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MEASURING BUBBLE POPULATIONS IN GASSY MARINE SEDIMENTS: A REVIEW

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1 INTRODUCTION

Gassy sediments have been observed at a large number of locations throughout the world[1]. These bubbles primarily consist of biogenic methane (*i.e.* methane generated by the anaerobic decomposition of organic matter in the sediments[2]), although thermogenic gases generated within deeper, higher temperature sediments[3] (>50 °C) may contribute to a lesser degree. At present regions of gassy sediments can be reliably mapped through the identification of "gassy" features[4], which include sub-surface features observed in high-resolution seismic records (*e.g.* acoustic turbidity, blanking and columnar disturbance), seabed features (pockmarks, active vents and biological / geological anomalies) and bubble-plumes in the water column.

While the spatial mapping of gassy sediment is itself useful, a number of applications require more detailed information. Of particular importance are measurements of the Void Fraction (*VF*), *i.e.* the fraction of the sediment volume that is composed of bubbles, and for many applications the Bubble Size Distribution (*BSD*). For example, climate modellers require *VF* to refine the, presently uncertain, estimates of the contribution of marine sources[5] to atmospheric methane (a major greenhouse gas). Further, as sediment pore pressures and sediment strengths are highly sensitive to both *VF* and *BSD*[6,7], knowledge of these parameters may allow the oil prospecting industry to site offshore structures more reliably and avoid blowouts which occur during drilling operations. Finally, the acoustic properties of gassy sediments are extremely sensitive to BSD and VF[8-10], hence making these important parameters to marine surveyors and sonar modellers.

This paper will review the state-of-the-art techniques available for measuring both VF and BSD in gassy sediments, with the aim of placing certain constraints on expected bubble populations. Such a review is both necessary, owing to the many published research projects that have examined gassy sediments, and timely, owing to the recent acquired ability to image bubbles on the 10s µm scale. As a considerable component of the existing measurement techniques are based on acoustic properties, Section 2 will review the current acoustic propagation theories. Section 3 will review available measurement techniques and conclusions will be drawn in Section 4.

2 THEORETICAL ASPECTS

Bubbles in marine sediments can be categorised into three generic categories[11] (Figure 1). Interstitial bubbles (Type 1) are confined to the pore space between the sediment grains, this places a maximum limit of approximately 100 µm on the bubble radius[12]. This scenario can be theoretically described by considering solid frame of sediment grains filled with a two-phase (gas-water) fluid. Reservoir bubbles (Type 2) displace only the pore water, while the sediment grains remain in place. The presence of sediment displacing bubbles (Type 3) describes the only scenario in which the structure of the sediment grains is modified, and can be theoretically described by isolated gas bubbles surrounded by a saturated medium.

A number of acoustic models have been developed for gassy sediments. As these have been used to infer *VF* and *BSD* from measured acoustic properties (see Section 3), the remainder of this section will briefly describe these models. The model that is most frequently used to interpret acoustic data from gassy sediments is that of Anderson and Hampton[8,9], which considers the acoustics of gas bubbles surrounded by a saturated sediment matrix, *i.e.* the scenario described by Type 3 bubbles. This modifies a resonance based acoustic theory for gassy water[13] to incorporate the effects of a finite shear modulus and differences in damping terms between sediment and water. Several assumptions are made, namely monchromaticity, linearity and non-interacting bubbles. In practical terms this will limit the model to small amplitude, single frequency signals and low *VF*.

Predicted compressional wave velocities and attenuations are displayed for a variety of monotonic populations[14], each with a *VF* of 0.1 % (Figure 2). At frequencies less than bubble resonance frequencies, pressure and volume changes in the bubbles are in phase and the bulk properties of the media dominate[8,15]. The low bulk modulus of the gas reduces the velocity of this low frequency region to less than that of saturated sediment. For frequencies near resonance the velocity is highly dispersive and the attenuation peaks. Above resonance the velocity approaches that of the saturated medium, while attenuations are dominated by scatter from non-resonant bubbles. Recent work[10,16] has focused on the best manner of determining the saturated bulk and shear moduli and dissipation factors for Type 3 bubbles, parameters which the Anderson and Hampton model is highly sensitive to.

