Delfim-Soares explicit time marching method for modelling of ultrasonic wave in microalgae pre-treatment

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Abstract. Ultrasonic wave is one of the most popular pre-treatment methods of lipid extraction in microalgae, due to its low energy supply requirement, eco-friendliness and excellent cell disruption capability. Although quite some number of experimental works were reported, the numerical modelling of the ultrasonic wave is limited so far, as to the knowledge of authors. The modelling is required for a more robust pre-treatment optimisation. Therefore, in current work, the numerical model of ultrasonic wave and its cavitation has been developed using the Delfim-Soares explicit time marching method (DSETM), which is proposed in recent years to solve structural vibration problem. The two dimensional wave equation in ultrasonic scale has been solved with the frequency of 20 kHz, 40 kHz and 60 Hz. Moreover, Rayleigh-Plesset equation is solved using the same method too to predict the growth of the radius of bubble due to different initial radius. It is found that higher wave frequency will not improve the speed of cavitation, but instead it can decrease the wavelength to increase the possibility of cavitation process occurrence in enhancing the pre-treatment efficiency.

1. Introduction

Microalgae are widely recognised with their high biomass production rate and energy efficiency among natural crops [1,2], and therefore they have been deemed as a promising source of renewable energy to substitute the fossil fuel [3]. The techniques of conversion of microalgae into biofuels can be found in many literatures [4–13] and prior to these biomass conversion, the extraction of lipids by breaking the lignin and hemicellulose structure is required [13,14]. In many conventional ways of lipids extraction (pre-treatment) such as mechanical milling [15], extrusion [16], acid/alkaline [17] and organosolv [18], high costing, environmental hazards and undesirable compounds formation appeared as their major disadvantages [13]. Ultrasonic irradiation is therefore one of the emerging pre-treatment technologies, which is eco-friendly and powerful in improving hydrolysis efficiency [19].

Ultrasonic wave is the acoustic wave with frequency ranging from 20 kHz and above, in which its compression and rarefactions will create acoustic bubbles. The bubbles will expand over time, and the void structure will implode as shock wave when critical bubble radius is developed. Such phenomenon is known as cavitation [20,21], and it will lead to temperature and pressure hike high enough to disrupt the lignocellulosic structure. The cavitation process can be illustrated in Figure 1. The point in which
the implosion occurs is named as hotspot. The details of the pre-treatment process due to ultrasonic wave can be further found in several archives [13,19–23].

Quite a number of works have been investigating on the production of biomass from different microalgae using ultrasonic method [24–28]. There are many other factors influencing the efficiency of ultrasonic pre-treatment such as biomass concentration, sonication intensity and power were reported [29–31]. However, most of the biomass investigations are limited to experimental works. The high cost and limited-variable range of experimental works will retard an in-depth study in microalgal pre-treatment. With this regards, the deployment of numerical mathematics for ultrasonic wave prediction could be an important alternative.

Nonetheless, the application of computational works in ultrasonic irradiation is in its infant stage. To the knowledge of authors, only Smithmaitrie and Tangudomkit [32] and Lais et al. [33] reported on the computational works in solving the fundamental ultrasonic equation with the assistance of finite element commercial software COMSOL. Moreover, the modelling of the ultrasonic cavitation is not available yet so far [33]. The very limited numerical works available in ultrasonic wave and cavitation have hindered further research on the computational prediction of pre-treatment efficiency and investigation on working principle of sonication. To fill the gap, Delfim-Soares explicit time marching (DSETM) Method will be applied in the current study. DSETM is proposed recently by Prof Delfrim Soares [34,35] to solve the structural mechanics problem, and now the method is being transplanted in current work to solve other hyperbolic equations in ultrasonic pre-treatment. Indeed, the method is simpler to be implemented compared with conventional time discretisation as used by COMSOL for hyperbolic equation.

Figure 1. Cavitation process due to ultrasonic wave [21].

