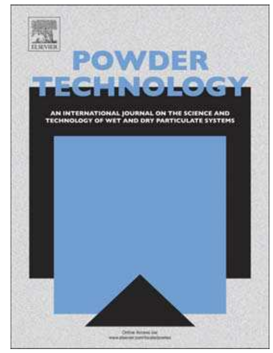

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PII: S0032-5910(16)30322-9
DOI: doi: 10.1016/j.powtec.2016.05.066
Reference: PTEC 11706

To appear in: *Powder Technology*

Received date: 4 January 2016
Revised date: 11 April 2016
Accepted date: 28 May 2016

Development of an open-ended microstrip stub apparatus and technique for the dielectric characterization of powders

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Abstract

A new apparatus and method to characterize the complex dielectric permittivity of powders is described. The apparatus and technique are used to determine the dielectric properties of detergent powder agglomerates at different conditions. The technique is based on the measurement of Scattering-parameters of an open circuit microstrip stub partly loaded with the test powder material. The scattering parameters relate the voltage waves incident on the ports of a microwave network to those reflected from the ports and can easily be measured with a vector network analyzer. A 3D finite element electromagnetic field simulation tool HFSS (High frequency structural simulator) is used to replicate the measured S-parameters and then extract the complex permittivity data from it. The method has been verified by measuring the dielectric properties of disks of known dielectric materials – specifically Duriod 5880 and Teflon. Results are in good agreement with manufacturer data sheets.

The complex permittivity of a range of detergent powder agglomerates with different moisture levels, at ambient and elevated temperatures, has been determined using this technique. Results are consistent with

predictions of how the water interacts with the different components of the detergent particles at these different conditions.

Keywords: Dielectric properties, Complex permittivity, Microwave dielectric characterization, Dielectric spectroscopy, Microwave measurement

1. Introduction

Microwave processing of materials is an area of on-going interest [1-6]. The dielectric properties of any material are obviously fundamental to its' suitability for any microwave/RF-based process [7, 8]. Having the capability to easily characterise the dielectric properties of a material over a range of conditions – such as those experienced during processing – will help to better understand and predict material behaviours during processing.

Detergent Powders – properties and characteristics

P&G (Procter & Gamble) makes many different types of detergent particles, such as agglomerates, blown powders, and extrudates. The most common manufacturing processes used are agglomeration and spray-drying. Agglomeration inherently makes higher density particles. Detergent particles will usually consist of surfactant(s) combined with organic polymers, such as polycarboxylates, soluble inorganic salts such as sodium carbonate, sodium sulphate and sodium silicate as well as insoluble materials such as silica and zeolite. Surfactants can have complex phase diagrams with a range of crystal, liquid crystalline and amorphous phases being present dependent on factors such as concentration, available water level, presence of electrolytes etc. Water can also be incorporated in inorganic hydrates formed from interactions with the non-surfactant components, such as sodium carbonate monohydrate.

Microwave radiation can be used to dry detergent powders using commercially available suitable equipment. However, microwave radiation can also be used to modify the properties of detergent particles, typically lowering the bulk density of the material by the formation of internal porosity. This is due to generation of steam inside the particles during heating. US Patent 6063751 [9] gives examples of this and the changes in the bulk density of the materials.

The water can be incorporated in multiple forms into a detergent particle –for instance, as non-associated free water, in surfactant liquid crystals or tightly bound in crystalline hydrates. The different strengths of these binding interactions mean that the impact of water on the dielectric properties of the particles is not straightforward. . Water that is tightly bound in a hydrate crystal lattice will not interact significantly with microwaves/RF. However water in a surfactant liquid crystal will be affected much more strongly. What will complicate the characterisation is that many of these hydrates have a temperature dependency – eg sodium sulphate decahydrate will decompose above 32 °C. Hence a priori knowledge of the composition is insufficient to predict dielectric properties during processing. Empirical determination over the range of conditions that will be experienced during processing is required.

Equipment Development

There have been several methods in the literature to measure the complex permittivity of the dielectric materials [10-16]. These methods could be classified mainly as either narrowband or wideband measurements. Each method has its advantages and disadvantages in terms of accuracy, ease of measurement, narrowband or wideband measurements, suitability for various liquids or solids, and frequency limitations etc[17].

