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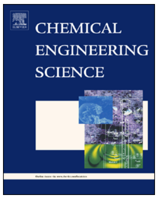

Sono-chemiluminescence (SCL) in a high-pressure double stage homogenization processes

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HIGHLIGHTS

- Hydrodynamic cavitation is investigated via sono-chemiluminescence (*SCL*).
- Second stage delivers back pressure and changes hydrodynamic cavitation.
- Cavitation intensity can be optically measured for different process conditions via *SCL*.
- *SCL* intensity values are in line with former theoretical assumptions.

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ABSTRACT

An applied back-pressure to a high pressure homogenization system influences the development of hydrodynamic cavitation and the collapse of cavitation bubbles. This paper focuses on the effect of back-pressure on hydrodynamic cavitation and how the intensity of collapsing cavitation bubbles can be identified. Based on sono-chemiluminescence (*SCL*), an optical measuring method was developed to detect cavitation induced formation of •OH radicals, which indicates the intensity of collapsing cavities. The results are discussed with the introduced dimensionless *SCL* intensity number $\bar{I}_{SCL}$. The $\bar{I}_{SCL}$ number increases with increasing inlet pressure as well as with increasing back-pressure and confirms therewith theoretical based assumptions by other studies. A maximum $\bar{I}_{SCL}$ at a back-pressure of approximately 30% was found and reduced to an intensified collapse of cavities in or after the first high pressure disruption unit. The maximum in *SCL* intensity is in line with the minimum droplet size of corresponding emulsification experiments.

1. Introduction

To produce dispersed products such as liquid foods, pharmaceuticals, fuels or paintings high pressure homogenization is a widely used technique. Pressure losses up to several 1000 bar over the disruption unit are responsible for elongation, shear and viscous forces in laminar and turbulent flows which disrupt the dispersed droplets. In industrial process a so called 'double stage' process is preferentially used as process intensification. Thereby two disruption units are installed in series in which the second unit realizes a 'back-pressure' p_{bp} towards the first one, while the total pressure loss remains the same compared to the 'single stage' process. The increase in droplet breakup efficiency compared to the single stage process is represented by a reduction of droplet sizes and most probably induced by an alteration of droplet disrupting forces. The resulting droplet sizes decreases at constant inlet pressure p_{inlet} with increasing p_{bp} until a minimum in droplet size is obtained at back-pressure to inlet pressure ratios (Thoma number *Th*) of 20–30% ($Th=0.2$–0.3) (Mohr, 1987; Freudig, 2004; Finke et al., 2014). In a former study we have shown that the droplet breakup at typical *Th* happens entirely in or after the first disruption unit (Schlender et al., 2015a). In addition to the above mentioned forces, several researchers argue that an intensification of hydrodynamic cavitation is responsible for the intensified droplet breakup (McKillop et al., 1955; Kurzhals, 1977; Treiber, 1979; Aguilar et al., 2004; Finke et al., 2014; Schlender et al., 2015a). In hydrodynamic reactors new approaches show the efficiency of cavitation on the droplet break disruption in venturi valves (Ramisetty et al., 2014). Collapsing cavities induce thereby shock waves and micro-jets that can be disruptive on the dispersed phase. The location of collapsing cavities as well as the intensity of induced shock waves and micro-jets ('cavitation intensity') are crucial for an intensified droplet breakup (Kurzhals, 1977). Until today, the exact location and intensity of cavitation collapses in

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single stage and double stage homogenization processes has just been poorly understood. For single stage processes our research group has recently shown that the development of cavitation pattern downstream of a cylindrical coaxial orifice has an immense influence on the droplet breakup and therefore on the emulsification efficiency (Schlender et al., 2015b). As principle finding the single bubble cavitation (jet-cavitation) is more effective in means of droplet breakup than coherent cavitation pockets (super cavitation or hydraulic flip). Cavitation was thereby characterized by shadow-graphic imaging (Schlender et al., 2015b). In regards to the characterization of cavitation a brief overview will be given of some basic approaches and investigations dealing with the location of cavity collapse and its intensity.

McKillop et al. (1955) stated that the appearance of hydrodynamic cavitation differs heavily while applying back-pressure to a high-pressure unit. In an optical accessible venturi tube he observed a suppression of cavitation bubbles with increasing back-pressure, but he did not make a correlation towards cavitation intensity. Recent investigations with µ-PIV techniques allow a more detailed visualization and location of cavitation pattern in rectangular micro slot orifices (Gothsch et al., 2014) in single and double stage processes. They have shown that hydrodynamic cavitation is quite sensible to low changes in p_{bp} and can be totally suppressed by a p_{bp} of 30–35% in respect to the total pressure loss.

Continuative research used acoustical measurements to indentify characteristic sound intensities induced by collapsing cavities inside high pressure valves (Kurzhals, 1977; Testud et al., 2007; Håkansson et al., 2010; Mizuyama et al., 2010; Nagaya et al., 2011). Amongst others, Håkansson et al. (2010) observed an increase of ultrasound amplitude at frequencies between $12\ \mathrm{kHz} < f < 45\ \mathrm{kHz}$ with increasing p_{bp}. Further, they detected a maximum amplitude at $15\% < p_{bp} < 20\%$ and correlated it with optical measurements obtained by laser scattering in an 55 µm gap valve (Håkansson et al., 2010). The increase in sound intensity was explained with an increase in single bubble cavitation instead of a coherent vapor pocket cavity or the decrease in overall vapor content while applying p_{bp} (Franc and Michel, 2004; Rus et al., 2007).

