Terahertz time domain spectroscopy and imaging: Emerging techniques for food process monitoring and quality control

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The Terahertz (THz) region of the electromagnetic spectrum, spanning the range between 100 GHz and 30 THz, has recently enjoyed a renaissance due to technological developments in source and detector components. With the development of THz instrumentation, applications of THz spectroscopy and imaging for quality control of food products have expanded in scope and improved in performance. This article gives an overview of the fundamentals of THz technology and a comprehensive review of applications of THz time domain spectroscopy and imaging for food quality and control. Technical challenges and future outlook for these emerging techniques are also discussed.

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Introduction

Once known as the Terahertz (THz) gap, the region of the electromagnetic spectrum from 100 GHz to 30 THz was largely ignored until the mid-1990s. This was due to the low efficiency of sources and detectors of THz energy which made it difficult to build efficient instrumentation in this wavelength range. The development of ultrafast lasers in the 1990s was a major turning point, leading to the development of modern day THz-time domain spectroscopy (Thz-TDS). THz systems can now be developed using a variety of sources, such as Gunn diodes (Chandra et al., 1997) and super-lattice electronic devices (Renk et al., 2007), which operate at the low frequency end of the THz band, or photonic sources, such as lasers and photoconductive dipole antennae (Tani et al., 2002), which are more widespread, and can generate radiation across a broadband frequency range. Other THz generation methods include optical rectification (Radhanpura et al., 2009) and mixing of light from two laser sources whose wavelength difference is in the THz region.

Terahertz systems can be designed to operate in transmission or reflection mode and in the time or spatial domain. Terahertz radiation has sufficient energy (1–10 meV) to promote rotations and vibrations of molecules and can thus be used for their identification; for example, Thz-TDS has been related to the function of biomolecules and conformations of proteins (Kim, Born et al., 2008; Kim, Jeon et al., 2008; Markels et al., 2002). It is non-ionising and sensitive to polar molecules such as water. At the same time, THz radiation interacts only very weakly with materials composed of non-polar molecules, such as plastics and ceramics and is reflected by most metals. This means that THz can 'see' through common packaging materials and textiles, making it well suited for security operations such as passenger screening at airport terminals. Many biological molecules have distinct THz signatures. He et al. (2006) demonstrated the potential of Thz-TDS in the range 0.1–2.5 THz for characterisation of animal tissue. Based on power absorption coefficients extracted from Thz-TDS spectra, they showed that skin, fat and lean tissues of the same animal exhibited different frequency dependent response, due to variations in water content, while similar tissues from different animals (rats and pigs in this case) showed similar responses. McAuley et al. (2006) demonstrated THz transmission imaging of bacon slices, and clear distinction between fatty and lean tissues was possible.

Spectroscopic methods such as NIR spectroscopy and chemical imaging are of major interest to industries such as pharmaceutical and food (Gowen *et al.*, 2007, 2008; Shen, 2011), as they facilitate rapid, non-destructive monitoring of inter and intra batch variability in processing methods. This is of major importance in quality control and allows manufacturers to benefit from the adoption of the process analytical technology (PAT) paradigm. The US Food and Drug Administration-led PAT initiative is transforming approaches to quality assurance in industry. Core to the PAT initiative is increased process understanding by monitoring of critical performance attributes, leading to better process control and ultimately improved product quality. Potential advantages of PAT implementation include: reduced production cycle times, prevention of rejects, reduction of human error and facilitation of continuous processing to improve efficiency. Compared with other vibrational spectroscopic methods, such as NIR and MIR spectroscopy, Teraherz spectroscopy has the distinct advantage that it can penetrate many organic materials (e.g. plastics, paper, skin). Commercially available Fourier transform infrared (FTIR) spectrometers can operate in the spectral range down to 1.5 Thz. However, the dynamic range of far infrared FTIR spectroscopy is limited due to source and detector efficiency. Unlike NIR chemical imaging, THz imaging enables direct measurement of sample thickness, refractive index and absorption coefficient. However, at the present time, commercial NIR imaging systems are at a more advanced state of development than THz systems. Examples of THz imaging applied to pharmaceutical quality control include tablet coating characterisation, evaluation of dissolution properties and characterisation of polymorphisms (Shen, 2011).