2. Mathematical modelling for ultrasonic wave equation
The modelling of two-dimensional ultrasonic wave is governed by the pressure acoustic equation which can be written as in Equation (1) [32,33,36]:

\[
\frac{1}{\rho c^2} \frac{\partial^2 P_t}{\partial t^2} - \frac{1}{\rho} \left[ \frac{\partial^2 P_t}{\partial x^2} - \frac{\partial q_d}{\partial x} \right] + \frac{\partial^2 P_t}{\partial y^2} \frac{\partial q_d}{\partial y} = Q
\]

(1)

where \( \rho \) is the fluid density \([\text{kg/m}^3]\), \( c \) is the speed of sound \([\text{m/s}]\), \( \rho c^2 \) is the fluid bulk modulus \([\text{kg/(ms}^2)]\), \( P_t \) is the total acoustic pressure \([\text{kg/(ms}^2)]\), \( q_d \) is the dipole source \([\text{kg/(m}^2\text{s}^2)]\), \( Q \) is the monopole source \([1/\text{s}^2]\), \( t \) is time \([\text{s}]\) while \( x \) and \( y \) represents the spatial coordinates \([\text{m}]\). Both monopole and dipole source are the directed acoustical sources [37], which influence the acoustic pressure at the far field as defined as in Equation (2) and (3) respectively:

\[
P(r) = Q \frac{\rho c}{4\pi r^2} \lambda
\]

(2)

\[
P(r) = q_d \frac{\rho c d \cos \theta}{4\pi r^2} \lambda^2
\]

(3)

in which \( r \), \( \lambda \), \( d \) and \( \theta \) is the radius from the source of the acoustic vibration \([\text{m}]\), wave length \([\text{m}]\), distance between the acoustical sources \([\text{m}]\) and angle between the acoustical sources respectively.
Equation (1) can be simplified into a simple wave equation by omitting the acoustical sources, as expressed in Equation (4).

\[
\frac{\partial^2 P}{\partial t^2} = c^2 \left( \frac{\partial^2 P}{\partial x^2} + \frac{\partial^2 P}{\partial y^2} \right)
\]  

(4)

The pressure compression and rarefaction will lead to acoustic cavitation and formation of bubbles. Therefore, upon numerical solution on Equation (1) or (4), the pressure field need to be corresponded with the bubble radius, which can be described by Rayleigh-Plesset equation [33,38] as in Equation (5):

\[
\frac{P(t) - P_\infty(t)}{\rho} = R \frac{\partial^2 R}{\partial t^2} + \frac{3}{2} \left( \frac{\partial R}{\partial t} \right)^2 + \frac{4\nu}{R} \frac{\partial R}{\partial t} + \frac{2\sigma_s}{\rho R}
\]

(5)

where \(P_\infty\), \(R\), \(\nu\) and \(\sigma_s\) represents the atmospheric pressure [kg/(ms^2)], bubble radius [m], kinematic viscosity of the fluid [m^2/s] and surface tension of the fluid [kg/s^2] respectively. The bubble radius will expand over time, and by reaching the critical radius, the bubble will collapse and release energy for cell wall disruption. Critical radius \(R_{cr}\) [39] can be expressed as in Equation (6):

\[
R_{cr} = \frac{9mBT}{8\pi\sigma_s}
\]

(6)

where \(m\), \(B\), and \(T\) is mass of gas in the bubble [kg], specific gas constant [J/(mol.K)] and absolute temperature [K] respectively. Equation (6) needs be modified into Equation (7), by assuming the geometry of the bubble as sphere:

\[
R_{cr} = \frac{9B_aT}{8\pi\sigma_s} \left( \frac{4}{3} \pi R_{cr}^3 \right) \iff R_{cr} = \frac{2\sigma_s}{3\rho_a B_a T}
\]

(7)

where \(\rho_a\) is the density of air [kg/m^3]. Upon explosion of bubble, the cavitation process will restart. By solving Equation (4) and (5), the contour of transient acoustic pressure and plot of bubble radius growth can be computed. The general numerical algorithm can be illustration in Figure 2.

However, the time required for the radius to reach its critical value is highly dependent on the initial radius [39]. Moreover, Rayleigh-Plesset equation is an initial value problem, but unfortunately there is no mathematical equation available to predict the initialisation of bubble formation. In the work of Chakma and Moholkar [40], the initial radius is prescribed instead of computed. The assumption on the initial radius is required prior to computation.

3. Physical Modelling

In current work, the ultrasonic transducer is located at the middle of the microalgae-fluid domain. The microalgae-fluid domain is set in square shape with area of 0.04 m^2 (20 cm x 20 cm), while the size of the transducer is at the middle of the domain, as shown in Figure 3. The ultrasonic transducer is supplied with the frequency \(f\) of 20 kHz, 40 kHz and 60 kHz. The implementation of the boundary conditions will be further discussed in the next section. Meanwhile the fluid domain properties applied can be summarised in Table 1.