In this paper, a new sophisticated low cost technique is introduced to precisely characterize the complex permittivity of dielectric materials in powder form. The complex permittivity of various commercially used detergent particles is determined using this technique. The resonant technique is used to perturb the resonant frequency of the open circuit stub by the dielectric material placed at its open end. The shifts in resonance frequency and dielectric absorption are measured to determine the complex permittivity of the material. The complete measurement procedure is explained in Section 3 and the measured results of different detergent agglomerates are presented in Section 5. The complex permittivity of different commercially used detergent particles is determined at room temperature and later on at elevated temperatures. This is relevant because the water-binding mechanisms can change at different temperatures – e.g. some hydrates will not form at elevated temperatures. This complex permittivity data can be used for further microwave processing of the detergent agglomerates.

2. Measurement Fixture

A quarter wavelength open circuit stub is designed on a microstrip line. The Duriod material used has $\varepsilon_r = 2.33$, $tan\delta = 0.0009$ and substrate thickness of 3.18mm. The width of the microstrip line can be calculated from [18].

$$Z_0 = \frac{120\pi}{\sqrt{\varepsilon_{eff}}\left[\frac{W}{d} + 1.393 + 0.667\ln\left(\frac{W}{d} + 1.444\right)\right]} \quad (1)$$

$$for \quad \frac{W}{d} \geq 1$$

Where

$$\varepsilon_{eff} = \frac{\varepsilon_r + 1}{2} + \frac{\varepsilon_r - 1}{2}\frac{1}{\sqrt{1 + \frac{12d}{W}}} \quad (2)$$

Where,

ε_r is the dielectric constant of the substrate material, W is the width of the conductor, d is the height of the substrate and ε_{eff} is the effective dielectric constant of a homogenous medium which replaces the dielectric and air region of the microstrip. The width W of the copper trace is selected to match the 50Ω input/output connectors and vector network analyser, this gives W=9.18mm.

The length of the open ended stub is selected as a quarter wavelength at the frequency of interest (2.45 GHz). A small adjustment in the length of the transmission line is required to account for fringing capacitance at the open end of the transmission line stub. The capacitive reactance due to fringing capacitance can be calculated as [19]

$$X_f = \frac{1}{j\omega C_f} \quad (3)$$

Where, C_f is the capacitance due to fringing fields.

When the sample holder is loaded with a dielectric material, the fringing capacitance increases and a down shift in resonance frequency of the quarter wavelength stub is noticed due to dielectric loading of the end of microstrip line. Fig 1 represents the fabricated microstrip stub along with the Teflon holder.

Fig 1 Microstrip dielectric measurement circuit

The substrate is placed on a 10mm thick aluminium block for proper grounding. The sample holder is made up of Teflon material with dielectric properties of $\varepsilon_r = 2.1$, $tan\delta = 0.001$. The sample holder has a wall thickness of 1mm, an internal diameter of 20mm, and a height of 20mm.

3. Measurement procedure

The process of determining the complex permittivity of a detergent powder agglomerates consists of two steps. First - measure the S-parameters of the transmission line with the test powder loaded in the sample holder. Second- replicate these results using the Ansys HFSS electromagnetic simulation tool. This software is widely used for very reliable electromagnetic field simulations of high frequency and high speed components. In the 3D EM simulation modelling step, the dielectric powder sample being measured is replaced with a homogeneous dielectric material having an arbitrary value dielectric constant and loss tangent. A series of iterative simulations is then carried out until the simulation shows a close agreement with the measured results. Figure 2 shows the flow chart of the procedure used to characterize the dielectric powder samples.

Fig 2 Dielectric characterization flow diagram

The open ended stub is measured without a sample holder and its S-Parameter results are compared with simulated results. An E5071C Network Analyzer by Keysight Technologies is used to measure the S-parameters of the circuit. Once an agreement between the simulated and measured S_{21} values is found, then the circuit is again measured with the Teflon sample holder glued to the design. The sample holder decreases resonance frequency and increases dielectric loss of the circuit due to dielectric loading of the transmission line (see Figure 3). A new series of simulations is then run to include the effects of the sample holder to the S- parameters of the circuit and a match between simulated and measured S-parameter response is found. Next, in order to verify the accuracy of the approach, some known dielectric materials (e.g. Duriod 5880 disks and Teflon cylinder) are placed in the sample holder and the S-Parameters measured. The complete setup including reference materials is then simulated using Ansys HFSS. A close agreement is found between measured and simulated results thus validating the technique. The comparison of measured dielectric constant and manufacturer data sheet of Teflon and Duriod 5880 is presented in Table 1.

