Incendivity of Ultrasound Applied in Explosive Atmospheres

Lars Hendrik SIMON, Thomas FEDTKE, Volker WILKENS, Michael BEYER Physikalisch-Technische Bundesanstalt Bundesallee 100, 38116 Braunschweig, Germany

2013-01-14

1 Introduction

Ultrasound is frequently used in explosive atmospheres especially in the processing, cleaning and measurement industries. However, little is known on the effectiveness of this ignition source. The current threshold value of 1 mW/mm^2 [1] is based only on theoretical estimations in analogy to other ignition sources rather than on experimental data. Therefore, it is assumed that this threshold includes a large safety margin. As high power ultrasound occurs in level measurement and can support or excite various processes such as parts cleaning, air cleaning, or chemical processes, an exaggeratedly large safety margin might result in a burden to potential innovations. An overview of potential ultrasonic high power applications for industrial processing can be found in [2]. For a comprehensive analysis on the hazards of ultrasound in different applications, ultrasound of 20 kHz and 1 MHz frequencies coupled into air and into liquid media was investigated.

2 Theory

For gaseous media the generation of high sound pressure amplitudes is technically limited. The coupling of high acoustic amplitudes is poor as a result of the large difference between the acoustic impedance of the solid transducer and the gas phase. For technical ultrasound, the amplitude is proportional to the deflection of the transducer material in resonance. At high frequencies, the transducer dimensions have to be diminished and, thus, the driving voltage has to be augmented, which can eventually destroy the transducer. The absorption by the gaseous medium of propagation at kHz frequencies is rather low [3] and with stepped-plate transducers sound pressure levels of up to $L_p = 170$ dB can be obtained at distances of the order of 0.5 m [2]. In the investigations of Lierke [4] concerning ultrasound standing wave fields, an amplitude gain of 20 dB compared to a propagating wave was reported. This means an augmentation in pressure by factor 10 since the dB scale is logarithmic: $L_p = 20 \cdot \lg(p/p_0)$, with $p_0 = 20 \ \mu$ Pa. Moreover, according to Nyborg [5], in standing wave fields strong heating rates can be expected in the sound pressure anti-nodes. Thus, a resonant standing wave field is considered to be the worst case situation when a highly sound absorbing target material is placed in the pressure anti-node which transforms the acoustic energy into heat. Thus, the target's hot surface could trigger ignition of a surrounding explosive atmosphere. In the absence of such an absorbing target the heating will be negligible because the acoustic attenuation in gases and dusts are of the order of 10^{-1} 1/m or less [3,6].

For liquids, two major aspects have been considered. First, at MHz frequencies, in liquids, high intensity ultrasound can be focused on focal areas with a width of a few millimeters. However, if this focus is placed on a liquid's surface, the acoustic radiation force will scatter the liquid's surface into droplets. Therefore, the situation becomes more critical if an absorbing material is fixed at this surface, whose acoustic impedance matches the liquid's. Then, again, this absorbing target material could transform the acoustic energy into a hot spot if it is temperature resistant and can therefore attain temperatures above the auto-ignition temperatures of burnable vapors or gases. In this case, the heated target could trigger an ignition of an explosive atmosphere above the liquid.

The second major aspect is acoustic cavitation within liquids. This phenomenon occurs if the stresses of the negative sound pressure phase exceed the binding forces and the liquid is ruptured or micro-fine gas bubbles which are dissolved in the liquid pulsate in the sound pressure field. These small cavities oscillate and grow under the influence of the ultrasonic wave. At a certain point, the bubbles collapse rapidly with collapse times of a few micro-seconds. Inside the collapsing bubbles, extreme conditions with temperatures of several thousand kelvin and pressures of more than 10 MPa are attained. Moreover, the collapsing bubbles excite shock waves of several 100 MPa in their vicinity [7] [8]. Finally, attraction forces between the oscillating bubbles cause them to build clusters [9]. Experiments concerning the exposure of polydisperse bubble media to shock waves showed explosions within the bubbles [10]. Moreover, the studies of so-called "cavitation-ignition bubble combustion" by Nguyen and Jacqmin at NASA [11] report evidence of combustion processes within acoustically generated cavitation bubbles. Thus, explosions in clusters of bubbles close to the liquid's surface might ignite an explosive atmosphere above the liquid.

