative MSD was less than 5%, which indicated that the image distortion due to uncompensated flow artifacts and misregistration was sufficiently low such that the images could be interpreted with an adequate degree of quantitative accuracy. For the 64×64 image raster, the relative MSD increases with increasing Reynolds number due to increased fluid displacement during the scan. Contrary to expectation, the relative MSD for the 32×32 image was less dependent on the Reynolds number. Despite having a shorter scan time to avoid fluid displacement artifacts, the relative MSD was higher than the higher resolution images. It is possible that the aggressive undersampling scheme utilized to attain a short T_R exacerbated perturbations to the signal caused by flow. This level of accuracy with the spiral imaging method was only possible due to the high homogeneity of the B_0 field and precise knowledge of the trajectory in k -space.

B. Turbulence statistics

The TKE was calculated using the 2D1C experiments and is plotted for both image rasters against the Reynolds number in Fig. 6(a). In some instances, the 32×32 px case predicted lower velocity mean and variance due to an increased partial volume effect due to the increased voxel

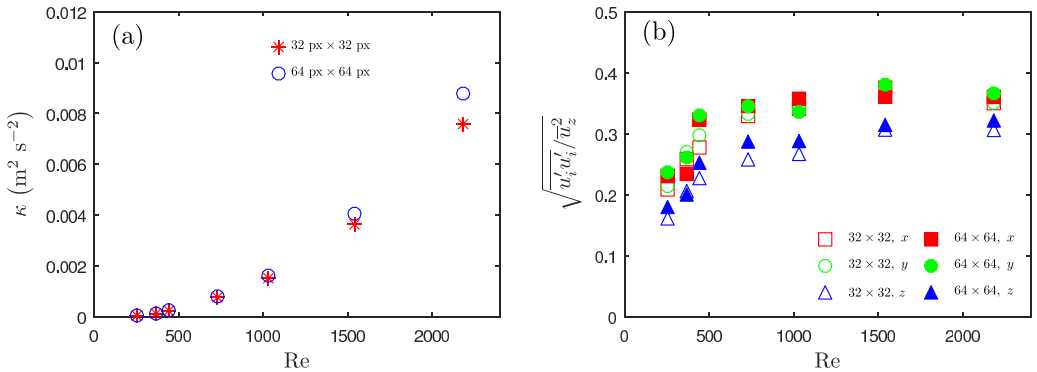


FIG. 6. (a) Plot of turbulence kinetic energy with respect to pore Reynolds number. (b) Turbulence intensity components vs pore Reynolds number.

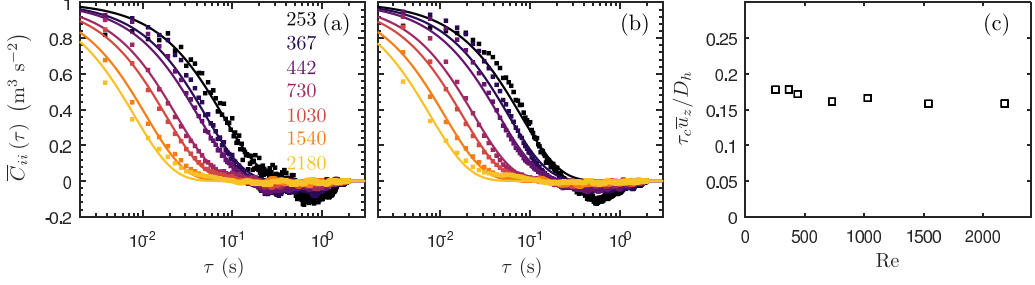


FIG. 7. (a) Temporal autocorrelation of x velocity fluctuations. (b) Temporal autocorrelation of z velocity fluctuations. Markers denote experimental data. Solid lines represent monoexponential decay fits to the data. The Reynolds number for each plot is indicated by the annotations in (a). (c) Normalized mean correlation time vs Reynolds number. All data shown are from the 32×32 image rasters.

size. To more clearly illustrate the scaling in TKE versus the Reynolds number, the turbulence intensity components are plotted against the Reynolds number in Fig. 6(b). The turbulence intensity components were determined by dividing the square root of the velocity variance by the mean axial velocity.