Material	Measured dielectric constant	Manufacturer data sheet value
Teflon	2.102	2.10
Duriod 5880	2.205	2.20

Finally, the test samples are characterized by measuring the circuit S-parameters with the sample holder filled with the test detergent powders. The exact setup is simulated in HFSS by replacing the powder sample with an arbitrarily selected dielectric constant and loss tangent. An iterative series of simulations is then carried out until simulations agree closely to the measurement.

Fig 3 Measured S-parameters of circuit with and without sample holder

Fig 4 Change in S-parameters of the circuit by loading reference materials

4. Materials

To characterise the range of dielectric behaviours, four different, commercially relevant particles were obtained from P&G (Table 2). Two particles were made by agglomeration and two were made by spray-drying. Two different surfactants have been used in these tests – sodium linear alkylbenzene sulphonate (LAS) and sodium alkyl ether sulphate (average degree of ethoxylation = 1). These particles cover a wide range of commonly used detergent particle compositions.

Agglomerate 1 is made by agglomerating micronized sodium carbonate with sulphonic acid. The carbonate neutralises the acid to form sodium LAS ("dry neutralisation") and the agglomerates are then cooled and sieved. Agglomerate 2 is made by agglomerating a concentrated aqueous solution of sodium AE_1S with silica powder and micronized sodium carbonate followed by drying and sieving. The blown powder particles are produced by making slurry of the solids with water (typically 20 – 40% water) followed by spray-drying. LAS is the most common surfactant used in detergents globally. Sometimes it is also used in combination with AE1S as a co-surfactant., Samples of different agglomerates were exposed to a high humidity (80% Relative humidity at 32 degrees C) for four hours in order to vary the water level in the samples without even altering the physical properties of the materials. The weight increases of the samples were recorded. The samples were then collected in sealed bottles and allowed to further equilibrate at room temperature.

% Equilibrium Relative Humidity (or % water activity) is commonly used as a measure of the water activity of a sample. It is a measure of the humidity of air above a sample that is in equilibrium with that sample[20]. ERH is used as it is a measure of the free water in a sample – water in a crystal structure will not contribute. The % equilibrium Relative Humidities (% eRH) of the samples were then measured (at room temperature) and the % eRHs before and after conditioning are recorded below as well as the % weight increases. The dielectric properties of the materials were then measured at ambient temperature. Much of the water would have become incorporated into a range of carbonate and sulphate hydrates at these temperatures.

5. Results

The complex permittivity of the detergent powders has been determined. It has been observed that the conditioned samples all have higher loss factors - compared to the unconditioned samples - because of the higher water levels. This is to be expected. However, the scale of increases for different samples shows the impacts of the different chemical compositions on how the extra water is bound. The unconditioned samples ag1, ag2, bp1 and bp2 are unsuitable for microwave treatment due to very low loss factors. In contrast, the conditioned samples ag1', ag2', bp1' and bp2' (especially ag2') have higher loss factors which make them more suitable for higher energy absorption (quick microwave heating) when exposed to microwave radiations. Fig 5 shows the measured S-parameters of the circuit when loaded with the different detergent samples. Adding powder samples increase the fringing capacitance of the open end of the microstrip transmission line stub. This increases the effective electrical length of the stub; therefore, the stub reflects the input energy at a lower frequency as compared with empty holder. The higher the dielectric constant of the sample the bigger is the down shift in frequency. Sample ag2' shows a much stronger effect of water due to the nature of the AES surfactant compared to the LAS surfactant used in the other samples. AES can readily form a range of liquid crystals and the surfactant is probably holding much of the extra water in these liquid crystal phases where it can interact more with microwave radiation. In contrast, much of the extra water in the other conditioned samples is probably in the form of inorganic hydrates with carbonate and sulphate as the LAS surfactant does not form these liquid crystal phases as readily as AES. Hence the tightly-bound water in these hydrates is much less available to alter the dielectric properties of the samples.

Fig 5 Measured S21 response with sample holder filled with different detergent powder samples

Fig 6 and fig 7 show the real and imaginary parts of the complex permittivity of different samples determined at room temperature, respectively. The samples with higher loss factor/ imaginary part of complex relative permittivity can absorb electromagnetic radiations more quickly and heat up even faster.