A range of additional theories have been presented, all of which entail the assumption of non-interacting bubbles, monochromaticity and linearity. A number of these are based on modified versions of the Biot theory, in which the pore fluid properties are adjusted to incorporate gas bubbles[17-22]. This intrinsically assumes Type 1 bubbles and hence limits the bubble radii to less than 100 µm. A theoretical approach for determining the compressional wave velocities of Type 2 bubbles has been presented by Brandt[23], who considers a random stacking of spherical grains of four different sizes. Lee[24] modifies the Biot Gassmann Theory to incorporate a differential pressure on velocity, which through alternative formulations for the fluid bulk modulus and density allows either Type 1 or Type 2 bubbles to be considered. The prediction of backscatter from gassy sediment has received much attention, with bistatic models developed for sandy and mud sediments[25-28], some of which include resonance effects.

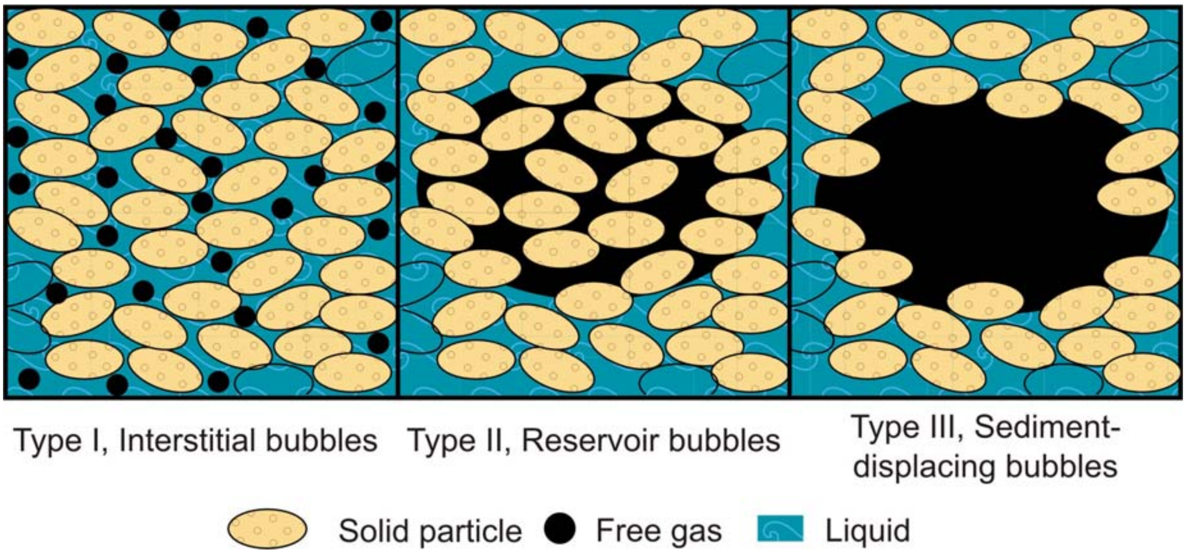

Figure 1. Generic bubble classification for bubbles in sediments. From Anderson *et al*[11].

