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ACCEPTED MANUSCRIPT

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The ultrasound-triggered explosion of an endoskeletal antibubble yields a predicable fragment size distribution

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Recently, the first high-speed video of a fragmenting antibubble was published. Nonetheless, this fragmentation process was not fully understood. Owing to a recent study on fragmenting glass, we can now conclude that an antibubble under tensile stress undergoes an exponential fragmentation process. This note gives a brief theoretical explanation and the first experimental data of a fragmenting antibubble. This is a highly relevant finding, as the fragmentation predictability of acoustically active antibubbles is required for their potential use in ultrasound-guided drug delivery.

Recently, the first high-speed video of a fragmenting antibubble was published in Ref. 1. Nonetheless, this fragmentation process was not fully understood. Owing to a recent study on fragmenting glass, we would like add a brief explanation on the fragmentation process of the antibubble. A very recent study by Kooij et al. showed that explosive fragmentation of Dutch tears leads to a predictable fragment size distribution.²⁾ In addition, the study showed that an increased stress regime of millimeter- to centimeter-size glass-like materials results in an exponential, rather than a power-law distribution of the fragments. Kooij et al. did not draw conclusions on the scale-invariance of this exponential process or whether microscopic materials might yield similar fragment distributions.

We hypothesised that this elegant explanation holds for the size distribution of explosively fragmenting microscopic silica-comprising endoskeletal antibubbles during sonic smashing.¹⁾

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This would be a highly relevant finding, as the fragmentation predictability of acoustically active antibubbles is required for their potential use in ultrasound-guided drug delivery.³⁾

Antibubbles are by definition gas bubbles containing at least one liquid core. Although antibubbles are typically highly unstable structures,^{4,5)} they may be stabilised by adsorbing colloidal particles at the interfaces, which is referred to as Pickering stabilisation.⁶⁾ The colloidal particles may form a solid skeleton structure, fixating the liquid core, as demonstrated with the aide of confocal microscopy and scanning electron microscopy.¹⁾ Stabilised antibubbles have been proposed as theranostic agents.^{4,5,7)}

We investigated an antibubble of 20- μm diameter comprising a solid skeleton and shell of agglomerated zinc oxide and fumed silica, whose dynamic fragmentation behaviour had been observed by us,¹⁾ but had not been properly understood. A still frame of the antibubble is shown in Fig. 1a. A scanning electron microscopy image of similar endoskeletal antibubbles has been shown by Kudo et al.¹⁾

To cause an explosion, the antibubble (Fig. 1a) had been subjected to a three-cycles pulse of 1-MHz, 1-MPa ultrasound, whilst recorded at ten million frames per second.¹⁾ As antibubble fragmentation has been associated with inertial cavitation, the medium was allowed to settle and released gas allowed to diffuse or buoy for an additional minute before a still frame was recorded, shown in Fig. 1b. The 56 antibubble skeleton fragments visible in this still frame were measured using MATLAB[®] (The MathWorks, Inc., Natick, MA, USA). Fig. 1c shows the size distribution of these fragments, with the same representation as Kooij et al.

Following the methodology of Kooij et al., an exponential curve

$$P(d) = Ce^{d/d_1} \quad (1)$$

was fitted through the fragment size distribution. This curve-fitting yielded the distribution parameter $d_1 = 0.9 \mu\text{m}$. The scalar value $C = 7$ is an indicator for the number of fragments included in the distribution and therefore of less interest than d_1 .

Our results presented in Fig. 1c are remarkably similar to the results for tempered glass presented by Kooij et al.²⁾ We attribute the overrepresentation of fragments $\frac{d}{d_c} > 3$ to the fact that a two-dimensional microscopic representation of a volumetric distribution automatically results in an overrepresentation of particles greater than the depth of field.

The exponential fit indicates that the antibubble was under tensile stress during fragmentation. We approximated the tensile stress during fragmentation by rearranging the equation relating tensile stress to characteristic length in Kooij et al.²⁾ and substituting parameter values

from literature:^{8,9)}

$$\sigma_t = \kappa_{Ic} \sqrt{\frac{15\sqrt{3}(1+\nu)}{4d_c}} \approx 0.0008 [\text{MPa m}^{\frac{1}{2}}] \sqrt{\frac{15\sqrt{3}(1+0.2)}{4 \cdot 2.5 [\mu\text{m}]}} \approx 1 \text{ MPa}, \quad (2)$$

where κ_{Ic} is the mode-I fracture toughness, ν is Poisson's ratio, and σ_t the tensile stress. Using conservative values when computing the tensile stress, it is clear that the stress is caused by the ultrasound field, rather than the Laplace pressure of < 15 kPa.