Fig 6 Real part of complex permittivity of detergent powder samples measured at room temperature

Fig 7 Imaginary part of complex permittivity of detergent powder samples measured at room temperature

6. Elevated Temperature Measurements

The S-parameters of the detergent powder agglomerates discussed in Section 5 were measured at the ambient room temperature of 25 ^{0}C. In order to find out the effects of the temperature on the dielectric properties of a detergent powder, two ag2 AE_1S agglomerates samples with different moisture levels are prepared. The dielectric properties of the samples are expected to change at elevated temperatures because of the fact that some sodium carbonate and sodium sulphate hydrates (that are commonly encountered at ambient temperatures) will decompose at elevated temperatures. For example, sodium sulphate decahydrate will decompose above 32° C, while sodium carbonate heptahydrate decomposes above 30 °C. Hence the dielectric properties of such surfactant agglomerates can be expected to change as the agglomerates heat up during (microwave) processing.

The moisture is again added by exposing two samples of ag2 to a high humidity environment for a limited period of time. The weight of each sample is measured before and after adding water. Two samples with 2.8% and 7.4 % moisture level are then heated in sealed containers in an oven. The micro strip circuit is also kept in the oven to get it to the same initial temperature as the samples. Fig 8 shows the measurement setup. An Omega FOB-100 fibre optic thermometer is used to continuously monitor the temperature over time as the samples cool down. The test is performed outside of the oven so as to get a range of measurements while the samples cool down.

Fig 9 and Fig 10 show that, the dielectric constant and loss factor of the AES agglomerates decrease, as temperature decreases. This must be mainly due to the fact that some of the free water must be taken up by the sodium carbonate as the sample cools down enough for higher carbonate hydrates become stable. This is very clear from the decrease in the real complex permittivity of the 2.8% water sample. This decrease happens between 35 °C and 30 °C. This is consistent with the formation of sodium carbonate hexahydrate which becomes stable at temperatures below 32 °C. Hence the loss factor of the agglomerate decreases.

Fig 8 Measurement setup

Fig 9 Real part of complex permittivity of detergent samples measured at elevated temperature

Fig 10 Imaginary part of complex permittivity of detergent samples measured at elevated temperature

7. Conclusion

In this paper, a new apparatus and method is introduced to precisely characterize the complex permittivity of detergent powder materials. Real and imaginary parts of the complex permittivity of different detergent powder agglomerates are determined. The samples were conditioned to different moisture levels to determine the effect of moisture on dielectric properties. The effect of temperature on the dielectric properties of detergent particles is also determined. It is observed that the dielectric constant and loss factor of the detergent powders increases by increasing the moisture level or temperature of the powders. The data on dielectric properties of detergent powders can be used for further microwave processing of the materials. In general, the technique presented here can be used to accurately characterize the complex permittivity of any powder material.

8. Acknowledgement

This work is funded by AMSCI and BIS as part of the Chariot project.

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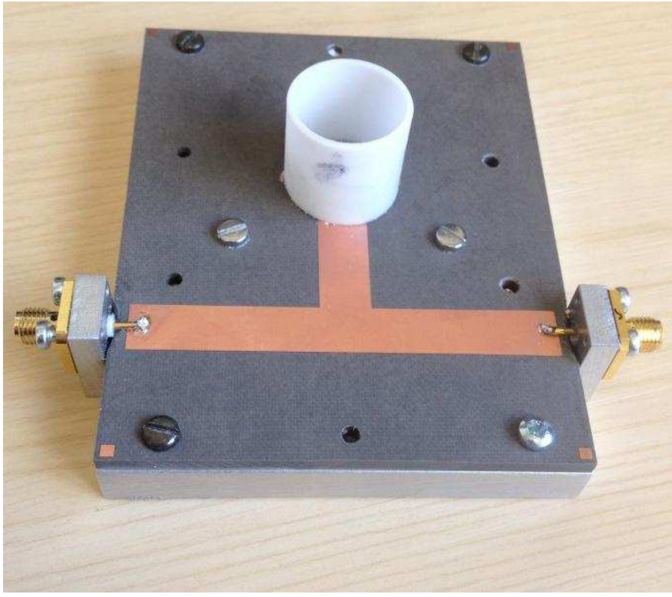