The number of reported applications of THz for food industry-related research has increased in tandem with those in the pharmaceutical industry. It therefore seems timely to provide a review of THz spectroscopy and imaging applications for food quality monitoring. This article is structured as follows: the next section gives an overview of instrumentation common in THz spectroscopy; this is followed by a short description of data analysis in THz. The main body of the article is found in Section 4, which compiles the reported applications of THz spectroscopy and imaging to food quality analysis (a summary is provided in Table 1). This is followed by a discussion of the challenges and limitations of this technology. Some brief conclusions end the article.

Instrumentation

Terahertz spectroscopy systems can be broadly divided into two categories based on their principle of operation: pulsed and continuous wave systems. A comparison of the advantages and disadvantages of each kind of system is given in Table 2. Pulsed systems rely on the generation of THz waves by ultrafast lasers which emit sub 100 fs pulses, focussed on photoconductive antennae (PCAs). There are typically two PCAs in a THz system: one emitter and one detector. An external electric field is applied to the emitter PCA in order to maintain an electrical bias. When excited by the fs pulse, the electrically biased emitter PCA emits a THz pulse (Fig. 1). The THz wave generated is typically re-focussed prior to passing through a sample using polymeric lenses or mirrors. After passing through a sample, the beam may be realigned, again by use of polymeric lenses or mirrors, and is directed to the detector PCA. Another portion of the optical pulse is used to gate the

Table 1. Summary of applications of THz in food quality monitoring.				
Product	**Author, year**	**Application**	**THz modality**	**THz range**
Wheat	Chua *et al.*, 2004, 2005	Moisture content	TDS	0.1–4
Wafers	Parasoglou *et al.*, 2009	Moisture content	TDS	0.1–2.4
Food powders (sticky rice, sweet potato, and lotus root)	Hua & Zhang, 2010	Pesticide detection	TDS	0.5–1.6
Animal feed	(Redo-Sanchez *et al.*, 2011)	Antibiotic detection	TDS	0.1–2
Milk powder	(Redo-Sanchez *et al.*, 2011)	Antibiotic detection	TDS	0.1–2
Egg powder	(Redo-Sanchez *et al.*, 2011)	Antibiotic detection	TDS	0.1–2
Chocolate	(Jordens & Koch, 2008)	Foreign body detection	Imaging	0.1–3
Vegetable oil (sunflower, peanut, soybean and rapeseed oil)	Li (2010)	Characterization of optical properties	TDS	0.2–1.5
Beverages (alcohol-free beer, Danish Pilsner, Danish Porter beer, French white wine, Irish cream liquor, German Bitter and Bulgarian Absinthe)	Jepsen *et al.* (2007)	Prediction of sugar and alcoholic content	TDS	0.1–1

Table 2. Comparison of pulsed and continuous wave THz systems.

	Pulsed	Continuous
Direct measurement of transient electric field	X	
Ambient noise	Low	High
Phase information retained	X	X
Monitoring of highly dynamic responses	X	
spectral resolution	Low	High
Intensity data		X
Frequency, time, depth data	X	
Speed	High	High
Cost	High	Lower
Data complexity	High	Low

detector PCA. The THz spectrum is generated by delaying the laser pulses going to the emitter and detector. This results in a time delay signal corresponding to one point in the THz spectrum. By changing the delay and measuring the signal corresponding to each delay the entire waveform is measured. The resultant signal is a measure of both the phase and amplitude of the THz field. Fourier transformation is used to convert the time-domain signal into the frequency domain. The absorption coefficient and refractive index of the material studied are directly related to the amplitude and phase of the transmitted field. This type of system may be used in transmission or reflection set-up, as shown in Fig. 1(a) and (b). Since Thz-TDS is a phase sensitive technique, in reflection or transmission mode, determination of the complex reflection coefficient can provide information on the dielectric constant of a sample. Recent developments in optical instrumentation have improved the speed of pulsed systems (Jin *et al.*, 2007; Kim *et al.*, 2010; Kim *et al.*, 2008).
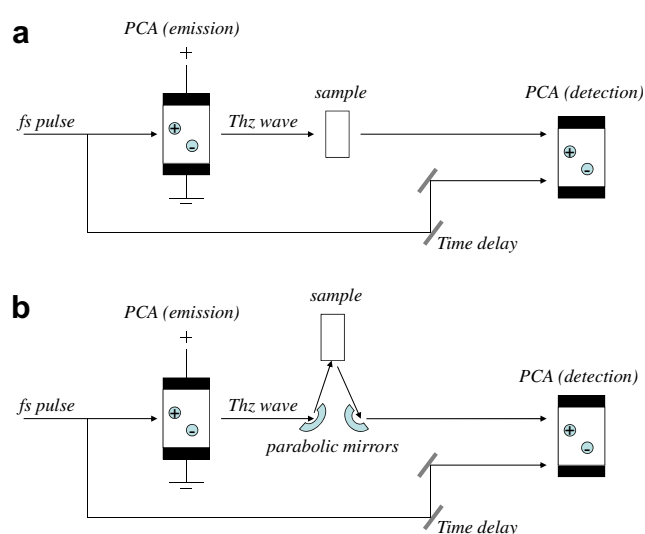
A typical Thz spectrum is shown in Fig. 2. In order to obtain a THz spectrum of a sample it is necessary to record a reference spectrum. In the case of transmission geometry, the reference is usually obtained by passing light through the system with no sample. For the reflection set-up, the reference is usually obtained by replacing the sample with a material of known reflectance, e.g. a mirror. Alternatively, it is possible to encase the sample in a container and measure the reflection from the air/container and container/sample interfaces. This method, known as self referencing, removes the need for a separate reference measurement (Jepsen *et al.*, 2007; Redo-Sanchez and Zhang, 2011).