3 Experimental setups for ignition tests

To model each of the three worst case situations described in chapter 2, an experimental setup was developed and ignition tests were conducted for each situation. For ignition tests with airborne ultrasound an ultrasound generating unit (USU) consisting of a piezoelectric transducer, a booster and a sonotrode was used (Figure 1a). It faced a reflector made of steel with an integrated pressure sensor. In this arrangement it was possible to generate sound pressure levels of up to 184 dB (re. 20 μ Pa) in the sound pressure anti-node at the reflector's surface. At this position, an alkaline earth silicate wool target was suspended. This material has an absorption coefficient close to 1 at frequencies above 1 kHz. For temperature monitoring, a thermocouple was positioned in the center of the target. The USU, reflector and target were integrated into a Hartmann tube, which is a standard apparatus for minimum ignition energy tests in dusts (Figure 1a). Dust filled into the tube could be dispersed by a compressed air shock of the dust disperser at the bottom of the tube. After dispersion it passes by the ultrasonically heated target at different concentration levels. As dusts, sulfur, maize starch and calcium stearate were used.

For ignition tests with liquid-borne ultrasound a setup as schematically illustrated in Figure 1b, was developed. First, a HIFU transducer (HIFU: high intensity focused ultrasound) was integrated to insonify a target made of polyetheretherketone (PEEK). This material is appropriate since plastic materials have an acoustic impedance relatively close to liquid media compared to solids. Moreover, their viscoelastic behavior is beneficial for the sound absorption and transfer of the acoustic energy into heat. The material has a melting point of around 340°C. Finally, it has a low thermal conductivity. Hence the generated heat within the material is thermally isolated from the liquid.

Demineralized and physically relaxed water was used instead of burnable liquids. This is due to the fact that the acoustic characteristics of the HIFU transducer refer to water as a medium of propagation.



Figure 1: Experimental setups for ignition tests in dust-air atmospheres in a Hartmann tube (a) and at liquid surfaces (b). For the ignition tests with focused ultrasound a HIFU transducer at 1 MHz was used. For experiments concerning acoustic cavitation this transducer was replaced by a 20 kHz power sonotrode.

If other liquids were used instead of water it would be necessary to calibrate the HIFU transducer and all the measurement equipment, for each liquid. Moreover, the use of a solvent with its own explosive atmosphere would result in atmospheric layering. For volatile liquids, directly above the liquid surface the vapor-air mixture would be too rich and the easiest ignitable mixture would be at some uncertain distance from this surface. Therefore, we used an external gas mixture preparation to supply the test vessel with a precisely premixed atmosphere. Carbon disulfide at concentrations from 2 % to 10 % and diethyl ether at concentrations from 10 % to 13 % with air were used as an atmosphere. These two vapors have low auto-ignition temperatures of 95°C (carbon disulfide) and 175°C (diethyl ether) [12]. In previous experiments concerning ignitions at small hot surfaces these concentrations turned out to be the most easily ignitable ones. To monitor the concentration within the test vessel we used a paramagnetic oxygen analyzer (Oxymat). The surface temperature of the PEEK target was measured with a thermograpic camera.

For ignition tests in cavitating liquids the HIFU transducer was replaced by a 20 kHz power sonotrode with a maximum electrical input power of 650 W at atmospheric pressure. It was designed to excite strong cavitation for liquid processing. Optionally, a cannula could be used to seed gas bubbles filled with an explosive gas mixture into the cavitation field. As explosive atmospheres, initially, hydrogen-air mixtures with 22 % hydrogen were used because this concentration has the minimum ignition energy [13]. In addition, again, a mixture of diethyl ether (13%) and air was used. The sonotrode was driven in the continuous as well as in the pulsed mode for one hour.

4 Results

For the ignition tests concerning airborne ultrasound, ignitions could be observed for sulfur dust. To the authors' knowledge, this was the first ignition of an explosive atmosphere ever triggered in an ultrasonic

Simon, L. H.

field. The results from the tests in dependence on the sound pressure level measured at the reflector are shown in Figure 2a. Up to sound pressure levels of 178 dB no ignition occurred. Target equilibrium temperature did not exceed 300°C. Above a transition range from 178 dB to 181 dB all ignition tests led to ignitions of the dust atmosphere. As shown in the Figure, the results of the ignition tests were statistically evaluated based on a logistic regression [14]. The curve shows a sharp separation of the probability of ignition at 178.5 dB. However, to evaluate the ignition probability more reliably, a larger sample is needed.

