Proceedings of 20th International Congress on Acoustics, ICA 2010

23-27 August 2010, Sydney, Australia

Transient Light Emission Measurement of Pulse Burst Sonochemical Luminescence

Yanagida, Hirotaka (1), Guo, Shuqiang (1), Fan, Honghui (1), Saitoh, Tadashi (1),

Masanaga, Ikegami (2), Takahashi, Tatsuhisa (2)

(1) Yamagata University, Jo-nan 4-3-16 Yonezawa 992-8510, Japan
(2) Asahikawa Medical College, Midorigaoka Higashi 2-1-1-1 Asahikawa 078-8510, Japan

PACS: 43.35.VZ; 43.25.ZX

ABSTRACT

The light emission measurement of sonochemical luminescence using an aqueous solution of luminol (5-amino-1,2,3,4-tetrahydrophthalazine-1,4-dione) is thought to be useful as a method to predict multi-bubble cavitation behavior because the time from the formation of active bubbles to light emission is very short compared to other methods using a chemical reaction. From this light emission phenomenon, we intended to predict the process of growth and dissipation of cavitation bubbles. To ascertain the optimum sample conditions for our system, we first measured luminescence intensity while changing the luminol concentration, sodium carbonate concentration and liquid temperature. The intensity of luminescence was highest when the sodium carbonate concentration was 450 mM. When the sample temperature was varied from 3 to 50 degrees, the intensity of luminescence was highest between 20 and 30°C. These were taken as the optimum conditions for the system and an experiment was conducted using pulse burst waves. In the experiment, the pulse duration time was set in the range from 0.3 to 8.0 milliseconds and the interval duration time was set in the range from 0.01 milliseconds to 10 seconds to measure the intensity of luminescence using pulse burst waves. It was ascertained that, for a pulse duration time of 4.0 milliseconds or less, the intensity of luminescence decreases to about 90 percent even if the pulse train has a duty ratio of 1:0.02, which is very close to a continuous wave. It was found that the interval duration time in such a state is fixed to 0.1 milliseconds regardless of pulse duration time. Based on these results, we identified the time of activation (the time taken by bubbles to become active during ultrasound irradiation) and the time of deactivation (the time taken by the activity to decay when the irradiation was stopped).

INTRODUCTION

Since the discovery of the excitation of chemical reactions by ultrasound in liquid, it has been expected that this would be used for applications such as cancer therapy [1] and the decomposition of industrial effluent [2]. A reason why these examples have never been applied practically is the poor energy efficiency and difficulty in control. For the application to cancer therapy, the spot of reaction and reaction time must be controlled, so it is necessary to elucidate the conditions for activating cavitation bubbles and the conditions for improving the efficiency in an experimental way. If ultrasound is to be used for diagnosis, irradiation must take place under conditions that cause no reaction. Elucidating the ultrasound irradiation conditions for the formation of active bubbles is important for developing a safe ultrasound diagnostic instrument. Henglein et al. have determined the conditions for the formation of active bubbles through experiments using pulse burst waves [3]–[7]. In their transient response measuring experiment using luminol (5-amino-1, 2, 3, 4-tetrahydrophthalazine-1, 4-dione), they use two pulse duration times (T_{on}) of 2 and 10 milliseconds and 5 interval duration times (T_{off}) in the range from 2 to 100 milliseconds. In the measurement of steady-state luminescence, they measured at various duty ratios ($R=T_{on}/(T_{on}+T_{off})$ from 1:1 to 1:500 in detail [3]. They conducted an experiment at T_{on} of 2 milliseconds that indicated the characteristic time (time of deactivation) of active bubbles to become inactive to be 20 milliseconds or less. In an experiment at 10 milliseconds, on the other hand, the characteristic time increased to 1 second or more [3]. For the active bubbles to become inactive, they presumably must float to the surface and leave the water or dissolve into the water. In either case, the rate of disappearance depends on the bubble size [8]. They ascribe the reason why the time of deactivation is different for different lengths of Ton to the fact that the size of active bubbles depends on T_{on} [3]. We think their outcome of identifying the conditions for forming active bubbles is very meaningful for finding the conditions for using ultrasound for medical diagnosis in a safe way.