Continuous wave systems do not require fs lasers, since the THz wave is generated by, for example, combining two frequency stabilised lasers, whose difference is in the THz region. Such systems are normally less complex and tend to be less expensive. than the pulsed systems, and are useful for detecting sharp spectral features, with resolution typically of a few MHz. THz imaging systems have also been developed, based on CW and pulsed systems described above. These are typically based on rastering, i.e. moving the sample along the x–y dimensions so that the THz beam passes through different spatial positions on the sample. An image is built up by recording a THz signal for each spatial position. This means that image acquisition is time consuming, varying from a few minutes to hours, depending on system characteristics and the desired spatial resolution. Due to diffraction, the image resolution obtainable is limited by the wavelength of the THz beam applied; however, various near-field set-ups have been proposed to reduce the pixel size to the order of 100 s of microns (Wang *et al.*, 2009; Yan *et al.*, 2009).
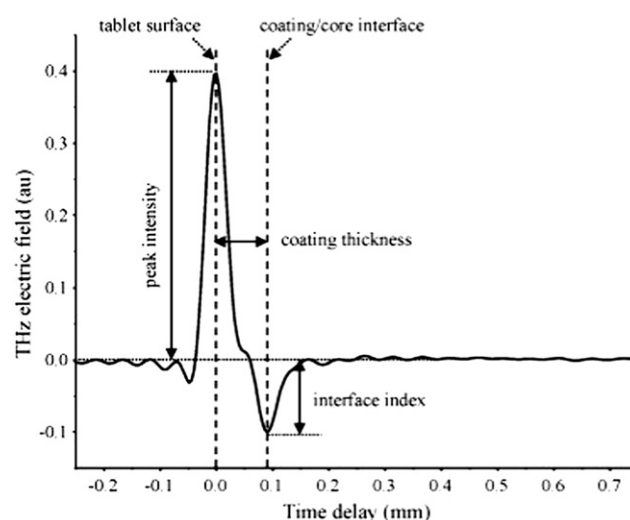

Fig. 1. Schematic showing pulsed THz acquisition in (a) transmission and (b) Reflection modes.

Fig. 2. Typical terahertz waveform. Reflection from the tablet surface (i.e. air/coating interface) and from the coating/core interface are indicated by dashed lines; drawn through arrows indicate how TPI parameters (peak intensity, interface index, layer thickness) are related to the waveform. (Maurer & Leuenberger, 2009).

Data analysis

Once the THz waveform has been acquired and transformed by Fourier transform into the frequency domain, it is necessary to calculate the absorption coefficient and refractive index for the sample under consideration. In the transmission set up, this is achieved using the sample $(E_s(v))$ and reference $(E_r(v))$ spectra. The transmission function in TDS is defined as follows (Hua & Zhang, 2010):

$$
\begin{aligned}
T(v) &= E_s(v)/E_r(v) \\
&= \left[4n/(n+1)^2\right]\exp\left[-\alpha d/2 + i2\pi v(n-1)d/c\right] \\
&= A\exp(i\Phi(v))
\end{aligned} \tag{1}
$$

where d is the sample thickness, v is the radiation frequency, c is the speed of light in a vacuum, n is the refractive index, α is the absorption coefficient, A is the amplitude ratio between the sample and reference spectra and ϕ is the relative phase difference. Equating the real and imaginary portions of Eq. (1), it is possible to calculate n and α for each frequency as follows:

$$
n = 1 + c\Phi/(2\pi vd) \tag{2}
$$

$$
a = (2/d)ln\left[4n/(n+1)^2/A\right] \tag{3}
$$

Certain compounds, such as pesticides or antibiotics, may exhibit sharp absorption peaks at specific THz frequencies. These features can be used for detection of contaminants in foodstuffs, and several examples are presented in the next section. Chemometric analysis (e.g. partial least squares regression) of the absorption coefficient or refractive index is also often applied for quantitative analysis of samples (Hua & Zhang, 2010; Nakajima *et al.*, 2007; Otsuka *et al.*, 2010; Shen, 2011).

Applications in food quality control

Moisture content determination

Due to the high absorbance of THz radiation by water, the most obvious application of THz spectroscopy would seem to be for quantification of moisture content in foods. This is especially important in controlling drying processes where low moisture content can alter the sensory qualities and shelf life of products. Moreover, the possibility of characterising the molecular nature of water in low moisture content products (e.g. free versus bound water) could be used for more accurate determination of shelf life.

Chua *et al.* (2004) and Chua *et al.* (2005) investigated the application of Thz-TDS in the 0.1–4 THz range for evaluation of moisture content of whole and crushed wheat. The crushed samples were housed in a PTFE sample holder during measurement, since this polymer is transparent to THz radiation, and the system was purged with nitrogen gas to avoid absorption by water vapour. Scattering of THz light due to the shape and orientation of grains led to the appearance of apparent peaks in the absorbance spectrum. Such artefacts were not present in the spectra of crushed wheat, and it was possible to identify water-specific absorption peaks in crushed samples varying in moisture content from 12 to 18%. Subtracting the THz spectrum of dry crushed wheat from that of wetted samples facilitated the development of a calibration model for prediction of moisture.

The moisture content of food wafers has also been examined using Thz-TDS (Parasoglou *et al.*, 2009). Dehydrated wafers were brought to various moisture contents (1–30%) by storage in a humid environment. Samples were then encased in sealed cells during for THz transmission measurement, in order to avoid dehydration by nitrogen purging of the THz system. A linear relationship between moisture content and THz time domain data (peak to peak amplitude) or frequency domain data in the range 0.2–0.6 THz was reported.

Although the potential for Thz-TDS has been demonstrated for several low moisture content foods, there have been few, if any, reported applications of THz imaging for moisture detection. Such investigations would be valuable for determining the spatial distribution of moisture in a food, which could be further related to processing conditions. A related application has been reported: THz imaging for detection of moisture content of paper (Banergee *et al.*, 2008). In this work, a raster based single pixel detection system was employed to map the distribution of moisture in paper at a spatial resolution in the mm range. An image representing 40×40 mm of paper was obtained within 2 min. Although the imaging of larger sheets of paper using this method would be prohibitively time consuming, these results suggest the feasibility of obtaining similar THz images for smaller, thin, low moisture content foods such as wafers, pasta or pastry sheets.

Residue detection

The presence of residues in foods is becoming a growing concern among consumers. Consequently, there has been interest in developing rapid non-destructive techniques for residue detection in foods, and THz spectroscopy is being evaluated for this purpose. Hua and Zhang (2010) examined Thz-TDS as a non-invasive tool for the detection of pesticides in food powders (sticky rice, sweet potato, and lotus root)(Hua and Zhang). They demonstrated that four pesticides (imidacloprid, carbendazim, tricylazole, and buprofezin) had specific absorbance peaks in the THz range (0.5–1.6 THz), while the spectra of the food powders themselves were similar to each other, exhibiting broad features. This indicated that the pesticide samples could be easily distinguished both from each other and from the food powder matrices. For instance, a linear correlation between absorbance at 0.89 THz and imidacloprid in sticky rice powder was reported. Partial least squares regression was carried out to predict imidacloprid concentration from the full spectrum, leading to promising results ($R^2 > 0.99$). However, it was noted that the physical properties of food

matrix (sticky rice powder in this case) can alter the signal due to scattering and absorption. In order to verify this method, the measurement of a wider range of physical conditions and food matrices is required.