The turbulence intensity became constant with respect to Reynolds number for $\text{Re} > 1000$. Similar scaling behavior was observed for pulsed-field gradient (PFG) experiments [32], although there was a systematic disparity between the plateau values obtained from PFG and time-resolved MRI. The reason for this disparity likely stems from the joint spatiotemporal character of the velocity variance measured by PFG that we briefly described in Sec. II B. In contrast, the velocity variance computed from time-resolved MRI only includes the temporal variance of the velocity.

To ascertain the characteristic timescales for the velocity fluctuations, we calculated the normalized autocorrelation coefficients for the fluctuating velocity in each voxel:

$$C_{ii}(\tau) = \frac{\overline{u'_i(t)u'_i(t+\tau)}}{\overline{u'_i u'_i}}. \quad (27)$$

We obtained the mean autocorrelation by then averaging over all fluid-containing voxels. Plots of $\overline{C}_{xx}$ and $\overline{C}_{zz}$ for the 32×32 px images are shown in Figs. 7(a) and 7(b). The autocorrelation function is modeled by an exponential decay, $C(\tau) = \exp(-\tau/\tau_c)$, where the decay constant is the mean correlation time. Although the monoexponential decay model is generally not suitable for describing Eulerian turbulence [72], we use it here because the model follows our data well, except for the negative correlation at long lag times. The plots for $\overline{C}_{yy}$ are similar to the plots shown in Fig. 7, so are not shown here for brevity. The correlation time varies from ~ 80 ms ($20T_R$) for $\text{Re} = 253$ to ~ 8 ms ($2T_R$) for $\text{Re} = 2180$. We plot the mean correlation time normalized by the mean velocity and the hydraulic diameter against the Reynolds number in Fig. 7(c). The normalized correlation time is insensitive to the Reynolds number, with an average of 0.183. This trend agrees with the findings from the PIV experiments in random packings by Patil and Liburdy [73], and shows that there is an inverse relationship between the mean correlation time and the mean velocity.

Using the 2D3C images to calculate the TKE dissipation rate via Eq. (25) makes the assumption that the velocity remains constant over the duration of a few frames such that the velocity gradients are simultaneous. In all cases, the repetition time was shorter than τ_c . However, for the 64×64 px raster for $\text{Re} = 2180$, $T_R \approx \tau_c$. Because three frames are required to construct a velocity vector map, the effective time scale of image acquisition can exceed the mean correlation time of the flow. Therefore, the assumption of constant velocity during the image acquisition may not hold for the highest Reynolds numbers.

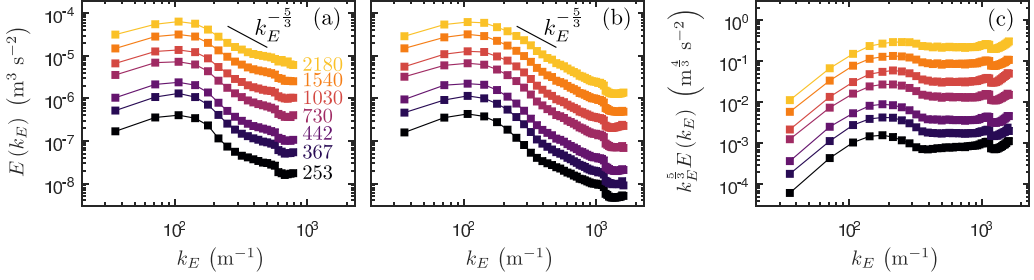


FIG. 8. Energy spectra of velocity fluctuations for several Reynolds numbers and image raster of (a) 32×32 px, (b) 64×64 px. (c) The spectra in (b) rescaled by $k_E^{-5/3}$. The inertial subrange region is indicated with the solid line following $-5/3$ scaling. The Reynolds number for each spectrum is indicated by the annotations in (a)

C. Energy cascade

The energy spectrum calculation described in Sec. II D was applied to the x , y , and z velocity 2D1C datasets. Plots of the energy spectra as a function of wave number for the Reynolds numbers studied are shown in Fig. 8. A line illustrating $-5/3$ scaling associated with the energy cascade in the inertial subrange is plotted for reference. All spectra exhibit a peak at $k_E \sim 100 \text{ m}^{-1}$, which corresponds to an eddy wavelength of 10 mm, which is equal to the hydraulic diameter of the Schwarz Diamond unit cell. This feature was expected because the size of the pores imposes an upper limit on the maximum attainable eddy length scale [74] which has been observed in other porous media experiments [73] and simulation [75,76]. However, the energy spectrum indicates that eddies with lower wave numbers still exist within the system, albeit with reduced power relative to the peak wave number. Due to the comparatively high voidage, the interconnectivity of the unit cells, and the lack of constrictions, the channel network could potentially support flow structures larger than the size of a single pore, such as across the spanwise cross-sections shown in Fig. 4. Such eddies would play an important role in transferring energy between unit cells, thereby enhancing the overall mixing performance.