| Fluid Properties                              | Value (Unit) |
|----------------------------------------------|--------------|
| Absolute temperature \(T\)                   | 300 K        |
| Density of microalgae-fluid \(\rho\)         | 1000 kg/m^3  |
| Kinematic viscosity of microalgae-fluid \(\nu\)| \(9 \times 10^{-2}\) m^2/s |
| Surface tension of microalgae-fluid \(\sigma_s\)| \(7 \times 10^{-2}\) kg/s^2 |
| Density of air \(\rho_a\)                    | 1 kg/m^3     |
| Specific gas constant \(B_s\)                | 8.314 J/(mol.K) |
Based on the properties set above, the critical radius \( R_{cr} \) is therefore \( 1.871 \times 10^{-5} \) m or 18.71 µm, according to Equation (7). As soon as the bubble radius develops as big as 18.71 µm, Equation (5) needs to be solved again.

\[
\begin{align*}
\text{Start} \\
\text{Initial values and boundary values} \\
\text{Solve Equation (4)} \\
\text{Solve Equation (5)} \\
\text{Reaching } R_{cr} \text{?} \\
\text{Yes} \\
\text{Yes} \\
\text{End} \\
\text{No} \\
\text{No} \\
\text{No}
\end{align*}
\]

**Figure 2.** Numerical algorithm for computation of ultrasonic wave and cavitation.

**Figure 3.** Physical modelling of problem domain.

4. Numerical Modelling

Time marching scheme for hyperbolic equation plays an important role in computational physics as the acceleration term always lead to numerical instability. Most of the time marching scheme applied is the central differencing time marching (CDTM), but the scheme requires three storage of memory in every iteration, i.e. the previous, current and future field variables. There are works [41,42] which applied the higher order time marching scheme, nonetheless, this will aggravate the complexity in the initialisation of computation. The complete review can be found also in the work of Tamma et al. [43].
To mitigate the issue, Soares proposed a novel explicit time marching scheme, \cite{34,35}, in which the current field variable is the only criteria to initial the hyperbolic computation. The method is simple to execute. Time integration of Equation (4) is:

$$\int_{t}^{t+\Delta t} \frac{\partial^2 P}{\partial t^2} dt = \int_{t}^{t+\Delta t} \frac{\partial^2 P}{\partial x^2} dt + \int_{t}^{t+\Delta t} \frac{\partial^2 P}{\partial y^2} dt$$

which will lead to

$$\left( \frac{\partial P}{\partial t} \right)^{n+1} = \left( \frac{\partial P}{\partial t} \right)^n + \frac{c^2\Delta t}{\Delta x^2} \left( P_{i+1,j}^n + P_{i-1,j}^n + P_{i,j+1}^n + P_{i,j-1}^n - 4P_{i,j}^n \right)$$  \hspace{1cm} (8)

$$P_{i,j}^{n+1} = P_{i,j}^n + \beta_1 \left( \frac{\partial P}{\partial t} \right)_{i,j}^n + \beta_2 \left( \frac{\partial P}{\partial t} \right)_{i,j}^{n+1} \Delta t$$  \hspace{1cm} (9)

where $\Delta x$, $\Delta t$ and $n$ is the node spatial distance, field variable time marching interval and current time step respectively, while $\beta_1$ and $\beta_2$ are time marching coefficients \cite{35}. The only variable needed to initiate the computation is the wave excitation at the middle of the domain, which can be described as:

$$P_{1,J} = P_{\text{max}} \sin \left( 2\pi ft \right)$$  \hspace{1cm} (10)

where ($I,J$) is the location of wave excitation while $P_{\text{max}}$ is the amplitude of acoustic pressure. Since the ultrasonic wave emits about 10 dB of sound pressure level, the corresponding $P_{\text{max}}$ is 3.1623 $\mu$Pa.

The non-reflecting boundary condition is applied. Now the DSETM approximation of Equation (5) is:

$$\left( \frac{\partial R}{\partial t} \right)^{n+1} = \left( \frac{\partial R}{\partial t} \right)^n - \left( R^n \right)^2 \left[ 1.5 \left( R^n \right)^2 + 4\nu \frac{\partial R}{\partial t} + \frac{2\sigma_s + \left( R^n \right)(P_{\infty}(t) - P(t))}{\rho} \right] = 0$$  \hspace{1cm} (11)

$$R^{n+1} = R^n + \Delta \left( \frac{\partial R}{\partial t} \right)^{n+1}$$  \hspace{1cm} (12)

During the computation of Equation (4) and (5), the Courant Number $C$ as defined in Equation (13), must be controlled within 1 to ensure numerical stability during time marching. In the current work, the Courant number applied is 0.5 to ensure the fulfilment of Courant-Friedrichs-Lewy (CFL) condition \cite{44,45}.

$$C = \frac{c\Delta t}{\Delta x} \leftrightarrow \Delta t = 0.5 \frac{\Delta x}{c}$$  \hspace{1cm} (13)

Note that the time step shall be sufficient to support the ultrasonic frequency, i.e. $\Delta t < 1/f$. Anyhow, Eq. (13) is ample to ensure time-marching stability indeed as $0.5\Delta x/c^2$ is always larger then $1/f$. Meanwhile for the simulation on Rayleigh-Plesset equation is conducted with initial radius of 1.8, 2.5, 3.4, 4.2 and 5.0 $\mu$m \cite{40}.

