Real-time *in situ* measurement of particle size in flowing powders by terahertz time-domain spectroscopy

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Abstract—**We present the results of real-time reflection terahertz time-domain spectroscopy (THz-TDS) measurements of flowing powders. The data shows a clear relationship between the sizes of particles flowing through a tube to measured scattering losses at terahertz frequencies. The particle size of the powder covers the range of typical length-scales used in the pharmaceutical and food industry.**

I. INTRODUCTION

THE measurement of particle size during powder processing is important for process control in a range of manufacturing industries in the pharmaceutical, chemical and food sector, among others[1]. For example, in the pharmaceutical industry blending uniformity is critical to ensure both uniform distribution of active pharmaceutical ingredient in each dose[2,3], as well as mechanical integrity of the tablet post-process. The wavelength of terahertz pulses generated in typical THz-TDS systems covers the range from 3 mm to around 75 μm (0.1 to 4 THz). This range includes length scales of particle sizes in typical powder samples which are relevant in powder processing. Strong scattering losses are expected when probing powders with such particle sizes[4]. In addition, terahertz radiation can penetrate a number of polymer materials that are used for tubes and pipes to feed powders into processing equipments and transport it between unit operations. The short acquisition times that can be achieved in THz-TDS make the technique ideally suited to monitoring properties of flowing powders in real-time.

II. RESULTS

In our experiments, five different fractions of glass ballotini (spherical beads) with diameters between 90 and 212 μm were used as model particles. The flow apparatus consisted of a plastic tube (9 mm I.D.) which was held in vertical orientation and through which powders were allowed to flow freely into a collection vessel placed below the tube. The terahertz measurements were performed using a fiber-coupled THz-TDS reflection set-up (TeraView Ltd., Cambridge, UK). Reflected terahertz pulse profiles were continuously recorded (every 100 ms) for the duration of the powder flow. Recorded time-domain profiles were averaged to produce profiles shown on the left in Fig. 1; corresponding power spectra are shown on the right. A scanning delay line length of approximately 18 ps was used to probe to a depth of about 4 mm inside the flowing powder. The first peak encountered (from left to right) is a reflection from the inner tube wall. Remaining reflections originate from within the flowing powder and rapidly decrease in intensity with increasing distance from the tube wall. Losses due to scattering from flowing particles are reflected in the gaps observed in the spectra, compared to the reference spectrum (not shown). In all measurements, the second most intense peak occurs at a distance which is separated from the first reflection by a delay close to the diameter of spheres flowing at that time. This distance between the first two peaks increases with particle diameter indicating that this feature is due to reflections from the back surface of a layer of spheres lining the inner tube wall while the particles move through the field-of-view.

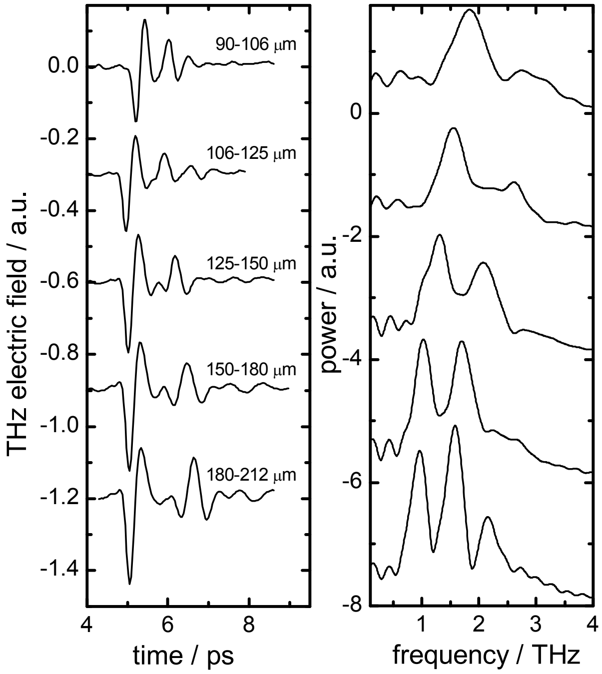

Fig. 1: Averaged terahertz time-domain waveforms (left) and corresponding power spectra (right) of glass ballotini of different sizes flowing through a plastic tube.