For maize starch and calcium stearate no ignition was observed. When the dusts were dispersed, a drop in the target temperature was observed. Magnification of the used absorbers under a microscope showed residues of smoldered dust which clogged the fibrous structure of the alkaline earth silicate wool.



Figure 2: a) Results of ignition test within sulfur dust - air atmospheres with airborne ultrasound and b) at the liquid surface with carbon disulfide - air mixtures at the liquid surface for focused ultrasound of 1 MHz coupled into water.

The ignition tests at the liquid surface in carbon disulfide also yielded ignition as shown in Figure 2b. When the PEEK target was insonified by the focused sound beam a small bubble of molten plastic erupted. The thermographic camera recorded bubble temperatures of up to 500 °C when the bubble was formed. These bubbles had a diameter of 2 mm to 5 mm. At high powers the carbon disulfide - air mixture ignited directly. Especially at lower power levels, however, the bubble did not directly trigger an ignition but rather cooled down while a larger hot surface around the bubble formed. Its diameter was approximately 10 mm to 15 mm in diameter. In the latter situation, in some cases, ignition still occurred a few seconds after bubble formation. Figure 2b shows that there is a transition zone between 287° and 327° where ignitions and non-ignitions occurred. In the case of ignition the temperatures signify the target's maximum temperature directly prior to ignition. In the case of non-ignition the temperatures signify the maximum target temperature averaged over a time period when no further temperature augmentation could be observed.

The ignition tests with diethyl ether yielded a threshold between ignition and non-ignition of 3 W of acoustic power. At lower power levels even for long insonification times of 3 minutes, no bubble was formed and the target's surface did not heat up above 150° C, which is below the standard auto-ignition temperature of 175° C.

The ignition tests in cavitating liquids yielded no ignition of the explosive atmosphere. The temperature recording of the PEEK target during insonification by the sonotrode showed no significant change in temperature. However, the water heated up in a homogeneous way to approximately 90°C within 1

Simon, L. H.

hour. High-speed camera recordings of the cavitation bubbles and the gas mixture bubbles seeded by the cannula showed that the former scattered the latter to smaller bubbles.

5 Discussion

The results from the ignition tests with airborne ultrasound show a sharp separation between non-ignition below 178.5 dB and ignition above 181 dB. However, as the dB scale is logarithmic, 3 dB means a factor of $\sqrt{2}$ in sound pressure. To confirm the threshold of 178 dB, the largest sample of ignition test was taken at this value. The temperature at this pressure level ranged from 240°C to 300°C. This broad distribution can be explained by the inhomogeneity of the used alkaline earth silicate wool as absorption material. The standard auto-ignition temperature of the sulfur dust used is 250°C. Thus, an ignition temperature of above 300°C in the setup used is reasonable. However, it has to be pointed out that for ignition, in addition to sound pressure levels above 178 dB, an absorbing material with an absorption coefficient close to 1 at frequencies above 1 kHz is needed to transform the acoustic energy into heat. Moreover, to heat up the target, insonification times longer than 5 seconds are needed and the target has to be fixed in the sound pressure anti-node of the resonant standing wave field. Finally, sulfur is a reactive dust with a comparatively low auto-ignition temperature.

The ignition tests with the HIFU transducer show that it is possible to ignite an explosive atmosphere at the liquid surface when focused ultrasound heats up an absorbing material which is fixed at the surface. Because the temperatures of these bubbles always exceeded the auto-ignition temperatures of carbon disulfide and diethyl ether ignition cannot be excluded. If the acoustic power of 3 W, which had to be exceeded for ignition, is related to the beam area at focus with a radius of 1.3 mm (-10 dB, i.e. decay of the intensity to 0.1 of the maximum intensity at focus), an spacial-averaged temporal-averaged intensity [15] of $I_{SATA} = \frac{P}{\pi r^2} \approx 600 \text{ mW/mm}^2$ can be derived which is larger by a factor of 600 than the current threshold value of 1 mW/mm² [1].