Following the peak power, the energy spectra exhibited an approximately power-law decay up to a wave number of $\sim 250 \text{ m}^{-1}$ for the 32×32 raster, or $\sim 400 \text{ m}^{-1}$ for the 64×64 raster. Beyond this point, there was an inflection towards a weaker decay rate, which we associate with the noise limit of the spectra. To illustrate the power-law scaling more clearly, Fig. 8(c) plots the energy spectra multiplied by $k_E^{5/3}$, where Kolmogorov scaling follows a horizontal line. At the lowest Reynolds numbers studied here, the energy spectra do not follow Kolmogorov scaling, and the stronger decay rate indicates that high wave numbers have low power due to the relative absence of small eddies. As the Reynolds number increases, high wave number modes gain more power, indicating that smaller scale eddies are forming as the flow takes on a more turbulent character. At the highest Reynolds number studied here ($\text{Re} = 2180$), the data approach a horizontal line between wave numbers of $150\text{--}400 \text{ m}^{-1}$, which demonstrates that the decay in the energy spectrum scales in accordance with the Kolmogorov theory. This result provides evidence in support of the flow regime classification outlined by [19], where the unsteady behavior widely observed at $\text{Re} \sim 10^2$ represents a transitional regime rather than a strictly turbulent regime.

We could not capture the dissipative regime, where the energy spectrum decays faster than $-5/3$, because the image resolution and signal-to-noise ratio were too low to provide access to sufficiently high wave numbers. A potential alternative is to study the energy spectrum in a Lagrangian reference frame via modulated-gradient diffusion MRI sequences [77].

The estimated TKE dissipation rate from the 2D3C datasets for both image rasters is plotted against the Reynolds number in Fig. 9(a). The 64×64 image raster gave a much higher dissipation rate due to better resolution of small scale features contributing to the mean-squared velocity

