

Magnetic resonance velocity imaging of turbulent gas flow in a packed bed of catalyst support pellets

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1 **Abstract**

2 Compressed sensing magnetic resonance methods have been used to image the time-averaged velocity
3 and turbulent kinetic energy in 3D for turbulent gas flowing through a bed of porous, hollow cylindrical
4 catalyst support pellets. Velocity and turbulent kinetic energy images were acquired at a spatial
5 resolution of 0.70 mm (x) × 0.70 mm (y) × 1.0 mm (z) for particle Reynolds numbers, Re_p , of 500,
6 2500 and 6500 in a bed with a tube-to-particle diameter ratio of 4.7. These data represent the first full-
7 field measurements of turbulent gas flow in packed beds of non-spherical pellets. The resulting images
8 reveal several interesting features of the hydrodynamics in this system. A large degree of flow
9 heterogeneity is observed in the bed, with regions of high-speed fluid observed near the walls and in
10 large voids, and regions of backflow observed in the wake of pellets, between pellets, and within the
11 pellet holes. For increasing Re_p , the normalized axial velocity at the wall is found to increase, and the
12 normalized turbulent kinetic energy becomes more homogeneous throughout the bed. The correlation
13 between the turbulent kinetic energy and time-averaged velocity shows that the highest turbulent
14 kinetic energy occurs in regions of intermediate time-averaged velocity. Further, the turbulent kinetic
15 energy profile at the pellet-scale is substantially different from the case of simple channel flow for
16 $Re_p \geq 2500$. Overall, these measurements clearly demonstrate the ability of magnetic resonance
17 methods for acquiring full-field flow data in packed bed systems using commercially-relevant pellets
18 and flow conditions.

19

20 **Keywords:** packed bed, MRI, magnetic resonance velocity imaging, turbulent flow, pellet shape

21

22

23 **1 Introduction**

24 **1.1 Hydrodynamics in packed beds**

25 Packed bed reactors are used extensively in the chemical process industry to facilitate many
26 important solid-catalysed reaction processes such as methanol synthesis, water-gas shift, steam
27 methane reforming and ethylene epoxidation, among others. Catalyst pellets used in these processes
28 come in a wide range of shapes and sizes [1,2]. Fluid is passed over the catalyst pellets, and reaction
29 proceeds on both the internal and external surface area of the pellet. Pellet size and shape have been
30 shown to significantly affect reactor performance [1–4], but due to the complex coupling between heat
31 transfer, mass transfer, and reaction rate, a unified rational approach to optimal pellet design does not
32 yet exist.

33
34 The overall performance of the reactor (conversion, selectivity) is strongly impacted by the local
35 (pellet-scale) heat and mass transfer properties in the bed, which are in turn impacted by the pellet-scale
36 hydrodynamics. Thus, to optimize the performance of packed bed reactors, it is imperative to
37 understand the hydrodynamics at the pellet scale. This is especially important for the narrow beds (tube
38 to particle diameter ratio, N , < 10) typically employed in highly endo/exothermic processes, where
39 pellet ordering effects at the wall strongly influence the bed hydrodynamics [5–7].

40
41 Most commonly, overall reactor metrics such as pressure drop, total temperature rise/drop,
42 average axial/radial dispersion, and feed conversion have been estimated using phenomenological
43 models with semi-empirical correlations developed from bed-scale experiments. Such experiments, and
44 thus the models developed from them, provide little information on the pellet-scale transport properties
45 that dictate the reactor performance [8,9]. To overcome this limitation, in recent years, numerous

46 researchers have conducted particle-resolved computational fluid dynamics (PR-CFD) simulations to
47 incorporate the effects of pellet-scale hydrodynamics and transport in reactor simulations, as recently
48 reviewed by Dixon & Partopour [10]. The accuracy of PR-CFD simulations is highly dependent on the
49 mesh quality, appropriate boundary conditions, and choice of turbulence model (where applicable).
50 Guardo *et al.* [11] evaluated different turbulent eddy viscosity models in the use of PR-CFD, finding
51 large differences in the resulting heat transfer coefficient and pressure drop. Recently, Karthik *et al.*
52 [12] and Ambekar *et al.* [13] compared several RANS based turbulence models to experimental and
53 direct numerical simulation (DNS) results, finding considerable differences in the turbulence properties
54 output from each model, and thus considerable differences in the predicted overall reactor performance
55 (measured as conversion and effectiveness factor). Therefore, it is important that experimental data are
56 used to validate, and improve the accuracy of, PR-CFD simulations, especially in the turbulent regime
57 where turbulence closure models are required. While bed-scale measurements such as pressure drop,
58 temperature rise/drop, dispersion and conversion can be used to evaluate CFD modelling, they are
59 macroscopic measurements that do not inform on the local, pellet-scale processes which influence and
60 limit the overall bed performance [8]. For a full, robust evaluation, experimental measurements at the
61 pellet-scale are required.

62

63 Numerous researchers have used particle imaging velocimetry (PIV) [14–20] (whereby statistical
64 ensembles of particles are tracked) and particle tracking velocimetry (PTV) [21,22] (whereby
65 individual particles are tracked) to experimentally study the velocity field at the pellet scale in narrow
66 packed beds. Wood *et al.* [14] compared 2D PIV measurements to CFD for laminar flow in a bed of
67 spheres, finding good agreement between experiment and simulation, with the deviations attributed to
68 geometrical mismatch. Thaker *et al.* [18] recently conducted 2D PIV measurements of turbulent flow

69 in a quarter-section bed of spheres and compared the results to corresponding PR-CFD simulations.
70 The authors found that the simulated time-averaged velocity field, strain rate and vorticity showed good
71 agreement with experiment, with small discrepancies attributed to geometrical mismatch and differing
72 upstream conditions. However, turbulent properties such as turbulent kinetic energy and dissipation rate
73 were significantly under predicted, attributed to limitations of the turbulence model used (SST $k - \omega$)
74 [18]. Further, the PIV setup used could only acquire two components of velocity, requiring the third to
75 be inferred for the calculation of turbulence quantities. Recently, a number of researchers have used
76 high resolution PIV to probe turbulent flows in small subsections of beds [19], and at the level of
77 individual pores [15–17]. Through the high spatiotemporal resolution afforded by state-of-the-art PIV
78 methods, these studies revealed fundamental insights into the spatiotemporal properties of the turbulent
79 flows, and the resulting pellet-scale transport properties, in model packed bed systems. Due to the
80 optical nature of PIV/PTV, all of the aforementioned studies were conducted for transparent spherical
81 packings with refractive index matching.

82
83 In contrast to PIV methods, magnetic resonance (MR) based methods do not require optical
84 transparency (rather only radio frequency (r.f.) transparency) making it possible to study flows, both
85 laminar and turbulent, in packed beds of real catalyst pellets. Additionally, MR measurements can be
86 made chemically-specific, making MR and in particular magnetic resonance imaging (or MRI), an
87 ideal tool for studying chemical engineering systems *in operando* [23,24]. Indeed, MR velocity
88 imaging (or MR velocimetry) has been extensively used to study various aspects of packed bed
89 hydrodynamics.

90

91 **1.2 Magnetic resonance velocity imaging in packed beds**

92 A brief review of relevant MR velocity imaging studies of flow in packed beds is given in this
93 section. For further details, the interested reader is referred to the reviews by Fukushima [25], Elkins &
94 Alley [26] and Gladden & Sederman [23].

95
96 MR velocity imaging has previously been utilized to study numerous aspects of packed bed
97 hydrodynamics. Early workers investigated pore scale flow profiles, and how the flow profile develops
98 with increasing Reynolds number in beds of spherical beads [27–29]. Sederman *et al.* [30] extended
99 this work by analysing correlations between the bed structure and hydrodynamics, finding that high
100 flow through a pore correlated with large pore cross-sectional area while high velocities were more
101 affected by the topology of the pore network. Sains *et al.* [31] extended MR velocity imaging to study
102 unsteady-laminar and weakly turbulent ($Re_p \sim 300$) single phase flows in packed beds using a rapid
103 acquisition technique to acquire 2D ‘snapshots’ of the unsteady velocity field. Aspects of multiphase
104 gas-liquid flow through packed beds have been investigated using MR imaging including the trickle to
105 pulse flow transition [32,33], and the investigation of interfacial momentum transfer by imaging both
106 the gas and liquid velocities [34]. MR velocity imaging has been used to complement other methods
107 used for investigating packed bed hydrodynamics including tracer experiments and CFD modelling.
108 Tang *et al.* [6] measured the radial velocity profile in narrow packed beds using MR velocity imaging
109 and subsequently compared these results to the residence time distributions (RTD) measured using
110 tracer methods in an effort to understand the influence of the bed aspect ratio on the RTD. Robbins *et*
111 *al.* [35] and Lovreglio *et al.* [5] compared CFD simulations of single-phase laminar and unsteady-
112 laminar ($Re_p < \sim 200$) flow through beds of spherical packings to MR velocity imaging
113 measurements, finding the (bed-scale) velocity distributions to agree well. Yang *et al.* [36] similarly
114 compared results obtained using MR velocity imaging and CFD for single phase laminar flow in a bed

115 of spheres, but found deviations between experiment and modelling at both the local and global scale.
116 These discrepancies were attributed to experimental limitations of the MR velocity imaging. Recently,
117 Clarke *et al.* [37] compared MR velocity imaging and CFD for laminar flow through a structured
118 packing ($Re_p < 30$), finding quantitative agreement between experiment and simulation.

119

120 As reviewed above, the vast majority of experimental studies of flow in packed beds, both MR
121 velocity imaging and otherwise, have been conducted in the laminar flow regime and on model (non-
122 porous, spherical) packings. MR is uniquely capable of acquiring full-field, spatially-resolved flow data
123 in beds of commercially relevant pellets, at the turbulent flow conditions of interest. However, to date,
124 this capability of MR has not yet been fully exploited, due in part to a number of technical challenges in
125 acquiring quantitative velocity images in heterogeneous, turbulent systems: namely, (1) difficulties
126 with low signal-to-noise ratio (SNR) in turbulent systems; (2) MR imaging artefacts inherent when
127 imaging unsteady systems; (3) magnetic field inhomogeneity issues when using real catalyst pellets; (4)
128 unfeasibly long acquisition times. Recent method developments and demonstrations have made
129 important steps towards tackling these challenges. Notably, the development and application of pure
130 phase-encode imaging techniques for robust image acquisition with high SNR [38–41], and the
131 development and application of non-linear compressed sensing strategies to accelerate image
132 acquisition times [42,43]. These recent developments now enable the experimental acquisition of full-
133 field turbulent flow data in packed beds using the real pellets and process conditions of true commercial
134 relevance.