More recently, Redo-Sanchez et al., 2011 investigated the potential of Thz-TDS for the detection of antibiotic residues in foodstuffs. Standard mixtures of antibiotics and powdered polyethelyne (transparent to THz energy) were prepared in pelletized form and transmission Thz-TDS spectra were obtained in the range 0.1 and 2 THz. Eight of the eleven antibiotics tested had specific signatures in the THz region, while two of them (doxycycline and sulfapyridine) were detectable when mixed with different food matrices (animal feed, milk, and egg powder). This indicates the potential for detection of some antibiotics in food substances using Thz-TDS. However, the mixture ratios used in this study were high (containing minimum 50% antibiotic content); further research is required to determine limits of detection for this application.

Foreign body detection

Detection of metallic contamination is rather straightforward in food processing; however, detection of non- metallic contamination, such as glass or plastic, is more challenging. The implementation of imaging systems with high spatial resolution would allow identification of the precise location of foreign bodies, thus minimising rejects. However, existing methods such as X-ray imaging do not perform well when the densities of the foreign body and the food product are similar. THz imaging, providing both phase and amplitude information, offers improved characterisation of foreign bodies. One demonstration of THz imaging for foreign body detection in foods has been reported, for the detection of contaminants in chocolate (Jordens & Koch, 2008). Due to its high fat and low moisture content, chocolate is relatively transparent to THz energy. When foreign objects, such as glass or plastic, are placed in chocolate, they alter the scattering profile of a transmitted THz wave and are thus detectable. This observation formed the basis of the work of Jordens & Koch, 2008. Foreign bodies (glass, stone and metal) were concealed within the interior of a bar of chocolate and THz images of the contaminated sample were obtained using a raster scanning Thz-TDS system operating in transmission mode. It was possible to identify foreign bodies in the chocolate sample, both in the presence and absence of its plastic foil packaging. However, variations in the chocolate thickness, caused by the morphology of the chocolate bar, resulted in variations in the transmitted signal. Another limitation highlighted in this study was the speed of THz image acquisition. The authors describe a method that could be implemented in order to speed up the process of imaging by sampling points rather than acquiring the entire waveform. In order to overcome the effect of sample thickness on signal variation, the authors suggest using two time delays. It was reported that such a method could facilitate scanning speeds up to 0.55 m/s.

Oil characterisation

Detection and characterization of water contamination of oils and other products is possible with THz spectroscopy. Gorenflo et al. (2006) employed Thz-TDS in transmission mode for characterization of the dielectric properties of oil–water complexes with water content ranging from 0.43 to 3.28%. This enabled determination of the absorption coefficient and refractive index of the samples, and thus determination of the amount and structure of water in these complexes. The results indicated the formation of a hydrogen bonded oil–water complex, rather than dissolved water clusters in the oil–water complexes. This represents an important advance in the understanding of the behaviour of water in oil, made possible by THz spectroscopy. More recently, Cunnell et al. (2009) developed a THz system capable of online measurements for water characterization in oil emulsions. This system, operating in transmission mode, included a quantum cascade laser source operating at 2.7 THz and a specially designed flow through cell for online measurements. It was reported that this system is capable of measuring variations in moisture content of approximately 0.1% in oil emulsions containing less than 3% water.

Li (2010) measured the optical properties of four types of vegetable oil using Thz-TDS in the range 0.2–1.5 THz. The oil samples (sunflower, peanut, soybean and rapeseed oil) were placed in a quartz cuvette for transmission measurement and the empty cuvette was used for reference measurement in this case. Due to multiple reflections at the air-cuvette and oil cuvette interfaces, it was necessary to develop a novel iterative algorithm for extraction of absorption coefficient and refractive index.

Beverage characterisation

It is important to characterise the dielectric properties of liquid foods in order to optimise certain processes, such as microwave heating. In addition, dielectric properties can be related to compositional information. Jepsen et al. (2007) developed a Thz-TDS system, operating in reflection in the 0.1–1.0 THz frequency range for measurement of the dielectric properties of liquids, in particular aqueous sugar and alcohol solutions. The developed system was employed for simultaneous determination of the sugar and alcohol content of commercial alcoholic beverages independent of other properties such as colour, organic matter content, carbonation and flavour.

Inspection of packed goods

Common packaging materials made from cardboard and polymers are transparent to THz radiation. This makes THz spectroscopy and imaging attractive tools for quality validation of packaged products. Although it is possible to apply some hyperspectral imaging techniques to packaged

goods, such analysis is typically limited to thin layer polymer packaging and imaging in the visible range (Gowen, O'Donnell, *et al.*, 2010; Gowen, Tiwari, *et al.*, 2010). As demonstrated by Jordens & Koch, 2008, it is possible to detect foreign bodies in packaged chocolate bars. Indeed, for most if not all of the applications listed in the preceding sections, it is possible to carry out the measurement while a product is cased in glass, plastic or paper. Another potential area of application is in the detection of production defects in packaging (e.g. holes or tears)(Morita *et al.* 2005).

