Journal of Colloid and Interface Science 462 (2016) 110–122

Contents lists available at ScienceDirect

Journal of Colloid and Interface Science

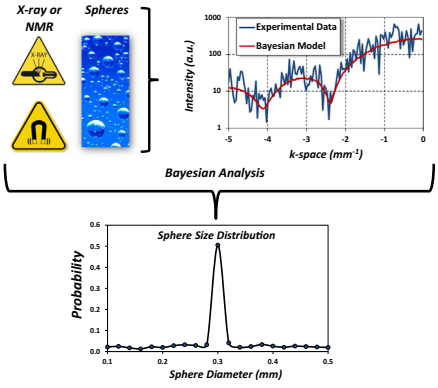
journal homepage: www.elsevier.com/locate/jcis

Rapid sphere sizing using a Bayesian analysis of reciprocal space imaging data

K. Ziovas [a], A.J. Sederman [a], C. Gehin-Delval [b], D.Z. Gunes [b], E. Hughes [b,1], M.D. Mantle [a,*]

[a] Department of Chemical Engineering and Biotechnology, University of Cambridge, Pembroke Street, Cambridge CB2 3RA, UK
[b] Nestlé Research Centre, Route du Jorat 57, 1000 Lausanne 26, Switzerland

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 16 July 2015
Revised 28 September 2015
Accepted 28 September 2015
Available online 30 September 2015

Keywords:
Bayesian analysis
Colloids
Dispersions
Size distribution
Nuclear magnetic resonance
X-ray
Foams
Bubbles

ABSTRACT

Dispersed systems are important in many applications in a wide range of industries such as the petroleum, pharmaceutical and food industries. Therefore the ability to control and non-invasively measure the physical properties of these systems, such as the dispersed phase size distribution, is of significant interest, in particular for concentrated systems, where microscopy or scattering techniques may not apply or with very limited output quality. In this paper we show how reciprocal space data acquired using both 1D magnetic resonance imaging (MRI) and 2D X-ray micro-tomographic (X-ray μCT) data can be analysed, using a Bayesian statistical model, to extract the sphere size distribution (SSD) from model sphere systems and dispersed food foam samples. Glass spheres-in-xanthan gels were used as model samples with sphere diameters (D) in the range of 45 μm $\leqslant D \leqslant$ 850 μm. The results show that the SSD was successfully estimated from both the NMR and X-ray μCT with a good degree of accuracy for the entire range of glass spheres in times as short as two seconds. After validating the technique using model samples, the Bayesian sphere sizing method was successfully applied to air/water foam samples generated using a microfluidics apparatus with 160 μm $\leqslant D \leqslant$ 400 μm. The effect of different experimental parameters such as the standard deviation of the bubble size distribution and the volume fraction of the dispersed phase is discussed.

* Corresponding author.
E-mail address: mdm20@cam.ac.uk (M.D. Mantle).
[1] Current address: Department of Chemistry, Durham University, Lower Mountjoy, South Road, Durham DH1 3LE, UK.

http://dx.doi.org/10.1016/j.jcis.2015.09.066
0021-9797/

1. Introduction

Dispersions are multiphase systems, where at least one phase is dispersed in a continuous phase. These systems are commonly found in many different industries ranging from the food, pharmaceutical and petrochemical industries. Two types of dispersions with particular interest and relevance to the food industry are foams and emulsions. In foams a gas phase is formed from spherical or near spherical bubbles within a continuous liquid phase whereas in emulsions both the dispersed and the continuous phases are liquids. The bubble or droplet size distribution, herein referred to as a generic sphere size distribution (SSD), is an important structural characteristics for foams and emulsions. The exact form of the SSD of dispersed systems may result from the production and treatment processes [1]. In addition, the contribution of the dispersed phase to the viscoelastic properties of emulsions and foams can also depend on the SSD [2,3]. Furthermore, the SSD can be related to the physicochemical properties and the stability of these systems [4,5]. Some examples from different industries include the use of foams in wet processing in the textile industry [6], where the foam SSD effects the efficiency of the process, or the pharmaceutical industry where emulsions are used as drug delivery systems [7]. In recent years foams find an increased application in the food industry with the emergence of functional foods [8]. Aerated food systems and emulsions can help to enhance sensory response whilst reducing the amount of flavouring used [9], reduce the calorific density of foods [10] and deliver nutrients or bioactive molecules much more efficiently [11]. Regardless of the specific application, it is highly desirable for scientists to be able to non-invasively characterise these systems and monitor their evolution over time. Currently there are a number of available methods which we outline briefly here.