135

136 **1.3 Structure and objectives of this work**

137 The objective of this work is to develop and demonstrate the novel ability of MR velocity
138 imaging methods to acquire experimental full-field data of the turbulent hydrodynamics in a

139 commercially-relevant packed bed system at the pellet scale. As identified in the recent review of
140 Dixon & Partopour [10] tomographic studies are required to better understand the hydrodynamics in
141 these systems, and to build up a database for the validation and calibration of numerical flow
142 simulations.

143

144 This paper is structured as follows. First, the relevant theory underpinning the quantification of
145 turbulent flows using magnetic resonance is presented. Details of the experimental rig and magnetic
146 resonance sequences used to acquire 3D images of the time-averaged velocity and turbulent kinetic
147 energy (TKE) in a narrow packed bed are presented. Porous α -alumina cylindrical pellets with an
148 interior hole are used as the packing material; α -alumina is a commonly used support material for
149 commercial processes, and this pellet shape has been the focus of recent PR-CFD studies [1,2,44–46].
150 The results acquired at three flow conditions ($Re_p = 500, 2500, 6500$) are presented and discussed.
151 The data are used to investigate the radial velocity profile and TKE distribution as a function of Re_p .
152 Finally, the results are discussed in the context of previously reported computational and experimental
153 studies. The results provide experimental insight into fundamental aspects of the hydrodynamics in
154 packed beds with the pellets and flow conditions of true commercial relevance.

155

156 **2 Theoretical Background**

157 In this section, a brief introduction to the theory of MR measurements of turbulent flows is given.
158 The theory presented here draws upon the works of Gao & Gore [47], Gatenby & Gore [48], Kuethe &
159 Gao [49], Dyverfeldt *et al.* [50] Elkins *et al.* [51], and Cooper *et al.* [41], as well as the theses of
160 Dyverfeldt [52] and Cooper [53].

161

162 In magnetic resonance, nuclei with non-zero spin (commonly ^1H , ^{13}C , or ^{19}F) are probed by
 163 measuring the precession of the nuclear spin in a strong magnetic field. The precession frequency, ω , is
 164 directly proportional to the external field strength, B_0 :

$$\omega = \gamma B_0 , \quad (1)$$

165 where γ is the gyromagnetic ratio of the nucleus under investigation. By varying the magnetic field
 166 strength in space, through the use of a magnetic field gradient \mathbf{g} , the precession frequency can be made
 167 spatially dependent:

$$\omega(\mathbf{r}) = \gamma(B_0 + \mathbf{g} \cdot \mathbf{r}) , \quad (2)$$

168 where \mathbf{r} is the position vector. The acquired MR signal is the ensemble average of all spins in the
 169 system, recorded as a complex signal with both magnitude and phase. The signal phase, ϕ , can be
 170 expressed by integrating Equation (2) and Taylor expanding the position \mathbf{r} (and dropping the B_0 term
 171 which merely provides a linear offset):

$$\phi = \gamma \int \mathbf{g}(t) \cdot \mathbf{r}(t) dt = \gamma \left[\mathbf{r} \cdot \int \mathbf{g}(t) dt + \frac{d\mathbf{r}}{dt} \cdot \int t\mathbf{g}(t) dt + \dots \right]. \quad (3)$$

172 Hence, it is seen from Equation (3) that the application of a gradient encodes for both the initial
 173 position of the spin, \mathbf{r} , and the velocity, $\mathbf{u} = \frac{d\mathbf{r}}{dt}$, (as well as higher order terms, which can
 174 typically be neglected for short gradient durations). To acquire spatially resolved measurements
 175 of velocity (velocity images), an MR sequence is utilized whereby position and velocity are
 176 encoded for by independently varying the 0th moment of the applied gradient, $\int \mathbf{g}(t) dt$, and the
 177 1st moment of the applied gradient, $\int t\mathbf{g}(t) dt$, respectively.

178

179 The well-known Reynolds decomposition can be used to decompose the velocity in a
 180 turbulent system, $\mathbf{u}(t)$, into a time-averaged component, $\bar{\mathbf{u}}$, and a fluctuating component, $\mathbf{u}'(t)$, such

181 that $\mathbf{u}(t) = \bar{\mathbf{u}} + \mathbf{u}'(t)$. The dynamics of $\mathbf{u}'(t)$ are typically described as a stochastic process with zero
182 mean ($\langle \mathbf{u}'(t) \rangle = 0$) and a characteristic correlation time, τ_c . There exist two approaches to probing
183 turbulent flows with MR velocity imaging: (i) the ‘snapshot’ approach, and (ii) the time-averaged
184 approach [26]. In the ‘snapshot’ approach, the acquisition time, TA , of the MR velocity imaging
185 experiment is short such that $TA \ll \tau_c$, giving an instantaneous measure of the velocity field, $\mathbf{u}(t)$, in
186 the system. In the time-averaged approach $TA \gg \tau_c$ and therefore the instantaneous nature of $\mathbf{u}(t)$ is
187 not accessible. However, the time-averaged velocity field $\bar{\mathbf{u}}$ and information on the statistical nature of
188 $\mathbf{u}'(t)$ can still be measured in the time-averaged approach. In the present work, the flows studied are
189 highly turbulent with τ_c on the order of 10 - 100 ms (Supplementary Information). In a packed bed,
190 where the catalyst pellets give rise to large magnetic susceptibility differences, it is not possible to
191 obtain a 3D image of suitable spatial resolution on the fast timescale required for the ‘snapshot’
192 approach. However, the time-averaged MR approach can be implemented. This is the approach used in
193 this work to measure both the time-averaged velocity, $\bar{\mathbf{u}}$, and the mean-square turbulent velocity
194 fluctuations, $\langle \mathbf{u}'^2 \rangle$.

195

196 **2.1 Measuring the time-averaged velocity, $\bar{\mathbf{u}}$**

197 For the time-averaged approach, the mean of the velocity fluctuations, $\langle \mathbf{u}' \rangle$, averages to zero
198 over the acquisition duration and the resulting signal phase contains information on the time-averaged
199 velocity field, $\bar{\mathbf{u}}$. For a bipolar pair of gradient pulses of amplitude g , duration δ , and separated by time
200 Δ , the moments of the gradient waveform are $M_0 = 0$ and $M_1 = g\delta\Delta$, and from Equation (3), the
201 resulting phase shift is:

$$\phi = \gamma\Delta\delta\mathbf{g} \cdot \bar{\mathbf{u}}. \quad (4)$$

202 Therefore, the phase of the MR signal, ϕ , can be used to quantify the time-averaged velocity, $\bar{\mathbf{u}}$,
 203 and, by combining with an appropriate imaging sequence, an image of $\bar{\mathbf{u}}$ is acquired.

204

205 **2.2 Measuring the mean square velocity fluctuations, $\langle \mathbf{u}'^2 \rangle$**

206 Information characterising \mathbf{u}' is also obtained using the same pair of bipolar gradients. Whilst
 207 the mean of the velocity fluctuations, $\langle \mathbf{u}' \rangle$, averages to zero over the acquisition duration, the mean
 208 square velocity fluctuations, $\langle \mathbf{u}'^2 \rangle$, can be measured. The mean square velocity fluctuations, $\langle \mathbf{u}'^2 \rangle$,
 209 cause a spread of displacements and thus a spread of phase shifts, leading to signal attenuation. The
 210 ability to measure the components of the Reynolds stress tensor, $\langle u'_i u'_j \rangle$, for turbulent flows using MR
 211 methods is well known for both medical [50,52] and engineering applications [54–56]. Here, we
 212 present the basic theory of these measurements, including accounting for the effects of molecular
 213 diffusion and intra-voxel mean velocity variance (shear).

214

215 For a Gaussian distribution of velocity (displacement), and equivalently for low gradient
 216 strengths (as employed in the present work), the signal attenuation is written:

$$|S| = |S_0| \exp \left[-\frac{\langle \phi^2 \rangle}{2} \right] = |S_0| \exp \left[-\frac{\langle (\gamma \int \mathbf{g}(t) \cdot \mathbf{r}(t) dt)^2 \rangle}{2} \right], \quad (5)$$

217 where $|S|$ is the signal magnitude acquired at gradient amplitude $g > 0$, $|S_0|$ is the signal magnitude at
 218 $g = 0$, and the angular brackets $\langle \cdot \rangle$ denote taking the ensemble average over all spins. Note that strictly
 219 speaking the phase dispersion term, $\langle \phi^2 \rangle$, should be written as the central second moment ($\langle \phi^2 \rangle -$
 220 $\langle \phi \rangle^2$), but since all attenuation processes in this work are analysed as fluctuations about the mean, only
 221 $\langle \phi^2 \rangle$ is written for notational simplicity. The assumption of a Gaussian turbulent velocity distribution is
 222 supported by the work of Gao and Gore [47], and further work by Dyverfeldt *et al.* [50] found Equation

223 (5) to be an excellent approximation for non-Gaussian distributions for small gradient amplitude, g .
 224 For the bipolar gradient pulse pair previously described, the phase shift of an individual spin, ϕ ,
 225 caused by application of the gradients along an arbitrary axis, denoted i , is written (using
 226 Equation (3)):

$$\phi = \gamma \int \mathbf{g}(t) \cdot \mathbf{r}(t) dt = \gamma g \int_0^\delta [z_i(t + \Delta) - z_i(t)] dt, \quad (6)$$

227 where g is the amplitude of the gradients applied along the i axis, δ and Δ are defined in Section
 228 2.1, and $z_i(t)$ is the position of a spin at time t along the i axis (i.e. in the direction of the gradients).
 229 Following Gatenby and Gore [48], the ensemble mean square phase, due to turbulent
 230 fluctuations, resulting from the bipolar pair is written [53]:

$$\langle \phi_{\text{turb}}^2 \rangle = \gamma^2 g^2 \int_0^\delta \int_0^\delta \langle [z_i(t_1 + \Delta) - z_i(t_1)] \times [z_i(t_2 + \Delta) - z_i(t_2)] \rangle dt_1 dt_2. \quad (7)$$

231 To compute the above integral, a model must be invoked to describe the statistical nature of the
 232 velocity fluctuations in the system. Taylor introduced the Lagrangian autocorrelation function to
 233 describe turbulent velocity fluctuations [57]. Further, Taylor found that this autocorrelation is
 234 well described by an exponential:

$$R(T) = \frac{\langle u_i'(t) u_i'(t + T) \rangle}{\langle u_i'^2 \rangle} = \exp(-T/\tau_c). \quad (8)$$