The ignition tests in cavitating liquids show that the heating of an absorbing material at the liquid surface can be excluded as an ignition mechanism. The major part of the acoustic energy is consumed by the formation of cavitation bubble clusters. In addition, the absorption increases with frequency. Thus, at 20 kHz it is much lower than at 1 MHz. Moreover, bubbles that rise to the surface accumulate beneath the target. This air cushion of bubbles shields the target from the acoustic wave and, therefore, reduces the heat production due to absorption.

The cavitation bubbles themselves do not trigger ignition even if merged with bubbles of explosive atmosphere at easily ignitable conditions. Their small size, the short collapse time and the strong cooling rate due to the surrounding liquid impede this mechanism.

6 Conclusion

The investigations showed that it is possible to ignite explosive atmospheres by airborne and liquidborne ultrasound. However, the ignition tests showed that this is only possible when many specific conditions are met at the same time. For airborne ultrasound a sound pressure level of 178 dB has to be exceeded, in liquids an acoustic intensity of 600 mW/mm^2 . In both cases, a specific target material has to be placed at a specific position. In a resonant standing wave field this was at the pressure anti-node, for focused ultrasound in liquids close to the focal length. At 20 kHz cavitation bubble clusters aimed at seeded gas bubbles with gas-air burnable did not trigger an ignition. In conclusion, these ignition tests show that the current threshold value of 1 mW/mm² comprises an exaggeratedly large safety margin and does not refer to conditions that are necessary to set off an ignition.

Acknowledgments

The support of the Deutsche Gesetzliche Unfallversicherung (German Social Accident Insurance) is gratefully acknowledged.

References

- [1] EN 1127-1, *Explosive atmospheres Explosion prevention and protection Part 1: Basic concepts and methodology*, Std., 2011.
- [2] J. A. Gallego-Juárez, G. Rodríguez, V. Acosta, and E. Riera, "Power ultrasonic transducers with extensive radiators for industrial processing," *Ultrasonic Sonochemistry*, vol. 17, pp. 953–964, 2010.
- [3] S. G. Ejakov, "Acoustic attenuation in gas mixtures with nitrogen: experimental data and calculations," Acoustical Society of America, vol. 113, no. 4, pp. 1871 – 1879, 2003.
- [4] E. Lierke, "Effekte und Anwendungen von Luft-Ultraschall hoher Schallpegel," Kolloquium der Abteilung 1 und 3, Physikalisch-Technische Bundesanstalt, June 2011.
- [5] W. Nyborg, "Heat generation by ultrasound in a relaxing medium," *Journal of the Acoustic Society of America*, vol. 70, no. 2, pp. 310–312, 1981.
- [6] A. Dukhin and P. Goetz, Ultrasound for Characterizing Colloids Particle Sizing, Zeta Potential, Rheology, M.-R. Mbius, D., Ed. Elsiver, 2002.
- [7] R. Hickling and M. S. Plesset, "Collapse and rebound of a spherical bubble in water," *The Physics of Fluids*, vol. 7, pp. 7–14, 1964.
- [8] E. A. Neppiras, "Acoustic cavitation," Physics Reports, vol. 61, pp. 159–251, 1980.
- [9] R. Mettin, Bubble and particle dynamics in acoustic fields: Modern trends and applications. Research Signpost, Kerala, India, 2005, ch. Bubble structures in acoustic cavitation, pp. 1–36.
- [10] K. Mitropetros, H. Hieronymus, and J. Steinbach, "Single bubble ignition after shock wave impact," *Chemical Engineering Science*, vol. 61, pp. 397–416, 2006.
- [11] Q. V. Nguyen and D. A. Jacqmin, "A study of cavitation-ignition bubble combustion," NASA Glenn Research Center, Tech. Rep., 2005.
- [12] E. Brandes and W. Möller, Safety Characteristic Data, Volume 1: Flammable Liquids and Gases, 2nd ed. Wirtschaftsverlag NW, 2008.
- [13] M. Hattwig and H.Steen, Handbook of Explosion Prevention and Protection. Wiley-VCH, 2004.
- [14] A. Wähner, G. Gramse, T. Langer, and M. Beyer, "Determination of the minimum ignition energy on the basis of a statistical approach," in *IX International Symposium on Hazard, Prevention and Mitigation of Industrial Explosions*, 2012.
- [15] IEC 62127-1, Ultrasonics- Hydrophones Part 1: Measurement and characterization of medical ultrasonic fields up to 40 MHz, Geneva: International Electrotechnical Commission Publication 62127-1, Std., 2007.