5. **Numerical Verification**

The numerical verification is done by comparing the results obtained for one-dimensional wave equation using DSETM and CDTM. By setting the initial and boundary conditions as described in Section 4, both DSETM and CDTM will produce the similar sinusoid curve as shown in Figure 4. Moreover, the wavelength of the wave is in accordance with the established relationship \cite{46} between the wavelength, frequency and speed of sound, i.e. for frequency of 20 Hz, the wave length, $\lambda$, is:

$$c = f \lambda \leftrightarrow \lambda = \frac{c}{f} = 0.0172m = 0.172 \times 10^5 \mu m$$  \hspace{1cm} (14)
Figure 4. Sinusoid curve produced by DSETM and CDTM for one-dimensional wave equation.

6. Results and Discussion
There are three possible ways in which the ultrasonic wave improves the pre-treatment: the initiation of cavitation, initial radius of acoustic bubble and its growth rate. The effect of wave frequency to these factors will be investigated.

The effect of frequency to initiation of cavitation can be studied via the modelling of ultrasonic wave produced by the excitation with different frequencies. Transducer frequency of 20 kHz, 40 kHz and 60 kHz within the time of 500 µs can be observed as in Figures 5 – 7 respectively. The higher the frequency, the smaller the wavelength. With the reduction of wavelength, acoustic pressure fluctuates at almost every location of domain within the same time frame. This will increase the possibility to implode the cavitation bubbles as the implosion is a direct outcome of pressure compression [47].

Figure 5. Development of ultrasonic wave of 20 kHz at the time of (a) 200 µs and (b) 500 µs.

Figure 6. Development of ultrasonic wave of 40 kHz at the time of (a) 200 µs and (b) 500 µs.
Figure 7. Development of ultrasonic wave of 60 kHz at the time of (a) 200 µs and (b) 500 µs.

The growth of radius of the cavitation bubble due to different initial radius has also been computed in Figure 8. The time required for bubble collapse increases with the radius of initial bubble formed. It can be observed that during the radius expansion, there are two patterns along the way: (a) the radius grows exponentially with time in general; (b) the radius expands and shrinks alternatively during the general exponential growth due to the pressure fluctuation. It can be clearly shown that the larger initial bubble will shorten the time for implosion. However, the formation of initial radius somehow is beyond the control of experiments [47]. Figure 8 can complement the unknown curve of radius expansion as in Figure 1.

The successful implementation of the DSETM Method to solve the wave equation lays a cornerstone for future manipulation of more variables to model the ultrasonic wave. This may include the effects of increment of pressure amplitude, interference modelling (coherent or non-coherent source) and various possible wall boundary condition and geometry.

However, the frequency does not influence the speed of growth of the bubble radius as shown in Table 2. The growth of radius is more dependent on other fluid properties. Ultrasonic wave is just a sparking factor to initiate the acoustic cavitation. However, it is noteworthy that frequency plays its role in such a way that: the higher frequency will enable more “dormant” bubbles to be geared towards implosion and energy release.

Figure 8. The computation radius expansion using DSETM at different initial bubble radius.
Table 2: Implosion time required for acoustic bubble with initial radius of 5 µm.

| Frequency (kHz) | Time required for implosion (µs) |
|----------------|---------------------------------|
| 20             | 233.7228                         |
| 40             | 233.7228                         |
| 60             | 233.7228                         |

7. Conclusion
In conclusion, DSETM scheme has been extended from structural mechanics for application in computation of ultrasonic wave and acoustic cavitation. It is found that the high frequency does not increase the speed of growth of acoustic bubbles, but it does enhance the possibility for the initialisation of cavitation process. This study has paved a computational basis for a more complex modelling in ultrasonic wave and cavitation. Nevertheless, the computational works is unable to predict the hotspot formation and location, due to the lack of mathematical description on the spatial information of acoustic cavitation occurrence. This could be a potential area for further investigation as well.

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