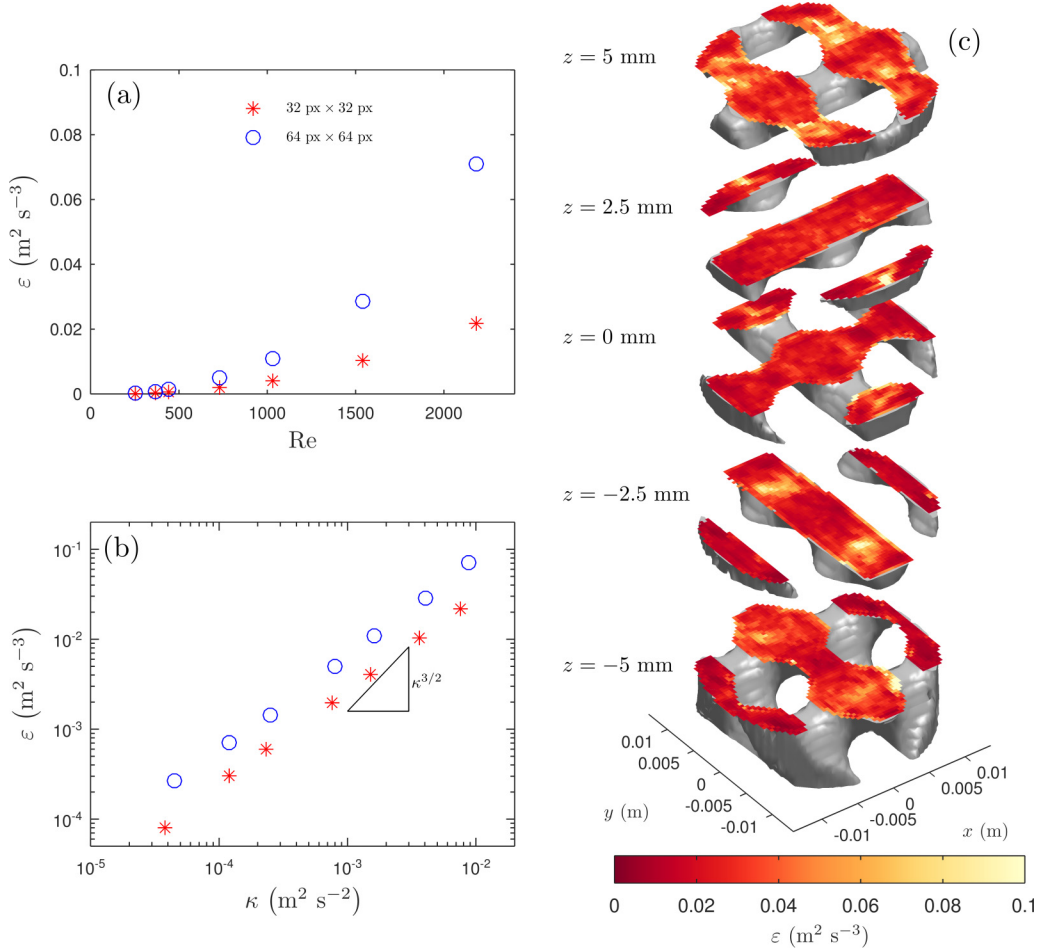


FIG. 9. (a) Turbulent kinetic energy dissipation rate vs Reynolds number. Markers represent MRV experiments. (b) TKE dissipation rate vs TKE. The triangle indicates $3/2$ exponent power-law scaling according to dimensional analysis. (c) Images of the local TKE dissipation rate for Reynolds number 1540.

gradients. The dissipation rate was plotted against the TKE in Fig. 9(b). Dimensional analysis predicts that $\varepsilon \propto \kappa^{3/2}/L$, where L is the integral length scale, which was approximately 7 mm in this case. The data follow a power law where the scaling exponent is reasonably close to the dimensional analysis prediction.

Using the dimensional analysis estimate of the dissipation rate, the smallest possible eddy size supported by the flow, the Kolmogorov length scale η , was between $30 \mu\text{m}$ at $Re = 2180$ to $170 \mu\text{m}$ at $Re = 253$. Because η was below the voxel widths in all cases studied here, the contribution from the smallest eddies was not included, leading to underestimation of the dissipation rate. This effect was most pronounced at the highest Reynolds numbers, where η was smallest. Mean-squared fluctuation gradient estimates of the dissipation rate are extremely sensitive to voxel size, and there is a trade-off between resolution and measurement error [66] (or signal to noise). Furthermore, our estimate of the dissipation rate does not include most gradient terms along the z axis. Although these measurements could not access the entire turbulence spectrum, they may have utility as a comparative tool to evaluate the mixing performance of various porous media geometries.

The local TKE dissipation rate is shown in the images for $Re = 1540$ in Fig. 9(c). These images show that the dissipation rate is highest in regions upstream of where pores split apart. For example, velocity gradients are prominent along the central channel of the $z = -2.5$ mm slices shown in Fig. 4, where fast-moving flow is in close proximity to swirling regions with low axial velocity. The exact shape of these features changes due to the unsteady nature of the flow field, which leads to fluctuating velocity gradients associated with TKE dissipation. Indeed, these areas correspond to local maxima in the dissipation rate in Fig. 9(c). The occurrence of these features is connected to the morphology of the porous media channel network, illustrating the role that channel geometry plays in generating flow patterns that dissipate TKE, which in turn influences mixing performance.

D. Turbulence anisotropy

Images of the full Reynolds stress tensor (RST) were determined from the covariance matrix of the 2D3C velocity vectors:

$$\mathbf{R}(\mathbf{x}) = \overline{\mathbf{u}'(\mathbf{x}, t) \mathbf{u}'^T(\mathbf{x}, t)}, \quad (28)$$

where $\mathbf{u}'^T$ denotes the transpose of the fluctuating velocity vector. An eigendecomposition of the RST returns the principal components $\lambda_1, \lambda_2, \lambda_3$ which describe the strength of turbulence along the principal axes. For isotropic turbulence, the principal components are equal. Anisotropic turbulence can be 1D where one principal component is larger than the others, or 2D where two principal components are similar in value. The eigendecomposition also returns a rotation matrix that describes the orientation of the aforementioned principal axes. This information contained in the RST can be represented visually via superquadric glyphs [78]. Isotropic (3D) turbulence is represented by a spherical glyph, 2D turbulence is represented by a disk-shaped glyph, and 1D turbulence is represented by a rod-shaped glyph. RST glyph maps are shown in Fig. 10 for three different slices and three Reynolds numbers. For additional clarity, the glyphs are coloured according to their dimensionality [76,79]. At the lowest Re , the glyphs near the wall are 2D in character, while the glyphs in the channel centres are 1D with the major axis perpendicular to the pore walls. For $Re = 2180$, the near-wall glyphs are more isotropic in character and the center-channel glyphs are more cigarlike in shape.