Fig 1 Microstrip dielectric measurement circuit

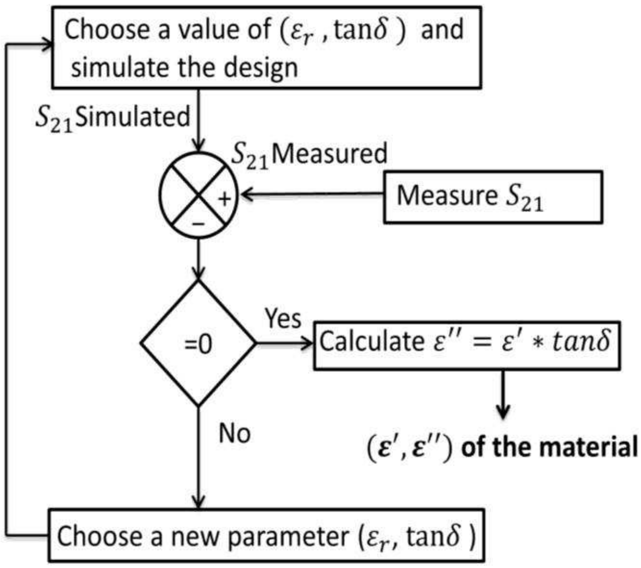


Fig 2 Dielectric characterization flow diagram

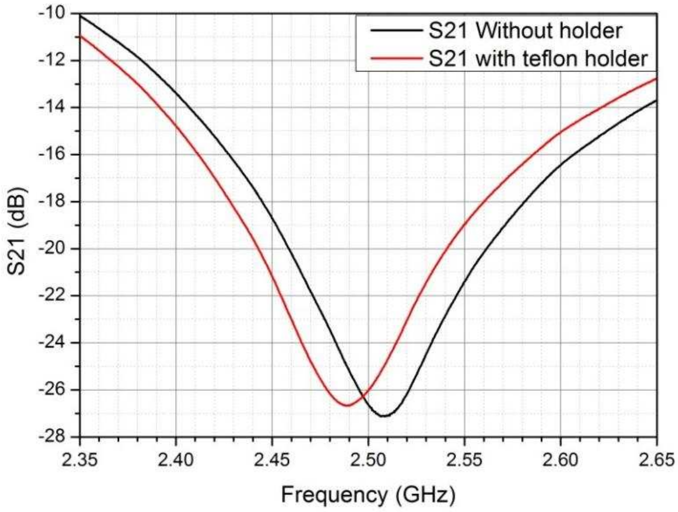


Fig 3 Measured S-parameters of circuit with and without sample holder

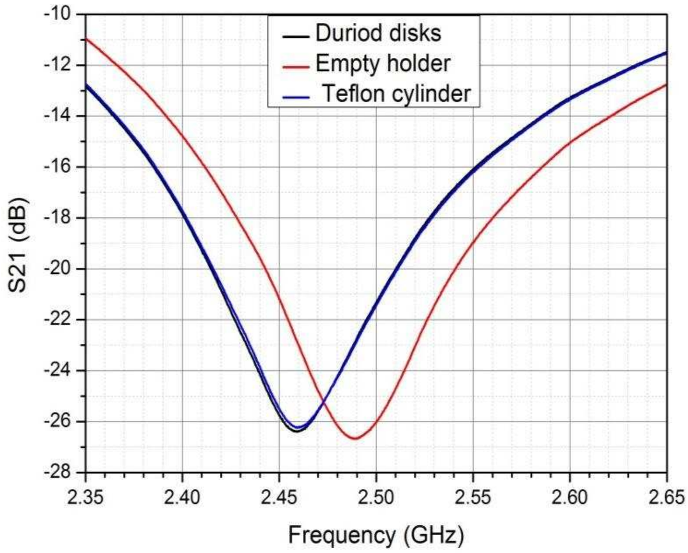


Fig 4 Change in S-parameters of the circuit by loading reference materials

Table1 Agglomerate compositions

Composition	**Agglomerate 1 (ag1)**	**Agglomerate 2(ag2)**	**Blown Powder 1 (bp1)**	**Blown Powder 2 (bp2)**
Sodium LAS	33		19.5	11.2
Sodium AE_1S		48		
Sodium Carbonate	64	33.5		0.6
Sodium Sulphate			67.75	70.1
Sodium Silicate (2.35R)			9	13.8
silica		14		
polymer			1.25	0.6
water	2	3.5	2	3.5
misc	1	1	0.5	0.5
% eRH before conditioning	36.2	31.5	18.6	29.2
% eRH after conditioning	64.6	64	80.9	69.4
% wt increase	10.4	10.6	9.9	16.6