235 The exponential form of the Lagrangian autocorrelation is well suited to describe a wide variety
 236 of turbulent flows, and the specific conditions where it holds are discussed by Taylor [57] and
 237 Landau & Lifshitz [58]. Expanding the integrand [47], and substituting in the exponential
 238 Lagrangian autocorrelation function (Equation (8)), the integration of Equation (7) yields [53]:

$$\begin{aligned} \langle \phi_{\text{turb}}^2 \rangle &= 2\gamma^2 g^2 \langle u_i'^2 \rangle \tau_c \left[\delta^2 (\Delta - \delta/3) - 2\tau_c^2 \delta \right. \\ &\left. + \tau_c^3 \left(2 - 2e^{-\frac{\Delta}{\tau_c}} + e^{-\frac{(\Delta-\delta)}{\tau_c}} + e^{-\frac{(\Delta+\delta)}{\tau_c}} - 2e^{-\frac{\Delta}{\tau_c}} \right) \right]. \end{aligned} \quad (9)$$

239 The terms in Equation (9) containing τ_c , δ , and Δ can be grouped together into a function
 240 $f(\delta, \Delta, \tau_c)$. Then, using Equation (5), the signal attenuation due to turbulence is written as [51]:

$$|\hat{S}_{\text{turb}}| = \frac{|S_{\text{turb}}|}{|S_0|} = \exp[-\gamma^2 g^2 \langle u_i'^2 \rangle f(\delta, \Delta, \tau_c)], \quad (10)$$

241 where $|\hat{S}_{\text{turb}}|$ denotes that normalized signal attenuation due to turbulent velocity fluctuations,
 242 and the ‘turb’ subscript denotes that only attenuation due to turbulence has been accounted for
 243 (other sources of attenuation are discussed later). In the limit where the turbulent fluctuations are
 244 constant over the encoding period, $\tau_c \gg \Delta$ and $\tau_c \gg \delta$, the temporal terms in Equation (9) reduce
 245 to (by taking terms up to 4th order) [47,50]:

$$f(\delta, \Delta, \tau_c) = \frac{\delta^2 \Delta^2}{2}. \quad (11)$$

246 Combining Equation (10) and (11) gives:

$$|\hat{S}_{\text{turb}}| = \exp\left(-\gamma^2 g^2 \langle u_i'^2 \rangle \frac{\delta^2 \Delta^2}{2}\right). \quad (12)$$

247

248 To this point, only the attenuation due to turbulent velocity fluctuations has been
 249 considered. As is clear in Equation (5), signal attenuation results from any process that causes a
 250 spread in displacements (and thus velocities). MR probes the total displacement variance within
 251 a voxel (which includes intra-voxel turbulence down to the Kolmogorov scale) [50]. In addition
 252 to turbulence, variation of time-averaged velocity across voxels of a finite width and molecular
 253 diffusion both contribute to the variance of molecular displacement within a voxel, and thus the

254 signal attenuation. These sources of phase dispersion, and thus signal attenuation, are depicted
 255 schematically in Figure 1. To accurately estimate the mean square turbulent velocity fluctuations
 256 in the present work, both of these sources of additional attenuation are accounted for. As
 257 discussed by Dyverfeldt *et al.* [50,52], this can be achieved by considering the intra-voxel mean
 258 velocity variance (due to shear across a voxel) as an additional attenuation process in addition to
 259 the stochastic turbulent fluctuations. Molecular diffusion can also be considered as a separate
 260 random process, and while its effect has been neglected in previous (liquid-phase) measurements
 261 of turbulence, we consider it here due to its significant effect in the gas phase. Therefore,
 262 following Dyverfeldt *et al.*, [50,59], the total attenuation considering the aforementioned
 263 processes is:

$$|S| = |S_0| |\hat{S}_{\text{turb}}| |\hat{S}_{\text{MVV}}| |\hat{S}_D|, \quad (13)$$

264 where $|\hat{S}_{\text{MVV}}|$ is the normalized signal attenuation from time-averaged mean velocity variance
 265 across a voxel, and $|\hat{S}_D|$ is the normalized signal attenuation due to diffusion. The mean velocity
 266 variance attenuation is written, combining Equations (4) and (5), as:

$$|\hat{S}_{\text{MVV}}| = \exp\left(-\frac{\gamma^2 g^2 \delta^2 \Delta^2}{2} \sigma_{\bar{u}_i}^2\right), \quad (14)$$

267 where $\sigma_{\bar{u}_i}^2 = \langle \bar{u}_i^2 \rangle_{\text{vox}} - \langle \bar{u}_i \rangle_{\text{vox}}^2$ is the time-averaged velocity variance within a voxel [50], which
 268 can be estimated from an image of the time-averaged velocity \bar{u}_i . The normalized attenuation due to
 269 diffusion is given by the well-known Stejskal-Tanner equation [60]:

$$|\hat{S}_D| = \exp(-\gamma^2 \delta^2 g^2 D_m (\Delta - \delta/3)), \quad (15)$$

270 where D_m is the molecular diffusion coefficient. Combining Equations (12) - (15) gives:

$$|S| = |S_0| \exp\left(-\gamma^2 \delta^2 g^2 \left[\frac{\Delta^2}{2} (\langle u_i'^2 \rangle + \sigma_{\bar{u}_i}^2) + D_m (\Delta - \delta/3)\right]\right). \quad (16)$$

271 Finally, rearranging Equation (16) for $\langle u_i'^2 \rangle$:

$$\langle u_i'^2 \rangle = \left[\frac{2}{\Delta^2} \left(\frac{1}{\gamma^2 \delta^2 g^2} \ln \left| \frac{S_0}{S} \right| - D_m (\Delta - \delta/3) \right) - \sigma_{\bar{u}_i}^2 \right]. \quad (17)$$

272 Equation (17) expresses the mean square velocity fluctuations (when $\tau_c \gg \Delta$ and $\tau_c \gg \delta$) in
273 terms of the signal attenuation from the application of a bipolar gradient pair, considering the
274 effects of both intra-voxel mean velocity variance (shear) and molecular diffusion.

275

276 In the context of modelling turbulent flows, Equation (17) is immensely useful as it
277 provides a means to experimentally measure the Reynolds stresses (mean square turbulent
278 fluctuations) [51]. The TKE, an isotropic measure of the local turbulence intensity, can be
279 computed from these measurements by:

$$k = \frac{1}{2} (\langle u_x'^2 \rangle + \langle u_y'^2 \rangle + \langle u_z'^2 \rangle), \quad (18)$$

280 where $\langle u_x'^2 \rangle$, $\langle u_y'^2 \rangle$, $\langle u_z'^2 \rangle$ are the velocity fluctuations in the x, y and z directions, measured in
281 separate experiments by applying magnetic field gradients independently in each respective
282 direction.

283

284 2.2.1 Estimating the intra-voxel mean velocity variance, $\sigma_{\bar{u}_i}^2$

285

286 To estimate the intra-voxel mean velocity variance, $\sigma_{\bar{u}_i}^2 = \langle \bar{u}_i^2 \rangle_{\text{vox}} - \langle \bar{u}_i \rangle_{\text{vox}}^2$, the intra-voxel
287 time-averaged velocity profile must be assumed. Naïvely, this can be assumed to be a linear velocity
288 profile, thus giving a boxcar-shaped velocity distribution. With this assumption, $\sigma_{\bar{u}_i}^2$ was estimated
289 from the time-averaged velocity images. To demonstrate, consider the variation in the velocity in the i

290 direction, \bar{u}_i , across a voxel. Considering the variance of a uniform distribution, the intravoxel variance
291 of \bar{u}_i due to variation in the z-direction is written:

$$\sigma_{\bar{u}_i}^{2z} = \frac{\left(\frac{\partial \bar{u}_i}{\partial z} dz\right)^2}{12}, \quad (19)$$

292 where the factor of 12 in the denominator arises from the variance of a uniform distribution. This can
293 be done in each direction and, since the directions are independent, the total intra-voxel variance for \bar{u}_i
294 can be written:

$$\sigma_{\bar{u}_i}^2 = \sigma_{\bar{u}_i}^{2x} + \sigma_{\bar{u}_i}^{2y} + \sigma_{\bar{u}_i}^{2z}. \quad (20)$$

295 Note that the derivatives in Equation (19) were taken numerically from the time-averaged velocity
296 images using 2nd order central differencing. The intra-voxel time-averaged velocity variance estimated
297 from Equation (20) was subsequently used to quantify $\langle u_i'^2 \rangle$, the mean square turbulent velocity
298 fluctuations (Equation (17)).

299 Typically, accounting for the intra-voxel velocity variance in Equation (17) had only a small effect on
300 the resulting value of $\langle u_i'^2 \rangle$. Indeed, simply neglecting the shear effect ($\sigma_{\bar{u}_i}^2 = 0$) rather than using the
301 estimate from Equation (20) was found to result in <10% change in $\langle u_i'^2 \rangle$ (calculated using Equation
302 (17)) for 97% of all voxels. Thus, although approximating the intra-voxel velocity profile as linear
303 might be considered a crude approximation, it is not considered to influence the conclusions drawn
304 from this work.

305

306

307

308

309 2.2.2 *Uncertainty analysis*

310

311 To estimate the uncertainty in measured vales of $\langle u_i'^2 \rangle$, and subsequently k , the following sources of

312 uncertainty were considered:

313 1. Uncertainty in $|S|$: following Elkins *et al.* [51], taken conservatively as the average signal
 314 magnitude outside the bed region in an image acquired at $g > 0$. This source of error was
 315 propagated as a random error.

316 2. Uncertainty in $|S_0|$: taken as the average signal magnitude outside the bed region in an
 317 image acquired at $g = 0$. This source of error was propagated as a random error.

318 3. Uncertainty in $\sigma_{\bar{u}_i}^2$: given the assumptions required to compute the intra-voxel velocity
 319 variance, this source of error was conservatively taken as $d\sigma_{\bar{u}_i}^2 = \sigma_{\bar{u}_i}^2$. This source of error
 320 was propagated as a systematic error.

321 4. Uncertainty in D_m : given uncertainty in the local temperature and pressure within the bed,
 322 the uncertainty in molecular diffusivity was taken as $dD_m = 0.1D_m$. This source of error
 323 was propagated as a systematic error.