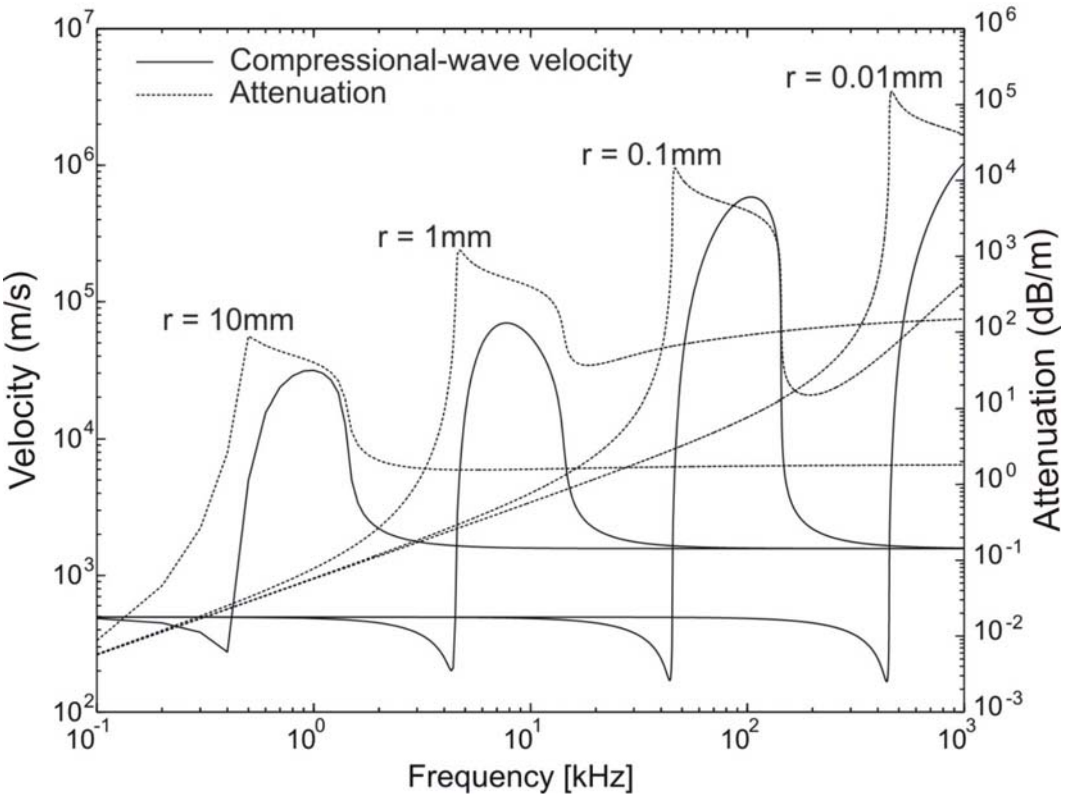

Figure 2. Compressional wave velocities and attenuations predicted by Anderson and Hampton model[8,9] for a variety of monotonic bubble populations with *VF* of 0.1 % and radii of 10 mm, 1 mm, 0.1 mm and 0.01 mm. From Best *et al.*[14]

3 MEASUREMENT TECHNIQUES

The present techniques available for measuring *BSD* and *VF* in gassy sediments can be classified into two main categories, namely density based techniques and acoustic based techniques.

The first class of techniques utilises the contrast between the densities of the gas bubbles[29] (densities of order 10^0 kg/m^3) and the densities of both the pore water[30] and sediment grains[31] (both of order 10^3 kg/m^3). Table 1 summarises *VF* and *BSD* obtained for gassy sediments using these density based techniques. Scanning-Electron-Microscope (*SEM*) images have been obtained for both un-pressurised cores from the Western Irish Sea[32] and resin-impregnated samples of artificial gassy sediments[33]. These results (Table 1) may however be compromised by depressurisation effects and the inability to determine between gas and water filled voids respectively.

The use of X-Ray CT scanners provides a more reliable manner in which to exploit this density contrast. Such scanners have been used to image both pressurised cores[11,14,34-38], un-pressurised cores[39] and artificial gassy samples[40-42], *i.e.* those generated under laboratory conditions through the growth of gas bubbles in, or injection of gas bubbles into, natural sediments and gels. Measured BSD display a decrease in the number of bubbles as radii increases[11,43], with the number of bubbles related to the radii, in mm, through

$$N = 10^a R^{-b} \qquad (1)$$

where *N* is the number of bubbles per m^3 at each radii *R*, and the parameters *a* and *b* control the VF and gradient of the distribution respectively. The only values available for the parameters *a* and *b* in the literature for 3-dimensional bubble distributions are based on cores collected from gassy sediments in Southampton Water, U.K.[43]. Information from 2-dimensional scans are converted into 3-dimensional populations under the hypothesis that the radii measured in the horizontal plane represents the shortest (a = 4.79, b = 2.30) and longest (a = 4.66, b = 1.92) dimension of the 3-dimensional bubbles (Figure 3).