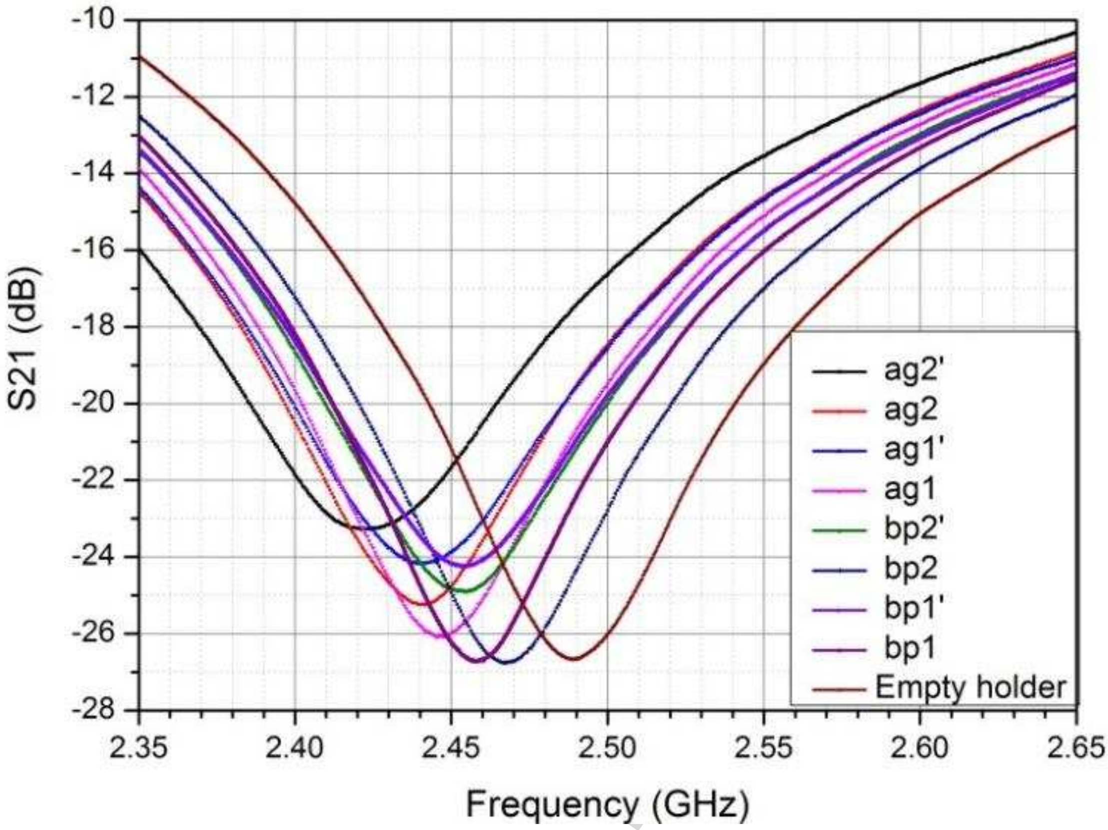


Fig 5 Measured S21 response with sample holder filled with different detergent powder samples

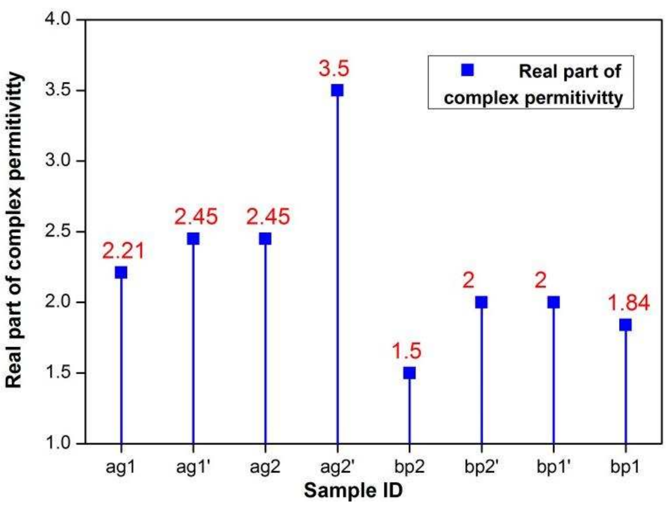


Fig 6 Real part of complex permittivity of detergent powder samples measured at room temperature