324 It is important to note that consideration of both $d|S|$ and $d|S_0|$ accounts for both random noise in the
 325 attenuation images, and pseudo-random ‘ghosting’ artefacts due to turbulent fluctuations during image
 326 acquisition [51]. Considering Equation (17) and using standard error analysis propagation [61], the total
 327 uncertainty in per-voxel values of $\langle u_i'^2 \rangle$ was then calculated as:

$$d\langle u_i'^2 \rangle = \left(\left(\frac{\partial \langle u_i'^2 \rangle}{\partial |S|} d|S| \right)^2 + \left(\frac{\partial \langle u_i'^2 \rangle}{\partial |S_0|} d|S_0| \right)^2 + \left(\frac{\partial \langle u_i'^2 \rangle}{\partial \sigma_{\bar{u}_i}^2} d\sigma_{\bar{u}_i}^2 \right)^2 + \left(\frac{\partial \langle u_i'^2 \rangle}{\partial D_m} dD_m \right)^2 \right)^{\frac{1}{2}}. \quad (21)$$

328 Subsequently, $d\langle u_i'^2 \rangle$ was used to estimate the uncertainty in TKE, dk , by considering Equation (18) in
 329 a similar manner. When considering the uncertainty in the bed-average value of k (which is the result
 330 of averaging the value of k over many voxels), random and systematic sources of uncertainty were
 331 considered separately and scaled in an appropriate fashion (random errors reduce $\propto N_{\text{vox}}^{-1/2}$ where N_{vox}

332 is the number of voxels averaged over, whereas systematic errors are simply averaged and do not scale
333 down with N_{vox}).

334 **3 Experimental**

335 **3.1 Materials and equipment**

336 Flow through the packed bed was investigated using an acrylonitrile butadiene styrene (ABS)
337 plastic pipe ($d_t = 41$ mm I.D., tube length of 2 m) packed with porous hollow cylindrical α -alumina
338 catalyst support pellets (Shell Global Solutions, Amsterdam), details of which are given in Table 1.
339 The spherical equivalent particle diameter, $d_e = 6V_p/A_p$ (e.g. the sphere with the same surface area
340 to volume ratio as the pellet), is also given in Table 1. The pellet volume, $V_p = \frac{\pi}{4}L(d_o^2 - d_{\text{hole}}^2)$, and
341 external surface area, $A_p = \pi L(d_o + d_{\text{hole}}) + \frac{\pi}{2}(d_o^2 - d_{\text{hole}}^2)$, were calculated based on the average
342 pellet shape without accounting for surface roughness. The resulting tube to particle diameter ratio, N ,
343 used in this study was $N = d_t/d_o = 4.7$. Pellets were loaded into the tube by pouring the pellets
344 slowly through a 25 mm funnel. To be consistent with loading practices for industrial-scale multi-
345 tubular reactors, no tapping for bed consolidation was conducted. Note that all flow imaging
346 experiments reported in this work were conducted using the same packed bed (i.e., the bed was not
347 repacked over the course of the experimental campaign), and thus all data were collected on the same
348 packing structure. Confirmation that the packing structure remained the same throughout the
349 experiments is shown in the Supplementary Information.

350

351 Sulphur hexafluoride (SF_6) was used as the nuclear magnetic resonance (NMR)-active gas in this
352 study, due to its high density, high gyromagnetic ratio, and favourable NMR relaxation properties, as
353 discussed by Sankey *et al.* [34] and Ramskill *et al.* [62]. A schematic of the closed-loop flow rig used
354 for MR velocity imaging is shown in Figure 2. The rig consists of an SF_6 gas cylinder (BOC, 99.9%

355 purity), a high and low pressure vessel, in-line pressure regulators, a DILO Piccolo compressor (model
356 B022R01), and a Bronkhorst mass flow controller (model F-113AC-M50-AAD-55-E). Gas was
357 delivered through 1/8" Swagelok tubing. Plastic perforated plate distributors with 2 mm holes were
358 glued into the pipe at the inlet and outlet to provide evenly distributed flow to the bed. The flow was
359 imaged approximately 1 m downstream of the tube entrance and approximately 0.95 m upstream of the
360 tube exit to ensure the flow was fully developed.

361

362 **3.2 Magnetic resonance**

363 All experiments were conducted using a 4.7 T vertical bore superconducting magnet (Bruker
364 Spectrospin DMX200), employing a 64 mm birdcage r.f. coil tuned to a ^{19}F resonant frequency of
365 187.64 MHz used for signal excitation and acquisition. A tri-axial gradient set was used (Bruker Mini
366 0.36) providing a maximum gradient strength of 13.1 G cm^{-1} in all three orthogonal directions.

367

368 Spin-echo single point imaging (SESPI) was used for all imaging experiments conducted in this
369 work [38,63]. This pulse sequence was selected due to the freedom it allows for designing compressed
370 sensing sampling patterns, as well as the robustness of this pure-phase encode method for imaging
371 turbulent flows with high velocities [40,54]. The SESPI pulse sequence shown in Figure 3 was used to
372 acquire 3D images of the bed structure, the three orthogonal components of time-averaged velocity,
373 $\bar{u}_x, \bar{u}_y, \bar{u}_z$ and the three components of the TKE, $\langle u_x'^2 \rangle, \langle u_y'^2 \rangle, \langle u_z'^2 \rangle$. To image each of the
374 aforementioned parameters at a given flow condition using the SESPI sequence, the only parameters
375 that were varied were the magnitude and direction of the flow encoding gradient, g . All other imaging
376 parameters were maintained constant and are described here. Images were acquired on a $64 \times 64 \times 64$
377 voxel matrix with a field of view (FOV) of 45 mm (x) \times 45 mm (y) \times 64 mm (z) resulting in images
378 with a resolution of 0.70 mm (x) \times 0.70 mm (y) \times 1.0 mm (z). Spatial resolution was achieved by

379 incrementing the position encoding gradient (G_{phase} in Figure 3) independently in each direction. The
380 duration of G_{phase} was 0.2 ms, with a ramp time of 0.02 ms and a gradient stabilization delay of 0.2
381 ms. A hard 90° r.f. pulse of duration $92 \mu\text{s}$ was used for signal excitation. To act as an effective
382 bandwidth filter (and avoid spurious background signals) a Gaussian shaped 180° soft pulse of duration
383 $512 \mu\text{s}$ was used for spin-echo refocusing. The echo time was 2.47 ms (with the exception of
384 experiments conducted at $Re_p = 500$, where the echo time was 5.45 ms), and a recycle time of 100 ms
385 was used (much longer than the measured $T_1 \approx T_2 \approx 15$ ms due to gradient cooling limitations). The
386 resulting signal was acquired with a sweep width of 100 kHz and 8 complex points from each free
387 induction decay were collected and averaged together to increase the SNR (adding more was found to
388 result in significant T_2^* weighting). Four scans were collected to complete a full phase cycle of the
389 SESPI pulse sequence.

390

391 For imaging of the bed structure, the flow encoding gradient was turned off, $g = 0$. For imaging
392 of the time-averaged velocity, $\bar{u}_x, \bar{u}_y, \bar{u}_z$, and the components of the TKE, $\langle u_x'^2 \rangle, \langle u_y'^2 \rangle, \langle u_z'^2 \rangle$, motion
393 encoding was achieved using gradient pulses (depicted as G_{flow} in Figure 3) of duration $\delta = 0.48$ ms
394 on either side of the 180° r.f. refocusing pulse, and a flow encoding duration of $\Delta = 1.69$ ms for
395 experiments at $Re_p = 2500, 6500$ and $\Delta = 4.67$ ms for experiments at $Re_p = 500$.

396

397 Details specific to the acquisition of the time-averaged velocity images and imaging of the TKE
398 components are now given. For time-averaged velocity imaging, the gradient strength, g , was set
399 empirically to avoid flow aliasing (phase wraparound), typical gradient strengths used ranged from $g =$
400 $0.2 - 2 \text{ G cm}^{-1}$. Two images were acquired at equal, but opposite gradient strengths ($+g, -g$), the
401 phase difference was taken, and the resulting phase difference map was used to compute an image of

402 the time-averaged velocity, \bar{u} , using Equation (4). Each velocity image acquired was corrected for
403 phase shifts caused by gradient eddy currents by acquiring images at zero-flow conditions and
404 subtracting the zero-flow phase map from the phase map acquired at flowing conditions. For imaging
405 of the TKE components, $\langle u_x'^2 \rangle$, $\langle u_y'^2 \rangle$, $\langle u_z'^2 \rangle$, identical flow encoding parameters were used except for
406 the gradient strength. As discussed by Dyverfeldt *et al.* [50,59] the optimal gradient strength to
407 maximize the trade-off between attenuation contrast and SNR occurs at an average (spatially
408 unresolved) signal attenuation of $|S|/|S_0| \approx 0.6$. The gradient strength was selected empirically to give
409 approximately this attenuation for each TKE experiment, with values ranging from $g = 1.5 - 5 \text{ G cm}^{-1}$.
410 Two images were acquired, one at $g = 0$ to give the unattenuated signal, S_0 , and one at finite g to give
411 the attenuated signal, S . An image of the mean square velocity fluctuation $\langle u_i'^2 \rangle$ was then computed
412 using Equation (17) and by repeating this in three orthogonal directions, a map of the TKE was
413 obtained as follows from Equation (18). Prior to acquiring images of the TKE, the long τ_c assumption
414 used to derive Equation (17) was verified by using variable Δ pulsed field gradient NMR to measure
415 the average τ_c in the bed at each flow condition. Full details are given in the Supplementary
416 Information. The resulting values of the correlation time ranged between $\tau_c = 7 \text{ ms}$ and $\tau_c = 142 \text{ ms}$
417 (Table S1). It was found that in all cases, use of the long τ_c approximation is expected to give <10%
418 error in the measurement of $\langle u_i'^2 \rangle$.

419

420 The uncertainty in the time-averaged velocity images, calculated based on the SNR of the
421 acquired images [64], was $< 0.2\bar{u}/U_{\text{int}}$. The uncertainty for the TKE measurements is presented in
422 Section 4.3.1. To further confirm the quantitative nature of the time-averaged flow images, the flow
423 rate at each axial position was calculated from the 3D images of time-averaged velocity in the axial
424 direction (\bar{u}_z), and compared to the known flow rate as set on the calibrated mass flow controller. As

425 shown in the Supplementary Information, in all cases the MR calculated flow rate agreed within
 426 $\leq 6.2\%$ discrepancy over a 45 mm central section of the imaging FOV (in the z-direction), thereby
 427 confirming the quantitative nature of the flow images. This central 45 mm section was used for all
 428 analysis and visualization shown in this work (providing a region approximately 5 pellet layers high).
 429 Further, for all image visualization and spatial analysis conducted, the \mathbf{k} -space data were zero-filled
 430 from $64 \times 64 \times 64$ to $128 \times 128 \times 128$ to increase the available spatial resolution. Image
 431 reconstruction and analysis were conducted using Matlab 2021 (Mathworks), and 3D image rendering
 432 and individual pellet segmentation were conducted using Amira-Avizo 2021.1 (ThermoFisher
 433 Scientific).