The fragmentation of the antibubble can be characterised by a single parameter, namely the distribution parameter d_1 . For sonic micromaterials smashing, this parameter is a more objective descriptor than the number of fragments or their geometric spread, since microscopic images only reveal a minute slice of the full event.

One might wonder, whether the droplet core inside an antibubble is of influence on the fragment size distribution. Although endoskeletal microbubbles without cores have not been produced, we may assume that their Laplace pressure is even lower than droplet-containing endoskeletal antibubbles.¹⁰⁾ Assuming no other tensile stress components of relevance and assuming identical acoustic conditions, the resulting fragment distribution should be exponential then, too. As the characteristic length follows from the solid material characteristics and the tensile stress applied, the characteristic diameter of an endoskeletal microbubbles should be on the same order as an endoskeletal antibubble. This is not necessarily the case, however, for the distribution parameter, as this parameter might be influenced by the exact geometry of the structure undergoing fragmentation.

Whilst Kooij et al. performed fragmentation experiments not only under tensile stress but also in absence thereof, demonstrating the influence of the tensile stress applied on the type of size distribution,²⁾ we could only fragment endoskeletal antibubbles under sonication. The strength of Pickering-stabilised microbubbles has thus far prevented them from being smashed at non-stress conditions. Therefore, there remains a need for control experiments to establish, whether the fragmentation distribution under such conditions would be a power distribution, indeed. In addition, it would be of great interest to investigate why Pickering-stabilised microstructures, notoriously hard to crack under isostatic conditions, are apt to fragmentation when applying moderate dynamic pressure amplitudes.

Since Kooij et al. used glass and sugar, it is most interesting to extend this research to applications of these materials that require a known fragment size distribution. These may include the controlled smashing of sugar-coated pills for consistence checks,¹¹⁾ the production of less-than-6- μm theranostic particles, and the controlled re-sizing of calibration microspheres. Furthermore, other brittle materials need to be included in such studies, such

as kidney or salivary stones.

In conclusion, the characterisation method proposed by Kooij et al. has been shown to hold for a silica-comprising antibubble. Whilst Kooij et al. found that macroscopic materials under tension yield an exponential fragment size distribution, we showed that a microscopic material under sonication yields an exponential fragment size distribution, as well.

Owing to this study, we are closer in understanding endoskeletal antibubble fragmentation and, with it, ultrasound-guided drug release from them.

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Figures

Fig. 1: Fragment size distribution of an antibubble: **a** an inlay showing a still frame of the antibubble before sonication; **b** an inlay showing a still frame of the antibubble skeleton fragments 1 minute after sonication; **c** the size distribution normalised by the characteristic diameter $d_c = 2.5 \mu\text{m}$. An exponential distribution fit $P(d) = Ce^{d/d_1}$ is indicated by a bold line.

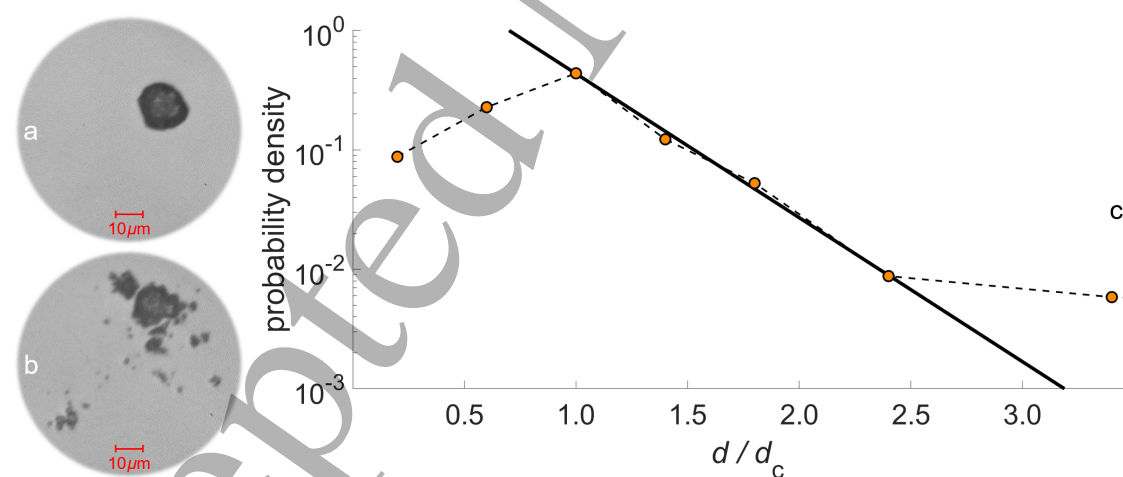


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