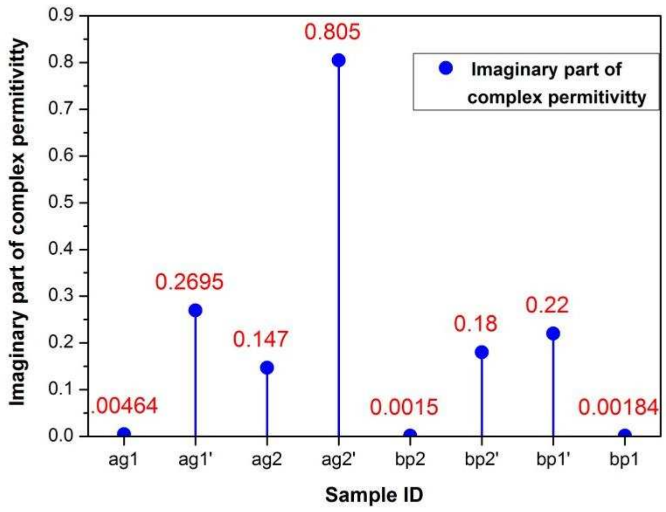


Fig 7 Imaginary part of complex permittivity of detergent powder samples measured at room temperature

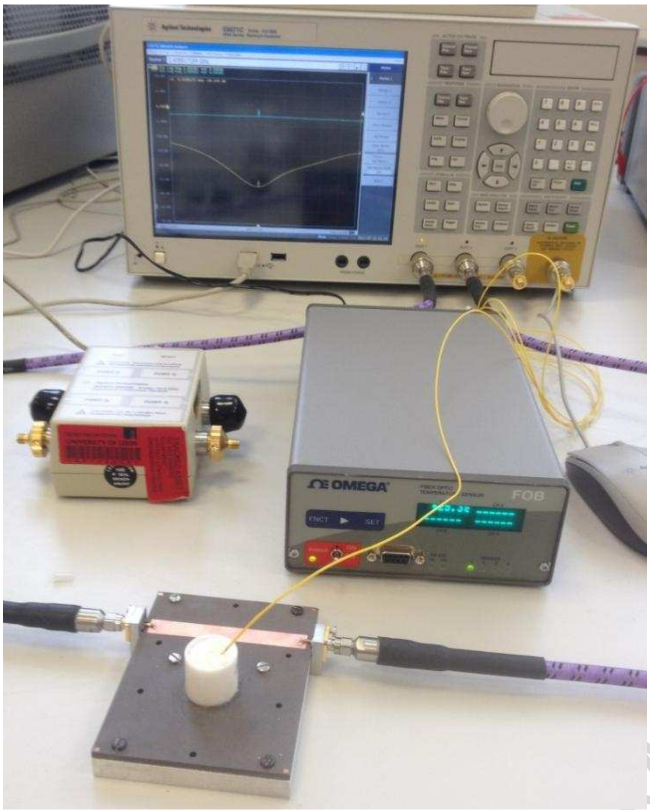


Fig 8 Measurement setup

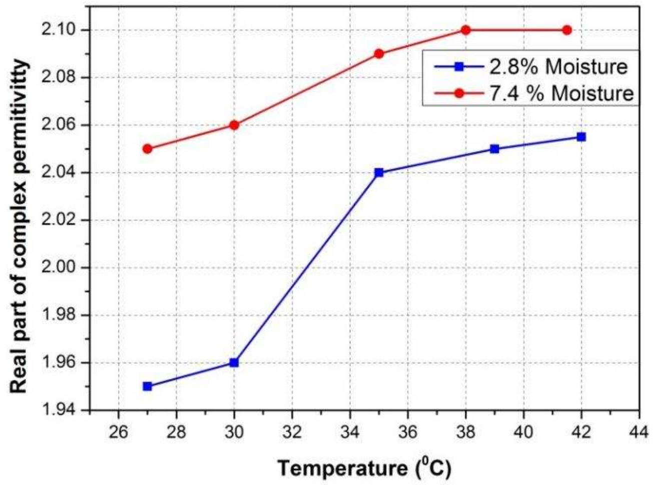


Fig 9 Real part of complex permittivity of detergent samples measured at elevated temperature