In recent years, a study has begun for the purpose of causing a sonochemical reaction efficiently without having to sacrifice energy efficiency [9]. For a sonochemical reaction to occur with high efficiency, it is necessary to clarify the mechanisms that determine the times of activation and deactivation of cavitation bubbles. To find the conditions under which the activation efficiency of cavitation is not inferior to a continuous wave, we undertook a steady-state luminescence measurement of an aqueous solution of luminol at a higher duty ratio, R, of 1:1 to 1:0.05 than the experiment by Henglein et al. Based on the results of this experiment, a model was created for the light emission of sonochemical luminescence using pulsed burst waves. The model was created by finding the degrees of activation and deactivation and considering these parameters.

EXPERIMENTAL PROCEEDURE

The same ultrasound irradiation cell as shown in Figure 1 of Reference [10] was used. The photomultiplier R464 made by Hamamatsu Photonics was placed 2 mm away from the cooling jacket to measure the amount of light of the sonochemical luminescence. For the measurement, the sampling rate of the photon counter was set to 20 Hz for steady-state luminescence measurement and 20 kHz for transient luminescence measurement. The element was driven at an ultrasound frequency set to 1 MHz and voltage set to 100 V. For waveform generation, two instruments were used: the Pulse Function Generator Model 8116 made by Hewlett-Packard and the Arbitrary Wave Generator AG4100 made by Yokogawa. For power amplification, the Model 4020 made by NF was used. As a hydrophone probe to measure ultrasound waveform, the MH28-10 made by Force Institute was used, and the waveform was observed on an oscilloscope DL1740 made by Yokogawa Electric. For pure water, luminol and sodium carbonate, the products by Wako Chemical were used. Luminol was prepared at concentrations of 0.03 to 0.91 mM and sodium carbonate at 45 to 900 mM. Circulating water for sample temperature control was adjusted in the range from 3 to 29°C. The experiment was performed at normal temperature, normal pressure and in an air atmosphere. The sample volume was adjusted to 10, 15 and 30 ml, which corresponded to liquid levels of 30, 45 and 90 mm from the bottom, respectively.

For the experiment, the pulse duration time and interval duration time of pulse burst waves were adjusted in the range from 0.1 to 8.0 milliseconds and from 0.1×10^{-1} to 2.0 milliseconds respectively.

RESULTS AND DISCUSSION

OPTIMUM SAMPLE CONDITIONS

Figures 1-a to 1-d show the time dependence of luminescence intensity at luminol concentrations of 0.03 to 0.46 mM. In this measurement, the sodium carbonate concentration was varied from the lowest 45 mM in Figure 1-a to the highest 900 mM in Figure 1-c. In all cases represented by Figure 1, the luminescence intensity decreases rapidly when the irradiation time exceeds 200 seconds if the luminol concentration is 0.11 mM or less. We attribute this decrease to the insufficiency of luminol that can react with the OH radicals formed by ultrasound irradiation or their adducts. If luminol is increased and its concentration exceeds 0.23 mM, no extreme decrease in luminescence intensity was seen even after ultrasound irradiation for 10 minutes in any of the cases from (a) to (c). At concentrations of 0.23 mM or more, the intensity of luminescence reaches its maximum in the first 50 seconds and decreases rapidly afterwards. After 200 seconds, it settles to a constant value and then decreases slowly. At concentrations of 0.11 mM or less where luminol is insufficient, the decrease in luminescence intensity does not become slow even after 200 seconds and the intensity becomes nearly zero. This decrease to 200 seconds is attributable to the temperature change in the cell. The luminescence intensity was the highest at a sodium carbonate concentration of 450 mM for both luminol concentrations. Thus, the optimum concentration was taken at 0.46 mM for luminol and 450 mM for sodium carbonate and it was decided that further experiments would all take place at these concentrations. Figure 2 shows the change in time of the solution temperature in the cell. The solution was irradiated with ultrasound for 900 seconds with the initial solution temperature set at 3 to 29 degrees. At whatever temperature the irradiation was started, a temperature rise of about 6 degrees was seen in the first 200 seconds. This is consistent with the 200 seconds of notable luminescence intensity change on the order of the time length. Then, luminescence intensity was measured at the various temperatures, the results of which are shown in Figure 3. As the sample temperature is raised from nearly zero degrees, the luminescence intensity takes a maximum at 20 to 30 degrees and decreases again after peaking.