434

435 **3.3 Image reconstruction**

436 Compressed sensing [65] was implemented in this work to accelerate data acquisition. 3D
 437 sampling patterns were generated as follows: a fully sampled image of the bed was collected at zero-
 438 flow and with no flow encoding gradient ($g = 0$) to obtain a 3D image of the bed structure. The
 439 parameter-free method outlined by Karlsons *et al.* [66] was then used to generate an appropriate 3D
 440 sampling pattern for the bed under study. Total variation (TV) regularization was used to reconstruct
 441 the undersampled images, due to its good edge preservation and to be consistent with previous work
 442 [41,62]. The reconstruction problem for TV regularization is expressed as:

$$\mathbf{x} \in \operatorname{argmin}_{\mathbf{x}} \left\{ \frac{1}{2} \|\mathbf{y} - W\mathcal{F}\mathbf{x}\|_2^2 + \alpha \|\nabla\mathbf{x}\|_2 \right\}, \quad (22)$$

443 where \mathbf{y} is the acquired (undersampled) MR signal in \mathbf{k} -space, W is the sampling scheme, \mathcal{F} is the
 444 Fourier transform operator, α is the regularization parameter, and \mathbf{x} is the reconstructed image.
 445 Equation (22) was solved numerically using the Object Oriented Mathematics for Inverse Problems
 446 (OOMFIP) code developed by Benning [67].

447

448 The fully sampled 3D image of the bed structure was used to optimize the compressed sensing
449 approach, namely the fraction of \mathbf{k} -space sampled, the regularization parameter α , and the number of
450 Bregman iterations used. Details of this optimization can be found in the Supplementary Information.
451 For optimal trade-off between image quality and acquisition acceleration, $\alpha = 0.1$, 8 Bregman iterations
452 were used, and 16% of \mathbf{k} -space was sampled. The acquisition time for the fully sampled image of the
453 bed structure was 29 h. For images of the time-averaged velocity, $\bar{u}_x, \bar{u}_y, \bar{u}_z$, and the components of
454 the TKE, $\langle u_x'^2 \rangle, \langle u_y'^2 \rangle, \langle u_z'^2 \rangle$ (where 2 images are required at different values of g) the implementation
455 of compressed sensing reduced the acquisition time for a 3D image of one component of the time-
456 averaged velocity or TKE from 58 h to 9 h, a $6.3\times$ acceleration in data acquisition time. Thus, the 6
457 flow images comprising the full time-averaged velocity vector, $\bar{u}_x, \bar{u}_y, \bar{u}_z$, and all three components of
458 the TKE, $\langle u_x'^2 \rangle, \langle u_y'^2 \rangle, \langle u_z'^2 \rangle$ took 54 h in total to acquire at a given flow rate using compressed
459 sensing. Note these long acquisition time arise as a result of sampling a large 3D raster of \mathbf{k} -space and
460 not from the need to sufficiently sample the temporal dynamics of the turbulent flows (whose
461 correlation times were on the order of 10-100 ms, see Supplementary Information).

462

463 **3.4 Experimental conditions**

464 Experiments were conducted at three flow conditions, $Re_p = 500, 2500, 6500$, where the
465 particle Reynolds number is defined as $Re_p = \frac{\rho d_e U_{\text{int}}}{\mu}$ [68] and $U_{\text{int}} = U_0/\epsilon$ is the interstitial velocity
466 computed by dividing the superficial (open-tube) velocity, U_0 , by the bed voidage, ϵ . These flow
467 conditions were chosen as they provide a wide range of turbulent flow conditions to study; $Re_p \sim 500$
468 is often considered the lower boundary of the turbulent regime in packed beds [69].

469 At each condition studied, 3D images of both the time-averaged velocity (components measured
470 separately in all three orthogonal directions) and TKE (constituent mean square fluctuating components
471 in all three orthogonal directions measured separately) were acquired. The experimental process
472 conditions used at each of the three flow conditions are reported in Table 2. All experiments were
473 conducted at 25 ± 5 °C. The viscosity of SF₆ at the process conditions used was taken as 1.5×10^{-5}
474 Pa s [70] whilst density data at the respective temperatures and pressure were taken from Guder &
475 Wagner [71]. The voidage of the bed was determined gravimetrically by measuring the mass of catalyst
476 loaded into the bed (of known volume). To determine the voidage gravimetrically, the effective density
477 of the pellets was required, which was derived from water absorption measurements provided by Shell
478 Global Solutions (not shown).

479

480 **4 Results & Discussion**

481 **4.1 Bed structure**

482 To obtain a 3D image of the bed structure, a 3D image of the signal magnitude obtained under
483 zero-flow conditions was segmented using Otsu's method [72]. The resulting bed structural image is
484 shown in Figure 4a. The external bed voidage, ϵ (ratio of the volume of inter-pellet space to total tube
485 volume), computed from the structural MR imaging results was 0.53 ± 0.05 as reported in Table 2,
486 where the uncertainty of ± 0.05 represents the uncertainty in the voidage caused by uncertainty in the
487 image segmentation (which was determined by successively segmenting the MR image with a range of
488 threshold parameters, and taking the resulting range of voidage values as the uncertainty in the
489 voidage). Although the MR imaging derived values carry an uncertainty due to the partial voxel effect
490 and the image segmentation process, this value is in good agreement (absolute voidage discrepancy of
491 0.04, relative percent discrepancy of 8%) with the gravimetrically measured bed voidage, $\epsilon = 0.49$,
492 reported in Table 2. Therefore, the MR image of bed structure can be used with some confidence to

493 analyse the bed structure and pellet ordering effects. The 3D structural image of the bed was averaged
494 in the axial and azimuthal directions to generate a radial profile of average voidage through the bed,
495 shown in Figure 4b. Further, to analyse the pellet orientation, individual pellets identified within the
496 structural image of the bed were segmented manually by identifying the hole in each of the shaped
497 pellets. The angle, θ , of each pellet with respect to the bed axis ($\theta = 0$ for vertically aligned pellets)
498 was computed from the segmented pellets, and the distribution of pellet orientation is shown in Figure
499 4c, alongside the distribution measured by Caulkin et al. [73] using $\mu - CT$ for smooth hollow
500 cylindrical pellets in a narrow packed bed of similar aspect ratio ($N = 6.1$), and the theoretical random
501 orientation distribution, $p(\theta) \propto \sin \theta$ [74]. Note that the position and orientation of each pellet within
502 the imaging section is tabulated in the Supplementary Information.

503

504 The radial profile of voidage in Figure 4b shows distinctive oscillations, a characteristic feature
505 of narrow packed beds. Note that the radial voidage profile carries an absolute uncertainty of ± 0.05
506 (approximately independent of radial position) based on the uncertainty from the segmentation of the
507 MR image data. The voidage shows a maximum close to the wall, at a distance of $\sim 0.1d_o$ from the
508 wall, before displaying a first minimum ($\sim 0.9d_o$), a second maximum ($\sim 1.2d_o$), and a second
509 minimum ($\sim 1.8d_o$). Further, the voidage profile exhibits a shoulder before the first minimum at
510 $\sim 0.5d_o$, indicative of the pellet hole. Due to the ordering effect of the wall in narrow packed beds, the
511 radial profile of voidage is well-known to show characteristic oscillations [75–77]. However, most of
512 these studies have focused on spheres or cylinders, and few have been conducted using the non-
513 spherical packings commonly employed in real processes. The voidage profile acquired here agrees
514 qualitatively with the study of Moghaddam *et al.* [44] who used DEM to simulate bed loading of
515 similarly shaped Raschig rings. Specifically, the first minimum in voidage was found at slightly less

516 than $1d_o$,ⁱ and a shoulder in the voidage profile due to the contribution of the pellet holes was observed
517 at approximately $0.5d_o$, consistent with the present observations. This is in contrast to spherical
518 packings, where the first minimum in voidage occurs around $0.6d_o$ and no such shoulder is observed
519 [75].

520

521 Qualitatively, the orientation distribution measured in this work using MR imaging appears
522 similar both to that measured by Caulkin *et al.* [73] and to the theoretical random distribution (Figure
523 4c). The limited number of pellets in the MR imaging analysis region ($n = 58$ pellets) makes it
524 challenging to draw quantitative conclusions from the orientation histogram, in which some bins only
525 contain 1-2 pellets. A Kolmogorov-Smirnov statistical test comparing the measured orientation
526 distribution with the theoretical random distribution did not yield a statistically significant difference
527 ($p > 0.05$). Note that a statistical test with the distribution measured by Caulkin *et al.* [73] was not
528 possible as the total number of pellets measured was not reported. The orientation of non-spherical
529 pellets within packed beds has been shown to significantly impact the pressure drop [74]. In previous
530 works, the pellet orientation distribution has been found to be affected by the tube to particle diameter
531 ratio, loading method, and inter-pellet friction [73,74,78], thus making a quantitative comparison with
532 previous works difficult. Niegodajew *et al.* [74] found the orientation distribution to deviate
533 significantly from random for narrow packed beds ($N < 15$), due to wall ordering effects, with
534 multimodal distributions observed for $N < 12$. However, Niegodajew used smooth polycarbonate
535 pellets, which have significantly different frictional properties than the porous α -alumina pellets used
536 here. Indeed, Jurtz *et al.* [78] have shown that the static friction between pellets can have a significant

ⁱ Note that Moghaddam *et al.* [44] use the sphere diameter corresponding to the same pellet volume, $d_{p,v}$, as opposed to the pellet outer diameter, d_o , to normalize the distance. However, for the pellets used by Moghaddam *et al.* $d_{p,v} \approx d_o$, so the results are simply compared here on a d_o basis without any need for conversion.