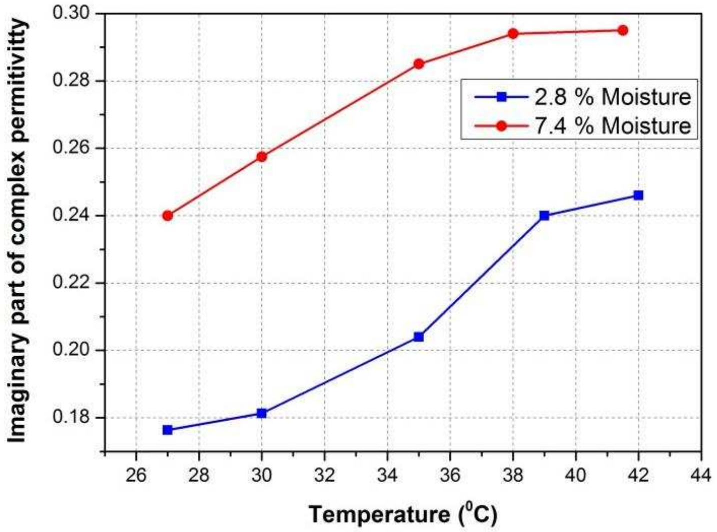
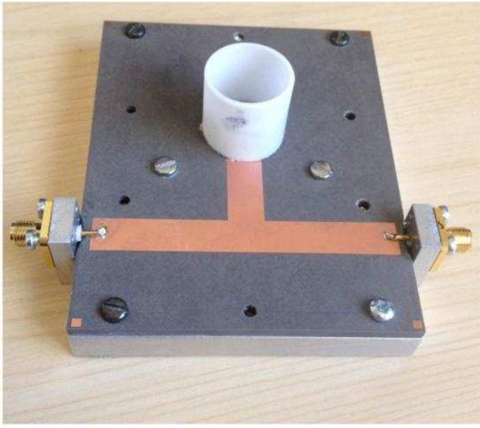
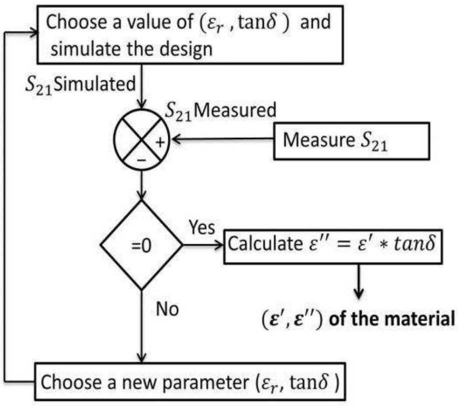


Fig 10 Imaginary part of complex permittivity of detergent samples measured at elevated temperature

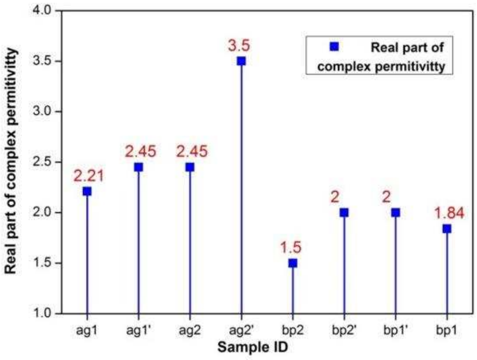


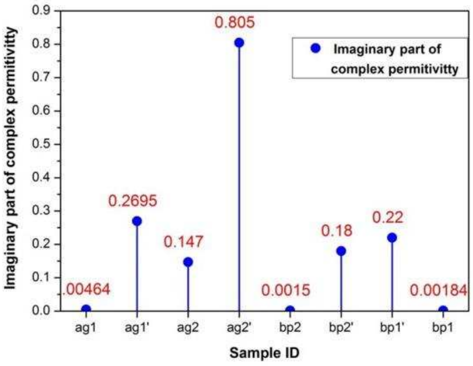
Microstrip dielectric measurement circuit



Dielectric characterization flow diagram

Results





Graphical abstract

Highlights

- A new method to characterize the complex permittivity of dielectric materials is presented.
- The complex permittivity of a range of detergent agglomerates with different moisture levels, have been determined using this technique.
- The effect of temperature on the dielectric properties of detergent particles is also determined.