In order to improve the signal-to-noise ratio, the raw data of each run was averaged over the duration of the powder flow experiment. The reflected pulse was truncated to a total length of about 5 ps before performing the FFT (Fig. 2). A feature at 2.1 THz was observed in all spectra recorded during particle flow. This peak originates from an oscillatory pattern at longer time-delays in time-domain waveforms (Fig. 2, bottom trace). The nature of this feature is not quite clear and further work is required to elucidate its exact origin. However since it was observed for all particle sizes its presence does not yield further information on flowing particles and indeed obstructs the peaks that are dependent on particle size. It was thus removed from spectra by truncating time-domain waveforms at a distance of about 3 ps beyond the inner tube wall.

The diameters of ballotini along with the frequency of the peak spectral features observed in the recorded spectra and their corresponding length scales for these features are shown in Tab. 1. It is interesting to note that in all cases not only scattering losses at the length scale of the particle size used were observed but also reflections which seem to have passed through more than one particle.

Size fraction	Peak / THz	corresponding length scale / µm
90-106 µm	1.80, 2.76, 3.21_s	167, 109, 93
106-125 µm	1.55, 2.62	193, 114
125-150 µm	1.30, 2.07	231, 145
150-180 µm	1.03, 1.70	291, 176
180-212 µm	0.95, 1.58, 2.15	316, 190, 134

Tab. 1: Particle size fractions, the frequencies of peak spectral features observed for each and their corresponding length scales.

Fig. 2 shows the time- and frequency-domain data obtained from a measurement of the fraction containing 106-125 µm particles. Two broad peaks are observed in the power spectra. The intensities of these peaks exceed that of the power spectrum from the time-domain waveform reflected from the plastic-air interface at the inner wall of the plastic tube at those frequencies.

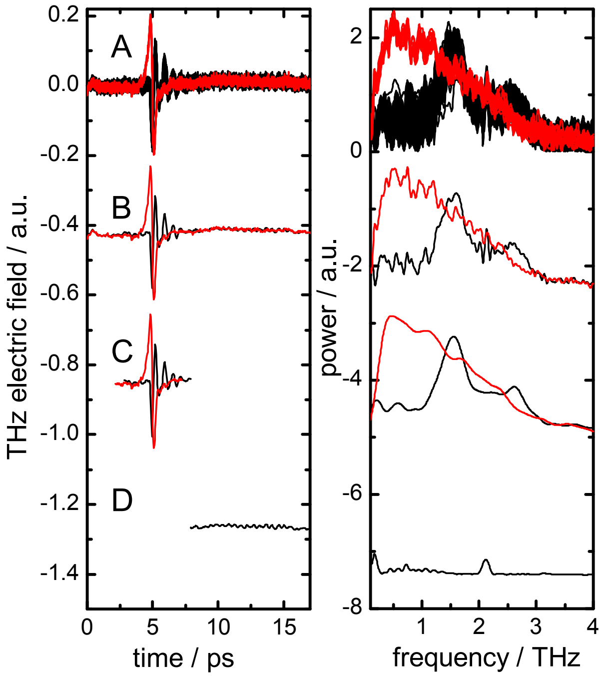

Fig. 2: Processing of the reflected terahertz data for the example of the 106-125 µm fraction. Shown are time-domain data on the left and corresponding power spectra on the right. A) Raw waveforms recorded during (black) and after particle flow (red). B) Averaged data; C) truncated time-domain data reveal two peaks in the power spectra; and D) time-domain signals at longer delay times produce a spectral feature at 2.1 THz common to measurements made at all particle sizes – as seen in the power spectra B).

The inner wall of the plastic tube with a refractive index of about 1.6 is by far not an ideal reflector hence the intensity of the power spectrum from this reflection is lower than that of the glass ballotini, which have a refractive index of about 2. Nonetheless, the air-plastic interface provides sufficient broadband reflections indicating the positions of relative losses due to scattering by flowing particles. The advantage of using the plastic tube reflection directly as the reference is that no additional reference mirror is required for a quantitative measurement of the particle sizes.

In Fig. 3 the time-domain waveforms during powder flow in real-time are presented. OOver the duration of the experiment the flow pattern indicates a very uniform flow behavior. The point where no powder passes through the tube any longer can be easily identified.