537 effect on bed voidage and pellet orientation distribution. The orientation distribution measured by
538 Caulkin *et al.* [73] for plastic hollow cylinders ($N = 6.1$) using $\mu - CT$ is qualitatively similar to the
539 distributions measured in this work (Figure 4c). It is important to note that the pellet aspect ratio is
540 approximately 1 ($\frac{L}{d_o} \approx 1$) in both the present study and that of Caulkin *et al* [73]. For different pellet
541 aspect ratios, the resulting pellet orientation distribution may differ. The results reported here provide a
542 useful experimental dataset for validating future numerical bed packing simulations. Further, these
543 results demonstrate the ability of MR imaging to serve as a tool to measure the radial voidage profile
544 and pellet orientation distribution for complex pellet shapes. The ability to use the pellets of
545 commercial interest is especially important given that the frictional properties of pellets has previously
546 been shown to impact both the bed voidage and orientation distribution [78].

547

548 **4.2 Time-averaged velocity**

549 Images of the normalized time-averaged axial velocity at each flow condition are shown in
550 Figure 5. Immediately apparent from the images is the heterogeneity of the flow field. Regions of fast
551 flowing fluid are observed near the wall and can also be found in the interior of the bed where large
552 voids exist. Also apparent from Figure 5 is the flow heterogeneity introduced by the pellet shape,
553 specifically the holes. High axial velocity is observed in some pellets parallel to the bed axis (top left
554 and bottom left in images shown in Figure 5a), while regions of near-zero axial velocity are seen in
555 pellets oriented perpendicular to the flow direction (Figure 5b).

556

557 The images of normalized velocity in Figure 5 exhibit a high degree of self-similarity across the
558 range of Re_p studied. Regions of high and low velocities can be found in approximately the same
559 position and many flow features appear visually similar. The self-similarity is especially strong when

560 comparing the results at $Re_p = 2500$ and 6500 , where the flow structures appear especially similar for
561 the different flow rates. This agrees well with previously reported experimental results for spheres
562 [16,19], where turbulent flows have been shown to be self-similar at $Re_p > 1300$. The results here
563 suggest that the self-similarity of turbulent flows applies not only to model porous media (spherical
564 pellets), but also hollow cylindrical porous pellets.

565
566 The distribution of time-averaged axial velocity, \bar{u}_z , transverse velocity, \bar{u}_y , and speed (velocity
567 magnitude), $|\bar{\mathbf{u}}|$, are shown as histograms in Figure 6. All quantities have been normalized by the mean
568 interstitial velocity, U_{int} , to facilitate comparison between different flow conditions. The normalized
569 axial and transverse velocity distribution at all flow conditions appears remarkably similar. The
570 distribution of normalized fluid speed, $|\bar{\mathbf{u}}|/U_{\text{int}}$, changes slightly as Re_p increases, with the peak
571 shifting to lower speeds (Figure 6c). This shift may be attributed to the pellet holes in this system. As
572 Re_p increases, inertia becomes more important and the holes become less accessible due to the
573 acceleration/deceleration needed for flow to enter a hole, due to the random orientation of the pellets
574 (Figure 4). The decreased relative flow through the holes makes the average flow in the bed less
575 tortuous, and thus the average normalized speed shifts downwards as Re_p increases. However, taken
576 together, the overall distributions in Figure 6 show a high degree of similarity at different Re_p . This
577 further supports the finding of self-similarity for turbulent flows (at sufficiently high Re_p) in packed
578 beds as also demonstrated in Figure 5.

579
580 Regions of the bed with zero, or negative, axial velocity, $\bar{u}_z/U_{\text{int}} \leq 0$, as well as regions of fast-
581 moving fluid, $|\bar{\mathbf{u}}|/U_{\text{int}} > 2.5$, are identified in Figure 7. The corresponding fractions of (inter-pellet)
582 fluid in the bed are reported in Table 3. From Figure 7 it is clear that regions of backflow ($\bar{u}_z/U_{\text{int}} \leq 0$)

583 are dispersed throughout the bed, whilst regions of fast fluid ($|\bar{\mathbf{u}}|/U_{\text{int}} > 2.5$) are primarily located near
584 the walls. The regions of backflow appear to be located in the wake of pellets, in throats between
585 pellets, and in the interior of some pellet holes. Both the proportion of backflow and high-speed fluid
586 are approximately constant with respect to Re_p (Table 3).

587

588 A number of previous experimental studies have reported backflow zones in both laminar
589 [27,28,79,80] and turbulent [18,19] flows in beds of spherical packings. Using PIV, Khayamyan *et al.*
590 [19] experimentally found the backflow fraction for spherical pellets to peak at $Re_p \approx 400$, beyond
591 which a very modest decrease in backflow was observed, which was attributed to the destruction of
592 recirculating zones by the increased inertia in the bed. At $Re_p > 1000$, Khayamyan *et al.* [19] found
593 the backflow fraction plateaued at approximately 10%, in good agreement with the results presented
594 here for hollow cylinders. Moghaddam *et al.* [44,81] simulated the flow through a bed of Raschig rings
595 (hollow cylinders with holes larger than those used here), and observed a backflow fraction of $\sim 20\%$ at
596 $Re_p = 790$,ⁱⁱ significantly larger than both that found here and those reported by Khayamyan *et al.* [19]
597 for spheres. Given that Moghaddam *et al.* also reported a large backflow fraction for spheres and solid
598 cylinders of $\sim 17\%$, the discrepancy between the backflow fraction measured here and the simulations
599 of Moghaddam *et al.* [44,81] is unlikely due simply to differences in pellet shape. The data presented
600 here suggest that further development of CFD models might be required if accurate prediction of the
601 backflow fraction is required in beds of non-spherical pellets. The regions of high-speed fluid occur
602 primarily at the wall, but pockets can also be found toward the centre of the bed, in general agreement

ⁱⁱ Note that Moghaddam *et al.* [44] defined the pellet Reynolds number as $Re_p^* = \frac{\rho d_p v U_S}{\mu}$, where U_S is the superficial velocity. To facilitate comparison with this work, the Reynolds number used by Moghaddam *et al.* $Re_p^* = 1000$ has been converted to the equivalent particle Reynolds number based on the definition used here, $Re_p = \frac{\rho d_e U_{\text{int}}}{\mu} = 790$.

603 with the CFD results from Moghaddam *et al.* [81]. The distribution of these regions of high-speed fluid
604 will inevitably be important for transport properties in the bed.

605
606 To further investigate the near-wall hydrodynamics and the radial trends in flow, the radial
607 profile of time-averaged axial velocity $\bar{u}_z(r)$ at $Re_p = 2500$, obtained by azimuthally and axially
608 averaging the 3D MR velocity imaging data, is shown in Figure 8a, superimposed on the radial voidage
609 profile $\epsilon(r)$. Here, r is defined as the distance from the central axis of the bed, ranging from $r = 0$ at
610 the centre of the bed, to $r = R_t$ at the tube wall, where R_t is the tube radius. The trend displayed in the
611 radial profile of axial velocity is seen to follow the voidage profile. Further, as shown in Figure 8a, the
612 range of velocity values found at a given radial position reveals that there are substantial local
613 deviations in flow of up to $\sim 2U_{\text{int}}$ compared to the bed-average axial velocity at a given position.
614 Interestingly, Figure 8a also shows that the highest values of axial velocity are observed to occur near
615 the wall and at the local maxima in voidage (which is also visually supported by Figure 7b). However,
616 the backflow (negative axial velocities) is distributed approximately evenly across the bed, independent
617 of radial position (also supported visually by Figure 7a). Both the backflow, and the regions of high
618 velocity, are not captured by the average radial profiles. The radial profiles of time-averaged axial
619 velocity $\bar{u}_z(r)$ at all flow conditions studied are shown in Figure 8b. For clarity, only the average
620 profile is shown, the 95% ranges at all flow conditions were found to display similar behaviour (not
621 shown) to those shown in Figure 8a at $Re_p = 2500$. From Figure 8b, it can be seen that, with the
622 exception of the near-wall region, the radial profiles of axial velocity are remarkably similar at all Re_p .
623 The velocity of the voxels adjacent to the wall increases from $1.2U_{\text{int}}$ at $Re_p = 500$ to $1.5U_{\text{int}}$ at
624 $Re_p = 6500$.

625

626 Overall, the observations from Figure 8a indicate that azimuthally and axially averaging the 3D
627 flow data is insufficient to capture the backflow and high-speed flow, and more generally the local flow
628 heterogeneity inherent in narrow packed beds of hollow cylindrical pellets. This conclusion was
629 similarly drawn from the CFD simulations of Moghaddam *et al.* [44,81] for spheres, cylinders, and
630 Raschig rings. This finding may help to explain why continuum and pseudo-continuum models are
631 insufficient for modelling narrow packed beds, and supports the need for particle resolved CFD to
632 accurately capture the hydrodynamics in these complex systems. Although the turbulent flows in the
633 packed bed are largely self-similar across different Re_p (Figure 5 and Figure 6) the changes near the
634 wall (Figure 8b) highlight the importance of matching the flow conditions used in studies of packed
635 beds with the process conditions of interest commercially in order to obtain representative
636 hydrodynamics at all scales in the bed.

637

638 **4.3 Turbulent kinetic energy**

639 *4.3.1 Turbulence at $Re_p = 2500$: measurement and uncertainty analysis*

640 An image of the turbulent kinetic energy in the bed at $Re_p = 2500$, is shown in Figure 9a. Note
641 that an example of the underlying attenuation images, and the images of Reynolds stresses used to
642 calculate the TKE are reported in the Supplementary Information. The associated uncertainty in the
643 TKE, dk , calculated from the uncertainty in each component of mean square fluctuating velocity
644 obtained using Equation (21), is shown in Figure 9b. Finally, the % uncertainty in the measurement of
645 k in each voxel, $dk/k \times 100$, is shown in Figure 9c. The image of TKE is highly heterogeneous. Small
646 clusters of high intensity turbulence are scattered throughout the bed, and larger clusters of low
647 intensity turbulence are found in the bed, both inside and outside pellet holes. The uncertainty in the
648 TKE image, dk , is seen to be approximately $dk/U_{int}^2 = 0.1 - 0.4$ throughout the image. The

649 uncertainty is slightly larger near solid surfaces, due to the high shear, but otherwise is relatively
650 homogenous. As a result, the relative % uncertainty for regions of low turbulence is large (Figure 9c).
651 The average relative uncertainty in a voxel decreases to $\sim 40\%$ at $k/U_{\text{int}}^2 = 0.5$ and decreases further to
652 $<20\%$ for $k/U_{\text{int}}^2 > 1$. The absolute uncertainty in k is sufficiently small to have some confidence
653 when interpreting the images of k . Uncertainties of these magnitudes are broadly consistent with the
654 uncertainty reported by Elkins *et al.*[51] for MR measurements of the mean square fluctuating velocity
655 (Reynolds stress) for flow in a channel. In the future, the more detailed multi-point non-Cartesian
656 scheme for turbulence encoding recently reported by Schmidt *et al.* [54] may be utilized to both reduce
657 the measurement error, and measure all components of the Reynolds stress tensor. The simple two-
658 point gradient encoding used in this work was selected for its simplicity and speed.