The fractional anisotropy (FA) for each voxel was determined according to $FA = \sqrt{1 - I_2/I_4}$, where I_2 and I_4 are invariants of the RST [80], given by

$$\begin{aligned} I_2 &= \lambda_1\lambda_2 + \lambda_2\lambda_3 + \lambda_3\lambda_1, \\ I_4 &= \lambda_1^2 + \lambda_2^2 + \lambda_3^2. \end{aligned} \quad (29)$$

The mean FA in each stack of images was plotted against the Reynolds number in Fig. 11. For both image rasters, the mean FA decreases with increasing Reynolds number, indicating that the RST becomes closer to isotropy as the flow becomes more turbulent. The 32×32 images exhibited higher mean FA than the 64×64 images. This disparity was likely caused by increased dispersion of velocity vectors within the larger voxels.

Because the RST is calculated from the velocity images, where each velocity vector is the mean value of the flow within a voxel, these estimates are representative of anisotropy at the voxel length scale. These FA measurements suggest that turbulence is anisotropic not just for the largest flow features at the integral length scales, but at smaller length scales as well. Therefore, turbulent eddies within the energy cascade regime are likely anisotropic, contrary to the assumption of isotropic, homogeneous turbulence made by the Kolmogorov scaling model. These data, in conjunction with Sec. III C, suggest that the flow converges to isotropic turbulence with increasing Reynolds number, which is associated with convergence to Kolmogorov scaling. However, testing this hypothesis requires experiments at higher Reynolds numbers than are possible with the experimental setup used in this work.