1.1. Traditional sphere sizing methods

Common analytical techniques that are used to determine the SSD in foams, colloidal dispersions, or emulsions are microscopy, light scattering methods, X-ray tomography, ultrasound spectroscopy and electrical conductivity measurements [12]. Optical microscopy is the most widely used technique as it produces images with very good spatial resolution. It is relatively simple to use and has been developed extensively for a wide range of systems and applications [13]. In a recent report an analysis of photographic images, acquired through an optical microscope, was performed to examine the bubble size distribution in cake batter and the SSD was found to be in the range of 25–80 μm [14]. Light scattering methods have also been used to gain SSD information and cover a wide range of sizes from sub-micron level up to centimetres [15]. Additionally, multiple light scattering techniques can provide additional information about the dynamics and the evolution of foams and emulsions [15,16]. For example, a commercial light scattering apparatus was used on a water-in-oil emulsion with a mean droplet size in the region of 1 μm [17]. Ultrasound spectrometry has also been suggested as a potential low cost method for analysing samples under a range of process conditions. However the method can be seriously affected by particle impurities and the thermo-physical properties of the continuous and dispersed phase, which need to be previously determined [13,14,18]. Finally, 3D X-ray tomography is a very powerful technique capable of providing high resolution spatial images of the system's microstructure [19,20]. However, it can be very time consuming, because of the image acquisition and reconstruction process.

1.2. Magnetic resonance sphere sizing methods

In recent years a number of nuclear magnetic resonance (NMR) techniques have been used to characterise the SSD in foams/emulsions and there is currently an expanding activity in this area [6,21,22]. The majority of the approaches have exploited pulsed field gradient (PFG) NMR techniques which measure molecular self-diffusion. This method relies upon the fact that the diffusion of molecules within the dispersed phase is restricted. By applying pulsed magnetic field gradients it is possible to measure the effect of the restriction that molecules experience [23]. The analytical solution of this behaviour is well established and has recently been reviewed [24]. Previous research using PFG-NMR methods to extract the SSD assumed, a priori, a log-normal form for the distribution [21]. This approach gives good estimations for many cases where droplet size distributions range from ~3 μm up to ~200 μm. In more recent studies, regularisation techniques have been implemented in order to determine the droplet size distribution of emulsions without assuming a specific functional form [25]. One drawback associated with the standard PFG-NMR method is the long acquisition times which can range from 5 to 20 min for a specific sample. It is clear therefore that the traditional techniques cannot be used to study transient processes which happen at much shorter time scales [26]. The Difftrain PFG-NMR pulse sequence was developed and used on various emulsions where the acquisition time was reduced down to 3 s but this sequence is difficult to implement and is not considered as standard [27]. Despite the fact that there are various applications of PFG-NMR to gain SSDs from emulsions in the literature, there appears to be very little research into gas/liquid foams, likely because of the poor signal to noise ratio that gas phase NMR suffers from. A recent study made use of PFG-NMR to determine the bubble size distribution in a non-overflowing pneumatic gas–liquid foam [28]. In this study the bubble size was in the range of 0.5 mm up to 6 mm and the bubble size distribution followed the Weibull model. Propane was used at the "NMR active gas" which represents a significant drawback of the PFG-NMR method with regard to health and safety.

The acquisition of spatially resolved 2D and 3D magnetic resonance images (MRI) of the microstructure of foam systems has also been demonstrated. In one study, researchers used susceptibility weighted MRI to investigate alveolar food products, e.g. bread dough [29]. Despite the fact that this method provides improved contrast compared to classical MRI the maximum spatial resolution they achieved (0.15 mm) was not enough to provide quantitative information for the bubble sizes which range from 0.05 mm to 0.3 mm, since the smaller bubbles could not be resolved [29].

NMR relaxometry techniques have also been used to measure the SSD in bubbles and droplets. Different microstructures result in different T_2 relaxation times which can then be related back to different droplet sizes. Emulsions have again been studied extensively [21], but there is a very limited amount of research performed on foams. In one reported study the ^{1}H T_2 relaxation time of water has been used to study hydrogel foams [30]. These foams were composed by gelatine and sodium dodecylsulphate (SDS) and the aeration was achieved with mechanical stirring. In that work it was found that it is possible to determine the size in the range of 100–200 μm with the time required for one measurement being 6 min and the total time for one experiment was 45 min. However, in order to correlate the T_2 relaxation with the bubble size distribution of the foam a set of computationally heavy simulations were required, which assumes that the structure of the sample is known. Additionally, parameters such as the surface relaxivity, the diffusion coefficient and the magnetic susceptibility have to be determined independently.

In the last decade there has been a rising interest in exploiting the Bayesian approach to data analysis in many fields of science [31,32]. Sphere sizing using Bayesian probability theory and NMR data has been developed recently and used to study the SSD in multiphase gas/liquid flow [33] as well as to estimate the pore size