659

660 4.3.2 Turbulent kinetic energy as a function of Re_p

661 Images of the normalized TKE, k/U_{int}^2 , at each flow condition studied in this work are shown in
662 Figure 10. The distributions of normalized TKE, k/U_{int}^2 , at each flow condition are shown in Figure
663 11. In contrast to the time-averaged velocity, the TKE does not appear self-similar as Re_p increases.
664 The structure, and position, of regions of high turbulence appear to change as Re_p increases, consistent
665 with pore-scale PIV findings reported by Patil & Liburdy [16] for spherical pellets. The distribution
666 changes shape as Re_p increases, from a bimodal distribution with peaks near $k/U_{\text{int}}^2 = 0$ and $k/U_{\text{int}}^2 =$
667 0.4 with a long tail at $Re_p = 500$, to a unimodal distribution with a peak near $k/U_{\text{int}}^2 = 0.7$ and a
668 shorter tail at $Re_p = 6500$. The average value of normalized TKE decreases slightly with increasing
669 Re_p ($k/U_{\text{int}}^2 = 0.76 \pm 0.11$; $k/U_{\text{int}}^2 = 0.71 \pm 0.04$; $k/U_{\text{int}}^2 = 0.60 \pm 0.04$ for $Re_p = 500, 2500, 6500$,
670 respectively).

671

672 PIV measurements from Patil & Liburdy have also been reported to show a similar decrease in
673 the average normalized turbulent velocity fluctuations with increasing Re_p in beds of spheres [16]. The
674 change of shape in the normalized TKE distributions shown in Figure 11 indicates that the TKE
675 becomes more homogeneous throughout the bed at high Re_p . Further, whilst some care is required in
676 interpreting low values of TKE due to the large uncertainty, the peak near $k/U_{int}^2 = 0$ at $Re_p = 500$
677 potentially indicates some regions of the bed that locally are not yet fully turbulent, which is possible
678 given that $Re_p = 500$ is often considered the lower limit of the fully turbulent regime in packed beds
679 [69]. The shapes of the normalized TKE distributions in Figure 11 appear qualitatively similar to PIV
680 measurements of spherical packings reported by Thaker *et al.* [18] who observed a similar change in
681 shape in the TKE distribution from $Re_p \approx 1100$ to $Re_p \approx 6600$. Notably, in the work of Thaker *et al.*,
682 the TKE predicted by the SST $k - \omega$ model was found to underestimate the experimental results by
683 approximately two orders of magnitude, demonstrating the importance of validating turbulence
684 modelling with experimental measurements of k .

685

686 Taken together, the measurements of TKE reported in this work reveal that the local turbulence
687 properties in the bed do not simply scale linearly with flow and are a complex function of the local flow
688 dynamics. However, qualitatively, the turbulence in the bed measured here shows similar
689 characteristics to that previously reported in beds of spherical pellets. Further investigation is required
690 to determine if this finding generalizes to other non-spherical, commercially-relevant pellet shapes.

691

692 4.3.3 Comparing turbulent kinetic energy with time-averaged velocity

693 To further investigate the turbulence in the bed, the TKE measurements were compared with
694 the time-averaged velocity measurements. Figure 12 shows a 3D visualization of the TKE and time-

695 averaged velocity at $Re_p = 2500$. Visually, the regions of high TKE appear close to locations where
696 the flow is impinging on pellets, and at some local necks between pellets (constrictions) in the bed.

697

698 To investigate any correlation between TKE and time-averaged velocity, Figure 13 shows a 2D
699 histogram of the normalized TKE, k/U_{int}^2 , and the normalized time-averaged velocity magnitude,
700 $|\bar{\mathbf{u}}|/U_{int}$. At all Re_p , it is seen that the low values of normalized TKE ($k/U_{int}^2 < 0.25$) occur with
701 highest probability in regions with low velocity magnitude. However, the highest values of TKE,
702 $k/U_{int}^2 > 2$ are most likely at intermediate velocity magnitudes ($1 < |\bar{\mathbf{u}}|/U_{int} < 2.5$). The 2D
703 distributions change shape slightly from $Re_p = 500$ to $Re_p = 2500$ and 6500 , with the highest values
704 of TKE occurring at slightly higher values of $|\bar{\mathbf{u}}|/U_{int}$ as Re_p increases.

705

706 It is interesting that the highest values of TKE occur at locations with intermediate, as opposed
707 to the highest or lowest, time-averaged velocity magnitude. Visually, this finding agrees with direct
708 numerical simulation (DNS) results of He *et al.* [82] for flow through a unit cell of face-centred cubic
709 packing, whose results visually appear to show the highest values of TKE occurring in regions with
710 intermediate time-averaged velocity magnitude. The 2D distributions in Figure 13 give a statistical
711 fingerprint of the pellet-scale turbulent hydrodynamics in a packed bed. This characterization could
712 prove useful in the future for validating the turbulence closure models used in porous media flow
713 simulations.

714 **4.4 Velocity and TKE trends at the pellet-scale**

715 To investigate the average flow field at the pellet-scale in this system, the time-averaged velocity and
716 TKE were analysed as a function of perpendicular distance from solid boundary (either wall or pellet).
717 Figure 14 shows the normalized time-averaged velocity magnitude and TKE plotted as a function of

718 the perpendicular distance from each voxel to the nearest solid boundary. These plots thus represent
719 average velocity and TKE behaviour at the pellet-scale within the bed. It is important to note that these
720 average profiles do not give the true velocity or TKE profile within a single void in the bed. Instead, the
721 average is taken over the entire bed, where a wide range of void geometries and sizes are present in the
722 3D void space. As expected, from Figure 14a, it is seen that the time-averaged velocity magnitude
723 increases as the distance from solid increases. The normalized velocity profiles collapse onto the same
724 curve for all Re_p studied, with the exception of the point closest to the solid. The normalized TKE
725 profiles shown in Figure 14b, show substantially different behaviour that changes with Re_p . At $Re_p =$
726 500, the normalized TKE decreases with increasing distance from the solid. At $Re_p = 2500$, the
727 normalized TKE profile shows a more gradual decrease with increasing distance from the solid.
728 Finally, at $Re_p = 6500$, the normalized TKE appears approximately constant with respect to distance
729 from the solid.

730

731 The time-averaged velocity profiles (Figure 14a) vaguely resemble the flow profile within a
732 channel/duct for turbulent flow, with an increase in velocity as distance from the solid increases. The
733 higher velocity for voxels closest to solid at $Re_p = 500$ is perhaps surprising. Again, these must not be
734 interpreted as the true velocity profile within a single pore/void in the bed. The decrease in velocity
735 closest to solid from $Re_p = 500$ to $Re_p = 2500, 6500$ is most likely due to the effect of narrow
736 constrictions (e.g. necks between pellets). As Re_p increases, the normalized velocity in narrow
737 constrictions in the bed (and thus many voxels adjacent to solid boundary) may decrease due to the
738 increasingly large resistivity to flow these regions present as compared to larger voids in the bed. With
739 the exception of the region closest to solid boundary, the velocity profiles in Figure 14a are self-
740 similar, further confirming the self-similarity of the velocity field at the pellet-scale.

741
742 In contrast to the time-averaged velocity, the TKE profiles (Figure 14b) are not self-similar with Re_p ,
743 providing further evidence that the TKE is not self-similar at the pellet scale (also shown in Figure 10).
744 Qualitatively, the TKE profile appears similar to that expected for channel/duct flow at $Re_p = 500$,
745 with a peak near the wall and a decrease with increasing distance from solid boundary. However this
746 decrease towards the centre of voids is much less than what one would expect in the case of channel
747 flow. As Re_p increases, the peak in TKE moves away from the solid boundary and the decrease is
748 more gradual, and at $Re_p = 6500$ the TKE is independent of the distance from solid. This is markedly
749 different from the case of channel flow, where the TKE reaches a maximum near the wall, and
750 decreases rapidly towards the centre of the channel. The simulations conducted by He *et al.* [82]
751 visually appear to show the normalized TKE in a pore becoming more uniform as Re_p increases (see
752 Fig. 2 in He *et al.* [82]), and qualitatively appear to reflect the trends shown in Figure 14b. Overall, the
753 TKE profiles indicate that the TKE at the pellet-scale in this packed bed is substantially different from
754 the case of simple channel flow. A final point to note is that in principle, the MR methods utilized here
755 may be applied to study any gas from which an MR signal during flow can be acquired (including
756 hydrogen, methane, ethane, propane, butane, hyperpolarized ^{129}Xe) [83–85]. However, the precise
757 pulse sequence and parameters employed will require modification subject to relaxation and diffusion
758 considerations.

759

760 **5 Conclusions**

761 A compressed sensing MR velocity imaging methodology has been implemented to acquire 3D
762 images of turbulent flows in a packed bed of porous, hollow cylindrical α -Al₂O₃ pellets over a range
763 of flow conditions $Re_p = 500 - 6500$. By imaging the flow in 3D, it is possible to assess the extent to
764 which the hydrodynamics in a commercially-relevant system differ from those in beds of smooth non-
765 porous spherical pellets, a system commonly taken to serve as a model packed bed. The 3D images of
766 the bed structure and turbulent flow reveal a number of interesting features.