Another method to indentify cavitation inside an orifice or a pipe is the detection of erosive effects (Benjamin and Ellis, 1966). Innings et al. (2011) investigated for high pressure flat valves, as used in dairy or fruit juice industry, that an increase in p_{bp} is accompanied by an increase of wear in all studied cases as a result of hydrodynamic cavitation. Other research describes a decrease of wear with decreasing surface tension in ultrasound cavitation devices (Iwai and Li, 2003) and explains it with an hypothetically lower 'cavitation intensity'. However, the detection of cavitation induced wear inside the high pressure unit is just a rough estimation on cavitation intensity. Since only cavity collapse at boundaries causes wear, cavities collapsing in the bulk fluid are neglected by this method.

Sonochemistry, chemical reactions induced by cavitation, is a quite common method to study cavitation applications. An extensive review over hydrodynamic cavitation reactors is given by Gogate and Pandit (2001). It is well known that the collapse of cavities induces high local pressure gradients and temperatures, which can result in the homolytic dissociation of water molecules into •OH radicals (Morison and Hutchinson, 2009). Certain additives react with the free radicals and form verifiable compounds. The quantity of these compounds can be associated with the formation of •OH radicals and therewith with the 'cavitation intensity'. Among others, the decomposition of potassium iodide (KI) to the tri-iodide complex (I_3^-), the so called Weissler reaction, is worth mentioning and well known in acoustic cavitation studies. For the Weissler reaction investigations on hydrodynamic cavitation show a characteristic behavior of I_3^- production with increasing 'cavitation intensity' (Suslick et al., 1997; Senthil Kumar et al., 2000; Vichare et al., 2000). Since the process time to receive a measureable amount of tri-iodide complex takes up to 60 min while side reactions cannot be excluded. Morison and Hutchinson (2009) critically annotated that the Weissler reaction is not suitable for a comparison of hydrodynamic cavitation setups and their parameters. Other possible reactions to detect cavitation are the degradation of Rhodamine B (Mishra and Gogate, 2010) or benzoic acid (Singla et al., 2004) and the salicylic acid dosimetry presented by Amin et al. (2010).

This work uses another sono-chemical reaction to detect intensities of hydrodynamic cavitation. While adding Luminol (5-amino-2,3-dihydro-1,4-phthalazinedione) to the processed fluid, sono-chemiluminescence (*SCL*), the emission of 'cold light', can be optically detected in the region of cavitation bubble collapse (Thiemann, 2011). Up to now, the indirect method of cavitation detection via *SCL* is mainly used for cavitation detection in ultrasonic devices. While cavity collapse takes place, the additive Luminol is oxidized by free •OH radicals to α-hydroxy hydroperoxide (α-HHP). The subsequent decomposition of α-HHP emits light at a wavelength of 425 nm (blue light). The highest light emittance appears instantly. The lifetime of light emittance lower intensities is reported up to several milliseconds. Detailed schematic reaction pathways of Luminol in cavitating fluids are given in the work of Rose and Waite (2001). The reported quantum yield of Luminol assisted *SCL* is differs between 1% and 2% (Gundermann, 1970).

Since the connection between 'cavitation intensities' in double stage homogenization processes and the resulting droplet size has not been sufficiently investigated yet, this study deals with optical and acoustical characterization methods of hydrodynamic cavitation downstream of orifices and corresponding emulsification results. By the use of an optical accessible orifice outlet channel, the region where droplet breakup in high pressure systems most likely occurs will be visualized (Galinat et al., 2005; Kelemen, 2014). This method was already applied in former studies (Schlender et al., 2015a, 2015b). Since Luminol is used as a *SCL* reactant, this work investigates if the detection of hydrodynamic cavitation is reasonable with this optical method. A suggestion for a dimensionless cavitation intensity number (I_{SCL}) based on *SCL* light emission is presented. Further inquiries regarding the location of cavitation appearance and on the variation in *SCL* intensity during the influence of p_{bp}. The results from I_{SCL} measurements are compared to those received by acoustical cavitation detection ($I_{acoustic}$). Finally, the resulting *SCL* intensities are discussed with corresponding emulsification results.

2. Calculations

The probability of cavitation appearance, represented by the ratio of local to hydrodynamic pressure, was calculated by the cavitation number σ (Yan and Thorpe, 1990; Brennen, 2005).

$$\sigma = \frac{p_{downstream} - p_{vapor}(T_1)}{0.5 \cdot \rho_l \cdot \overline{u}^2_{orifice}} \qquad (1)$$

Used pressures are the static pressure downstream of the orifice $p_{downstream}$ and the vapor pressure of the used liquid p_{vapor}. In back-pressure experiments $p_{downstream}$ describes the pressure measured between after the first homogenization unit (p_{bp}). ρ_l represents the density of the liquid at experimental conditions. Since the mass flow rate can be accurately measured, the fluid mean velocity $\overline{u}_{orifice}$ in the orifice throat $d_{orifice}$ can be calculated. For single stage processes the atmospheric pressure is used for $p_{downstream}$. In theory cavitation can start for $\sigma < 1$ and below due to