The major limitation of this work has been the minimum bubble size which could be detected. This is controlled by the resolution of the scanner, which pre-2003 varied from 4x10^2 to 10^3 µm. However, the recent development of higher resolution scanners (< 10 µm), offers the ability to detect much smaller bubbles. Recently published work which utilises such scanners, observe bubble radii as low as 10s of µm in natural[41] and artificial sediments[38,40].

VF (%)	Radii observed (µm)	Measurement technique	Gassy sample	First author and reference
-	-	SEM image of un-pressurised core	Gassy mud from W. Irish Sea	Yuan[32]
0.4-19.8	-	SEM images of resin impregnated samples	Artificial gassy sediment	Sills[33]
6.0	*5x10^2 - 2.1x10^4	X-Ray CT scanning of pressurised cores	Gassy mud from Southampton water	Best[14]
0.5 – 4.5	*5x10^2 - 5x10^3	X-ray Ct scan of pressurised core	Gassy mud from Eckernforde Bay	Abegg[35]
-	*5x10^2 - 8x10^3	X-ray CT scan of pressurised core	Gassy mud from Eckernforde Bay	Lyons[37]
< 2.0 mean of 0.1	*5x10^2 - 5x10^3	X-ray CT scan of pressurised core	Gassy mud from Eckernforde Bay	Wilkens[34]
2.4	-	X-ray CT scan of pressurised core	Hydrate sediment from Cascadia Margin	Abegg[36]
>6.0	*1x10^3 – 2.5x10^3	X-ray CT scan of unpressurised core	Gassy mud from Chesapeake Bay	Hill[39]
< 9.0	*2x10^2 - 1x10^4	X-ray CT scan of pressurised core	Gassy mud from Eckernforde Bay	Anderson[11]
-	Many in 10s µm range	X-ray CT scan of pressurised core	Gassy mud from Eckernforde Bay	Reed[41]
-	3x10^1 - 9x10^3	X-ray CT scan	Lab. based bubble growth in natural samples	Reed[38]
-	-	X-ray CT scan of pressurised core	Bubbles injected into mud and gel	Boudreau[40]
-	-	X-ray CT scan of pressurised core	Bubbles injected into mud and gel	Johnstone[42]

Table 1. Void fractions (VF) and range of bubble radii observed in gassy sediments using techniques based on the density contrast between gas bubbles and the surrounding medium ('-' denotes no available information, while '*' denotes the lower limit set by the resolution of the scanner used.)

Knowledge of the shape and types of bubbles present is also important, as this will also affect physical and acoustical properties of the sediment. While all of the investigations listed in Table 1 have observed sediment displacing (Type 3) bubbles, recent work[33] has provided evidence for Type 1 bubbles in natural sediment samples[38]. Although the resolution of scanners has been sufficient no observations of Type 2 bubbles have been reported in unconsolidated sediments. Concerning bubble shape, both spherical[14,38] and non-spherical bubbles[11,14,37,38] have been observed in sediments. The bubble shape depends on both the sediment type and the bubble size, with spherical bubbles more commonly observed in sands / silts. Elongated bubbles which are orientated with their longest axis in the vertical plane are observed in the more fine-grained muds[14], with bubbles becoming more elongated as bubble size increases[38].