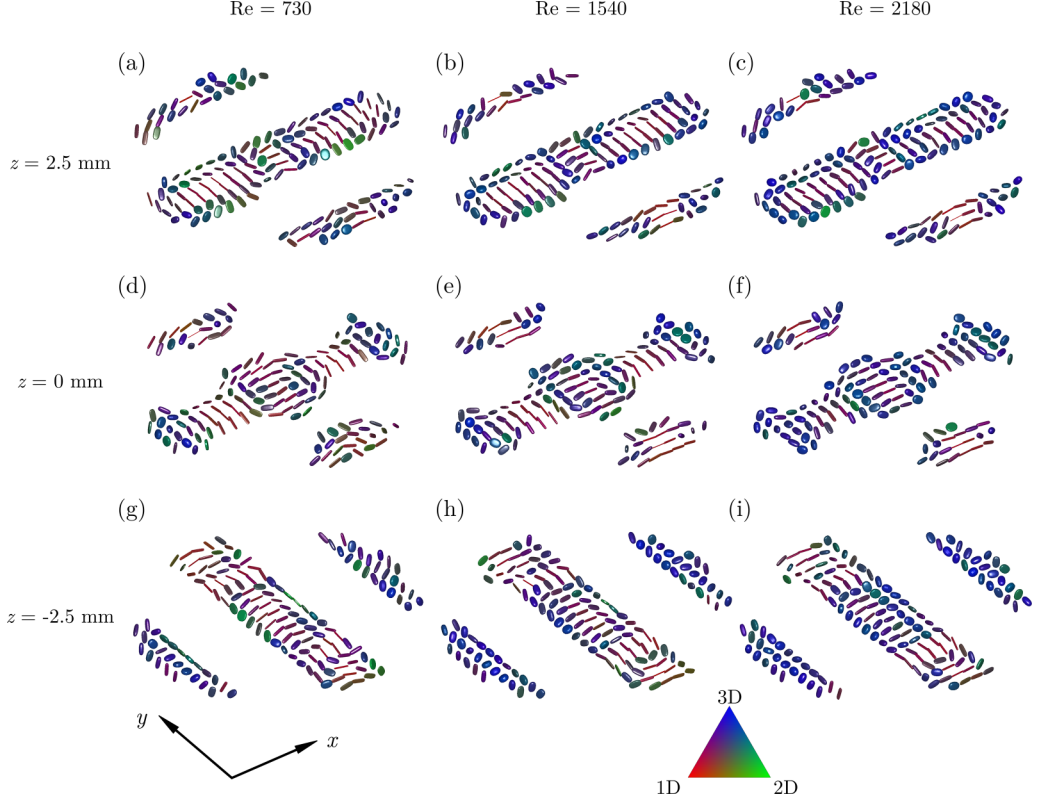


FIG. 10. Map of Reynolds stress tensor glyphs representing the anisotropy and orientation of local turbulence in the Schwarz Diamond column for three different Reynolds numbers at offset slice positions of (a)–(c) +2.5 mm, (d)–(f) 0 mm, (g)–(i) –2.5 mm. The ternary color map indicates if the turbulence is one-dimensional (rodlike), two dimensional (disklike), or three-dimensional (spherelike, isotropic).

From an engineering perspective, the transport of heat and material to the monolith surfaces may be affected by the turbulent structures in the channel centers due to the RST glyphs aligning perpendicular to the channel surfaces such that velocity fluctuations towards the surfaces are comparatively high. This feature is an example of how monolith structure affects prevailing flow phenomena. Hence, time-resolved MRI velocimetry may be a useful method to assist in the development of intensified process devices by informing the design of the surface geometry.

IV. CONCLUSIONS

To understand the suitability of 3D-printed porous materials for process intensification, we implemented a time-resolved spiral MRI velocity imaging sequence to study turbulent flow in a Schwarz Diamond TPMS monolith. This method enabled us to noninvasively measure the velocity field in a 2D plane at up to 260 frames per second. We confirmed that the experiments accurately measured the velocity within experimental uncertainty for mean channel velocity below 0.15 ms^{-1} ($\text{Re} = 1540$) and that image distortion was kept sufficiently low. From the velocity images, we could compute the full Reynolds stress tensor, which returned information about the turbulence kinetic energy, the local orientation, and the anisotropy of the turbulence. These data illustrated that the turbulence was anisotropic even at the highest Reynolds number studied here, where some regions in the column may have desirable mixing characteristics.

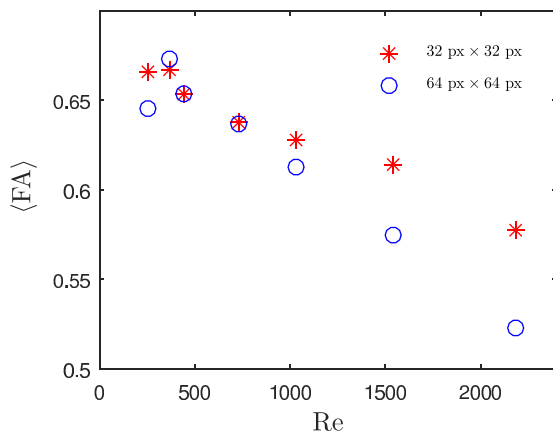


FIG. 11. Mean fractional anisotropy of the voxel-scale Reynolds stress tensor vs the Reynolds number.

Spectral analysis of the velocity images was used to compute the turbulent energy spectrum. The energy spectra showed that the modal energy was contained in eddies with length on the order of the hydraulic diameter due to the constraint imposed by the channel geometry. The energy cascade scaling exponent converged towards the $-5/3$ value predicted by the Kolmogorov law. The TKE dissipation rate was calculated using the mean square of the velocity fluctuation gradients. However, it is likely that this method underestimated the dissipation rate due to the image voxel size being considerably larger than the Kolmogorov length scale.

This study illustrates how time-resolved spiral MRI velocimetry can be used to characterize porous media flow phenomena in greater detail than is usually possible with standard imaging sequences by observation of millisecond-level flow features, provided that the main field is suitably homogeneous and that the k -space data are appropriately mapped. We envision that this approach could be extended to measuring other quantities, such as temperature, combined with spatial/temporal resolution enhancement, which would enable correlation of turbulent velocity fluctuations with the local heat transfer rate. The insights gained about turbulence properties may inform the design of intensified heat exchanger and reactor configurations that offer enhanced process performance compared to traditional devices.

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DATA AVAILABILITY

The data that support the findings of this article are publicly available on the Zenodo repository, Ref. [69].

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