Challenges and limitations

A major barrier to the adoption of Terahertz imaging systems at present is the high cost of detectors and sources. Although new developments have drastically decreased these costs, they are currently too high to render the technology economically beneficial for many applications. Related to this is the low acquisition speed typical of most systems. Although it has been demonstrated by Jordens & Koch, 2008 that high speed systems are feasible, many of these are based on sampling a sub-region of the THz waveform. The development of such systems is generally application-specific, since the sampled sub-region would need to match the desired response. Relatively high signal to noise ratio is also still problematic for certain regions of the THz spectrum, although this may be ameliorated by acquisition of multiple scans and averaging (although this increases the acquisition time). A number of technology providers are addressing these issues and developing improved, turnkey THz systems (Mantsch & Naumann, 2010).

Wu *et al.* (2007) listed a number of challenges, in the form of questions, to be addressed prior to the adoption of THz as a process analytical tool for the pharmaceutical industry. These challenges are equally relevant for the adoption of THz in the food industry, and are paraphrased below:

- How do measurement conditions affect quality, precision, accuracy and repeatability of THz spectra? For instance, in many cases, the THz system is encased and flushed with nitrogen gas – this may not be feasible in a food processing scenario.
- How can these measurement conditions be optimized for process monitoring? These will vary according to the process of interest; therefore an extensive research and development phase will be required for each application. This is often seen as a barrier in industry as it is expensive in terms of time and money.
- How can process and product knowledge be implemented into the data analysis to improve understanding of the THz spectra? Again, this is a process/product specific question and is best answered by collaborative research between the instrument provider and user.
- Can other spectroscopic techniques and chemometric modelling be integrated with THz for improved understanding of product and process interactions? Fusing complementary spectroscopic techniques to optimize

process monitoring capabilities could be regarded as the next major challenge in the implementation of PAT strategies for food quality control.

One of the limitations of THz spectroscopy for moisture content detection is that it is not suitable for high moisture products of thickness greater than 1 mm. This is due to the high absorption of THz radiation by water. For example, He *et al.* (2006) reported that the THz spectra of thin slices of skin and lean tissue samples were very weak due to water absorption. Even thin samples (approx 1 mm in thickness) were approximately opaque to THz above 0.7 THz. Reflection THz imaging is another option; however, the appearance of standing waves due to differences in the optical path length can produce image artefacts (McAuley *et al.*, 2006). Another challenge facing practitioners of THz spectroscopy is the effect of physical variations in a sample (e.g. particle size) on the refractive index of a material. This is particularly pertinent in the case of quality monitoring of fresh produce which shows high variability in this respect. Scattering effects may adversely affect measurements of Thz absorption in certain materials. When the grain size of solids is comparable to the THz wavelength, extinction spectra are severely influenced by scattering losses. Bandyopadhyay *et al.* (2007) investigated the effects of scattering on Thz-TDS spectra (in the range 0.2–1.2 Thz) of granular solids (Ammonium Nitrate, flour and salt) as a function of grain size (Bandyopadhyay *et al.*, 2007). Mie theory was applied to separate the contributions to the signal arising from absorption and scattering, thus enabling the total extinction spectra for various materials of different grain sizes to be extracted. However, the authors demonstrate that Mie theory cannot be successfully applied in the case of large non-speherical particle sizes.

Conclusions and future outlook

The ability of THz waves to pass through a wide variety of packaging materials, combined with their ability to characterise the molecular structure of many biological substances makes it an attractive process analytical tool for enhanced monitoring of food production. Although the potential of Thz-TDS spectroscopy and imaging has been demonstrated for a number of issues in food quality control, it is clear that much remains to be investigated in this area. The development of THz spectral library databases is something that is still in its infancy. Important work has already commenced on this, as evidenced by the applications described in the previous section. Although plenty more remains to be done, with the increased availability of turnkey THz systems, it is likely that the establishment of a library for the wide group of compounds related to food safety will be accomplished in the not too distant future. This, combined with lowering costs for sources and detectors, and faster systems, should lead to the increased adoption of THz spectroscopy and imaging for food process monitoring and control.

Acknowledgements

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