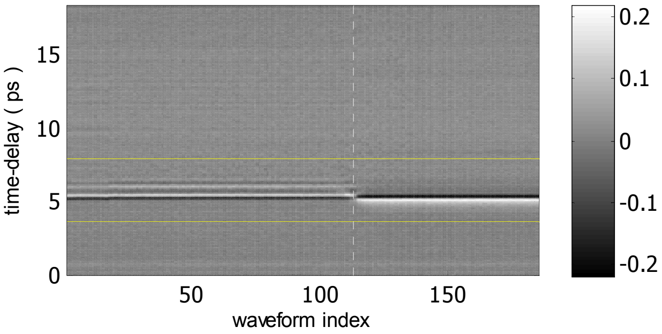

Fig. 3: Time-domain waveforms of the 90-106 µm fraction. The dotted white line indicates the point after which particles cease to flow. The yellow lines indicate the region of interest in the time-domain, for the Fourier transform to extract the power spectra. The colour bar represents the THz electric field in arbitrary units.

In addition, by being able to probe into the powder bed rather than only collecting data from the immediate interface at the tube wall itself, the terahertz data reveal that the powder in the centre of the tube moves faster than the powder towards the tube walls (Fig. 4).

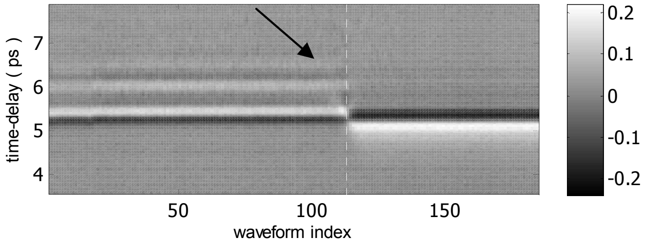

Fig. 4: Detail of cropped time-domain waveforms from Fig. 3 containing reflections from flowing particles (left) and from the plastic-air interface at the inner tube wall after particles have stopped flowing (right of vertical dashed line). The arrow highlights the region towards the emptying of the tube where the signal from deeper into the powder bed decays quicker compared to the signal on the tube walls.

In frequency domain the corresponding data of the flowing powder is represented in Fig. 5 and 6.

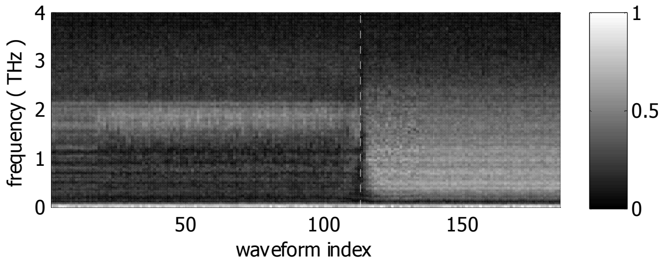

Fig. 5: Power spectra extracted from the full 18 pstime-domain waveforms (as shown in Fig. 3). Notice the difference between spectra before and after the 10th waveform, which is the point at which particles began to flow. The colour bar represents the power in arbitrary units.

In this particular powder flow experiment the tube was initially filled with the powder and data acquisition was started before the flow was initiated. While it is difficult to spot a difference between the stationary and flowing powder bed in the time-domain waveform (Fig. 3), the spectra show the difference quite pronounced (Fig. 5 and 6).

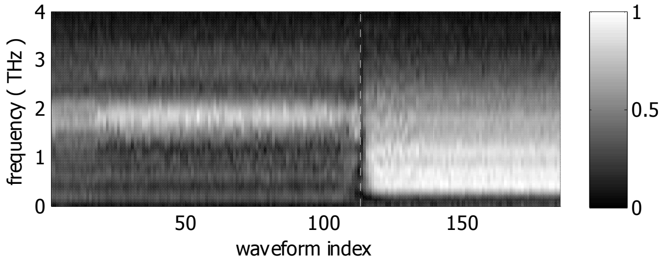

Fig. 6: Power spectra of cropped time-domain waveforms (Fig. 4).

THz-TDS was found to be highly sensitive to monitor particle size and powder flows in optically opaque tubing. The fast data acquisition together with the high penetrative power of terahertz radiation makes it possible to study the dynamics of powder flow in industrially relevant materials

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