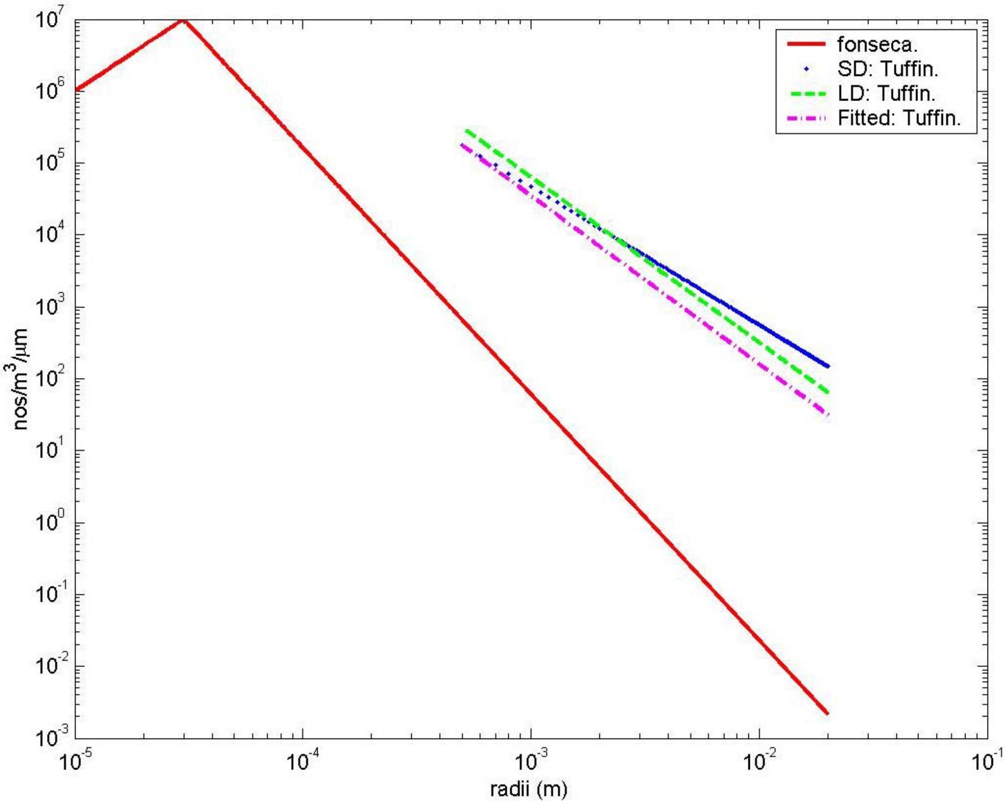

Figure 3: Bubble Size Distributions available from the literature, including: *BSD* used by Fonseca[47] to model backscatter from gassy sediments; *BSD* determined by Tuffin[43] from CT scans (using Smallest dimension (SD) and Longest Dimension (LD)); and fitted *BSD* determined by Tuffin[43] through the comparison of predicted and measured acoustic properties.

Those techniques which are based on acoustics focus on the use of a variety of acoustical theories to infer VF and BSD from compressional wave properties. As compressional wave properties are strongly dependent on the gas content these are favoured over shear wave properties, which are primarily controlled by the properties of the sediment frame[34]. Although a large number of authors have measured the compressional wave properties of gassy sediments, only a limited number of these have computed *VF* or *BSD*. Wilkens and Richardson[34] use *in situ* measurements (from Eckernforde Bay) of compressional velocities and attenuation spanning 5 to 400 kHz to determine a "qualitative" estimate for the smallest bubble size present. Between 21 and 25 kHz velocities change from being dependent on *VF* to being independent of *VF*. These researchers interpret this frequency range as the transition from using driving frequencies coinciding with the dominant resonances exhibited by the polydisperse bubble population, to driving frequencies greater than this, and therefore assume that there are negligible bubbles with radii less than 0.3 mm. Unfortunately, this cannot be confirmed though the X-ray CT scanning of pressurised cores owing to the resolution of the scanner being limited to 0.4 mm. Andreassen *et al.*[44] interpret the reduced velocities below bottom simulating reflectors associated with gas hydrates as corresponding to a *VF* of 2 %. Similarly, Tinkle *et al.*[45] interpret low velocities measured *in situ* at sediment depths of 60 m as representative of a *VF* of 1 %, while Edrington and Calloway[46] convert velocities measured frequencies below frequency in gassy marine sediments in the Mississippi delta to a *VF* of 0.065 %.