767 The measured pellet orientation distribution was found to be qualitatively consistent with
768 previous x-ray tomography measurements of similarly shaped packings [73]. The radial profile of
769 voidage is different from that previously reported for spheres, showing a shoulder at a distance of
770 $\sim 0.5d_o$ from the tube wall which is indicative of the pellet hole and consistent with computational
771 findings for a similar pellet shape. The time-averaged velocity in the bed shows significant flow
772 heterogeneity, with regions of backflow distributed through the bed, and regions of high-speed fluid
773 primarily close to the wall and at regions of high voidage. The amount of backflow in the bed was
774 found to be approximately 10% of the total inter-pellet fluid in the bed, independent of Re_p , and
775 significantly lower than that predicted by previous CFD simulations for spherical, cylindrical, and
776 hollow cylindrical pellets [44]. The velocity images and distributions at different Re_p appear
777 remarkably similar when normalized, demonstrating that the turbulent flows in packed beds of
778 commercially-relevant pellets are self-similar, as has been previously reported for model packed bed
779 systems. The radial profile of axial velocity reveals significant flow heterogeneity, and demonstrates
780 the inadequacy of axial and azimuthal averaging in capturing the flow heterogeneity present within
781 narrow packed beds, in agreement with recent simulations [44,81]. Further, the radial profiles show the
782 near-wall flow behaviour changes with Re_p , with the velocity near the wall increasing as Re_p

783 increases. 3D images of the turbulent kinetic energy reveal that the local turbulence does not scale
784 linearly with the flow. The TKE distribution becomes more homogenous as Re_p increases from 500 to
785 6500, indicating the turbulence in the bed is more uniform at higher Re_p . The distributions of TKE
786 agree qualitatively with previous experimental measurements [18] made in a bed of spheres, suggesting
787 that the TKE in beds of hollow cylinders is similar to model packed bed systems. Interestingly, the
788 highest values of TKE in the bed are associated with fluid having an intermediate value of time-
789 averaged velocity, as opposed to the fastest or slowest fluid in the bed. The TKE at the pellet-scale
790 shows markedly different behaviour from simple channel flow, with the TKE becoming approximately
791 independent of distance from solid interface at $Re_p = 6500$.

792

793 Application of 3D MR imaging techniques to measure the structure and flow in a commercially
794 relevant packed bed reveals a number of insights not accessible by studying model spherical pellet
795 systems, specifically the pellet orientation distribution, the radial voidage profile, the decreased
796 backflow compared to previous computational studies and the near-wall flow behaviour. Further, the
797 MR imaging measurements provide a full-field dataset that can be used to validate and calibrate future
798 numerical flow simulations. The direct study of hydrodynamics in commercially-relevant packed beds,
799 as realized in this work, is an important step forward for understanding fundamentally how pellet shape
800 impacts reactor performance, and ultimately in advancing towards the rational design of the catalyst
801 pellets and process conditions used commercially.

802 **6 Acknowledgements**

803 The authors thank Shell Global Solutions International B.V. for funding this work, provision of the
804 catalyst pellets used in this study, and providing water absorption measurements for determining pellet
805 material density. SVE further thanks the Sir Winston Churchill Society of Edmonton and the Natural
806 Sciences and Engineering Research Council of Canada (NSERC) for additional funding. For the

807 purpose of open access, the authors have applied a Creative Commons Attribution (CC BY) licence to
808 any Author Accepted Manuscript version arising from this submission.

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1054 8 Figure Captions

1055
1056 **Figure 1.** Sources of phase dispersion, $\langle \phi^2 \rangle$, within a voxel that contribute to signal attenuation.

1057 **Figure 2.** Schematic diagram of closed-loop SF₆ flow rig used for MR velocity imaging in a packed
1058 bed. (a) flow loop. 1 – low pressure vessel; 2 – compressor; 3 – high pressure vessel; 4 – needle valve;
1059 5 – regulator valve; 6 – perforated plate distributor; 7 – catalyst support pellets; 8 – ABS pipe, $d_{\text{tube}} =$
1060 41 mm ID; 9 – superconducting magnet, 4.7 T; 10 – mass flow controller; 11 – digital pressure gauge;
1061 12 – SF₆ gas cylinder. (b) packed bed. Red boxes denote the MR imaging region (note: drawings not to
1062 scale).

1063
1064 **Figure 3.** Schematic of SESPI pulse sequence used for all MR imaging experiments in this work.

1065
1066 **Figure 4.** Bed structure from segmentation of 3D intensity image under zero flow conditions. (a) 3D
1067 segmented image of bed structure (image rendered in Amira-Avizo); (b) radial voidage profile showing
1068 average voidage as a function of normalized distance from the wall; (c) histogram of pellet orientation
1069 with respect to vertical. Experimental distribution from MR imaging (this work) (–); distribution for
1070 smooth plastic hollow cylinders (termed Hama beads in [73]) in a narrow packed bed ($N = 6.1$) from
1071 Caulkin et al. [73] obtained using $\mu - \text{CT}$ (–); theoretical random distribution $p \propto \sin(\theta)$ (–). Note
1072 that the vertical direction in (a) represents the z-axis.

1073
1074 **Figure 5.** Central slice of 3D time-averaged axial velocity images at each flow condition. (a) xy
1075 images. Images shown have a FOV of 45 mm (x) \times 45 mm (y) and an in-plane spatial resolution of
1076 0.35 mm (x) \times 0.35 mm (y). Direction of net flow is into the page. (b) xz images. Images shown have a
1077 FOV of 45 mm (x) \times 45 mm (z) and an in-plane spatial resolution of 0.35 mm (x) \times 0.5 mm (z).
1078 Direction of net flow is in the +z direction.

1079
1080 **Figure 6.** Distribution of (a) time-averaged normalized axial velocity, \bar{u}_z/U_{int} , (b) time-averaged
1081 normalized transverse (y) velocity, \bar{u}_y/U_{int} , and (c) time-averaged normalized speed, $|\bar{\mathbf{u}}|/U_{\text{int}}$. (–)
1082 $Re_p = 500$; (–) $Re_p = 2500$; (–) $Re_p = 6500$.

1083
1084 **Figure 7.** 3D visualization at $Re_p = 2500$ of (a) regions of the bed with zero or negative velocity
1085 (backflow) in the axial direction (all voxels with $\bar{u}_z/U_{\text{int}} \leq 0$ are visualized by blue shading); (b)
1086 regions of the bed with high-speed fluid, $|\bar{\mathbf{u}}|/U_{\text{int}} > 2.5$. Note that the vertical direction represents the
1087 z-axis.

1088

1089 **Figure 8.** Radial profile of voidage, $\epsilon(r)$, and normalized time-averaged axial velocity, $\bar{u}_z(r)/U_{\text{int}}$,
1090 as a function of distance from the tube wall, $R_t - r$, normalized by pellet outer diameter, d_o . (a)
1091 Velocity at $Re_p = 2500$ (—) plotted alongside voidage (—). Shaded regions represent 2.5 percentile to
1092 97.5 percentile of velocity values found at each respective radial position in the 3D flow images (thus
1093 the shaded region covers 95% of the velocity values found in the bed at a given position). (b)
1094 comparison of radial velocity profiles (◆) $Re_p = 500$; (●) $Re_p = 2500$; (■) $Re_p = 6500$.

1095

1096 **Figure 9.** Turbulent kinetic energy image and uncertainty analysis at $Re_p = 2500$. (a) Central slice of
1097 3D image of TKE, k , and (b) uncertainty in TKE, dk . Both quantities have been normalized by the
1098 square of the mean interstitial velocity, U_{int}^2 . Images shown have a FOV of 45 mm (x) \times 45 mm (y)
1099 and an in-plane spatial resolution of 0.35 mm (x) \times 0.35 mm (y). Direction of net flow is into the page.
1100 (c) % uncertainty in TKE measured from each voxel in the 3D dataset, calculated as % uncertainty =
1101 $\frac{dk}{k} \times 100$, plotted as a function of normalized TKE. (●) uncertainty in each voxel, (—) average
1102 uncertainty at a given value of k/U_{int}^2 .

1103

1104 **Figure 10.** Central slice of 3D image of normalized turbulent kinetic energy, k/U_{int}^2 , at each flow
1105 condition. (a) xy images. Images shown have a FOV of 45 mm (x) \times 45 mm (y) and an in-plane
1106 spatial resolution of 0.35 mm (x) \times 0.35 mm (y). Direction of net flow is into the page. (b) xz images.
1107 Images shown have a FOV of 45 mm (x) \times 45 mm (z) and an in-plane spatial resolution of 0.35 mm
1108 (x) \times 0.5 mm (z). Direction of net flow is in the +z direction.

1109

1110 **Figure 11.** Distribution of normalized turbulent kinetic energy, k/U_{int}^2 , in the bed at: $Re_p = 500$ (—);
1111 $Re_p = 2500$ (—); $Re_p = 6500$ (—).

1112

1113 **Figure 12.** 3D visualization of (a) normalized turbulent kinetic energy, k/U_{int}^2 , and (b) normalized
1114 time-averaged velocity magnitude, $|\bar{\mathbf{u}}|/U_{\text{int}}$ at $Re_p = 2500$. Note that the vertical direction represents
1115 the z-axis.

1116

1117 **Figure 13.** 2D histogram of normalized turbulent kinetic energy, k/U_{int}^2 , and time-averaged velocity
1118 magnitude, $|\bar{\mathbf{u}}|/U_{\text{int}}$, at (a) $Re_p = 500$; (b) $Re_p = 2500$; (c) $Re_p = 6500$.

1119

1120 **Figure 14.** Average (a) normalized time-averaged velocity magnitude, $|\bar{\mathbf{u}}|/U_{\text{int}}$, (b) normalized TKE,
 1121 k/U_{int}^2 , plotted as a function of perpendicular distance from the nearest solid boundary l , normalized
 1122 by pellet outer diameter d_o . Error bars represent the uncertainty of the average value at each position.
 1123 $Re_p = 500$ (\blacklozenge); $Re_p = 2500$ (\bullet); $Re_p = 6500$ (\blacksquare).

1124 9 Tables

1125 Table 1. Average pellet properties.

pellet	hollow cylinder – large hole
material	α -alumina
pellet length, L [mm]	9.1
hole diameter, d_{hole} [mm]	3.2
mean OD, d_o [mm]	8.7
spherical equivalent diameter, d_e [mm]	6.3

1126

1127

1128 Table 2. Process conditions used.

Re_p , nominal [-]	500	2500	6500
P [bar(a)]	7	7.1	7.2
\dot{m} [g s^{-1}]	0.833	3.88	10.0
ρ [kg m^3]	45.2	45.9	46.7
U_0 [m s^{-1}]	0.014	0.064	0.162
ϵ , gravimetric [-]	0.49 ± 0.02		
ϵ , MRI [-]	0.53 ± 0.05		
Re_p [-]	544	2537	6533

1129

1130 Table 3. Fraction of fluid with zero or negative velocity in the axial direction (backflow), $\bar{u}_z/U_{\text{int}} \leq 0$,
 1131 and high-speed fluid, $|\bar{\mathbf{u}}|/U_{\text{int}} > 2.5$, in the bed at each flow condition.

Re_p [-]	backflow	high-speed fluid
	$f(\bar{u}_z/U_{\text{int}} \leq 0)$ [%]	$f(\bar{\mathbf{u}} /U_{\text{int}} > 2.5)$ [%]
500	12	9.1
2500	11	7.4
6500	12	8.3

1132