A number of researchers use the comparison between observed acoustic properties and theoretical predictions to infer both *VF* and *BSD*. Note that the range of radii used in each comparison is set according to additional information and the effect of radii outside this region is not examined. Comparison of measured and predicted backscatter from sands[26] and muds[47] indicates *VF* less than $1\text{x}10^{-3}$ % in sands and up to 9 % in muds. In both sediment types a distribution of bubble radii, which range from 10 μm to approximately 2 mm and peaks at approximately 30 μm, is assumed (Figure 3). This is obtained by combining *BSD* measurements of bubble radii greater than 0.2 mm in gassy sediments obtained using X-Ray CT scanning techniques[11] with those measured in the ocean water column[48]. *In situ* bubble populations are also obtained for a region of gassy mud in Southampton Water, U.K. through the comparison of *in situ* phase velocities and attenuation measured from 1 to 11 kHz with the predictions of the Anderson and Hampton model[14,43]. This uses radii from 0.5 to 20 mm (determined from CT scans of pressurised cores) and predicts a *VF* of 6 % and a *BSD* in which the number of bubbles decreases as radii increase. This is described by Equation 1 with a = 4.54 and b = 2.57.

Additional researchers adopt the use of combination frequency techniques to infer *VF* from non-linear scattering terms[49,50]. Such techniques have been successfully used in the water column to measure bubble populations[51,52] and involve the simultaneous insonification of a bubbly medium with two sound fields at different driving frequencies. If one of these frequencies corresponds to a bubble resonance, non-linear terms will be generated in the scattered field. Although this represents an extremely promising technique for measuring bubble populations in gassy sediments, the results obtained for Eckernforde Bay are unexpectedly low and ambiguous (*e.g.* *VF* could be either $3\text{x}10^{-4}$ or $7\text{x}10^{-3}$) and are therefore treated with caution.

A cautionary note should be made be concerning *VF* and *BSD* information obtained from acoustic measurements. These will only be as reliable as the theoretical model used. At present the only theoretical model available for the more common Type 3 bubbles is limited to the assumptions of monochromaticity, linearity and non-interacting bubbles. These may be violated by the use of multi-frequency, high amplitude signals and the observation of void fractions up to 9 %. A number of researchers have noted that the Anderson and Hampton model[8,9] considerably over predicts attenuations measured at resonance[34,52,53].

Theoretically there exists a minimum and maximum bubble size that can be supported by a marine sediment. The minimum size will be controlled by the ability of bubble stabilisation mechanisms to counteract the effects of surface tension, which tends to drive free-gas into solution and increases as bubble size decreases[54]. The maximum size will be determined by the ability of the sediment to resist the increasing buoyant force associated with larger bubbles. Unfortunately such a theoretical approach is not possible owing to a lack of information concerning the nature of the bubble skin and the highly variable rigidities of marine sediments. Even for the much simpler scenario of bubbly water it is uncertain what the minimum stable bubble size is. This will depend on the concentration of organic matter[55] and hydrophobic suspended particulate matter[56], both of which may act to stabilise microbubbles, *i.e.* bubbles with radii less than 15 μm.

4 CONCLUSIONS

This review allows certain constraints to be placed on bubble populations in gassy sediments. Bubbles predominantly possess radii from tens of μm to 10 mm, while *VF* lies between 0 and 10 %. The number of bubbles generally decreases as bubble size increases, as described in Equation 1. This trend has been confirmed for bubble radii from 0.2 to 10 mm, with the trend followed at smaller radii uncertain. Although Type 3 bubbles are the dominant type observed, recent results using higher resolution CT scanners suggests that Type 1 bubbles are also present. There has been no evidence observed for Type 2 bubbles in unconsolidated sediments (these may be more applicable to consolidated sediments). Spherical bubbles are more common for smaller bubble sizes and in more-coarse grain sediments, while vertically orientated, elongated bubbles dominate in the more fine-grained muds.

The continued use of recently developed higher resolution X-ray CT scanners, with resolutions of sub 10 μm, presents an obvious manner in which our knowledge of *BSD* and *VF* in gassy sediments can be improved. However, this should be accompanied by the further development of acoustic theories, which omit the assumptions of monochromaticity, linearity and non-interacting bubbles.

5 ACKNOWLEDGEMENTS

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