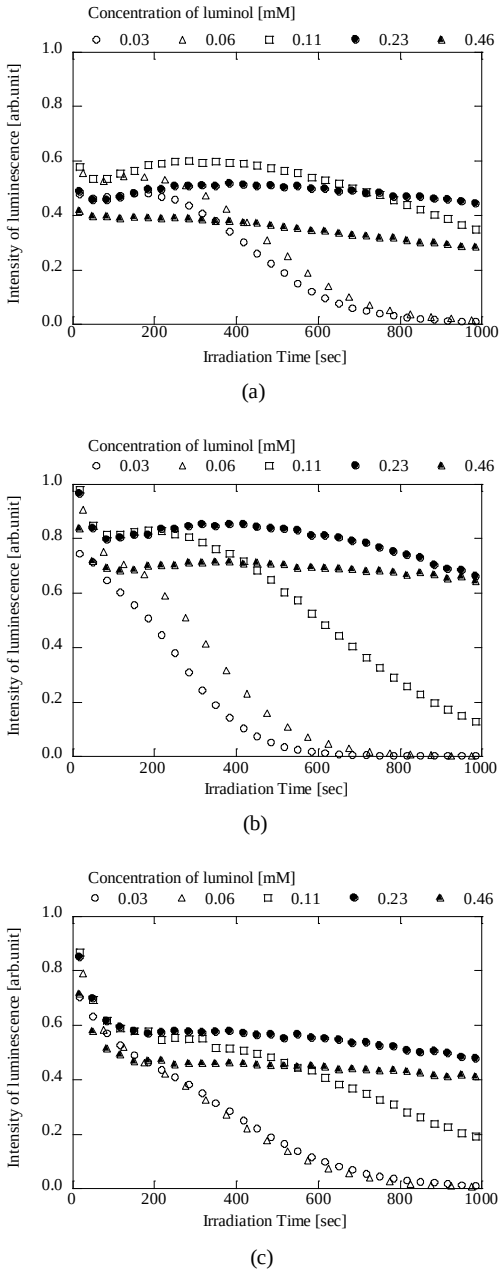


Figure 1: Time development of sonochemical luminescence on various luminol concenration (a)Na_2CO_3=45mM, (b)450mM , (c)900mM.

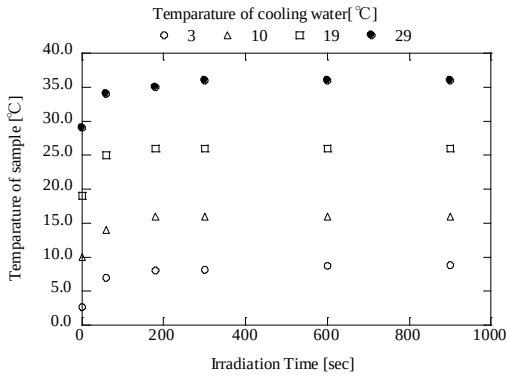


Figure 2: Temparature of luminol solution as a function of irradiation time.

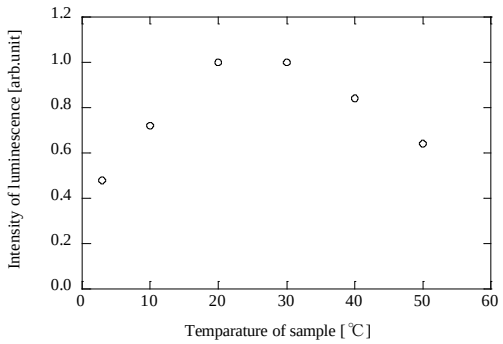


Figure 3: Effect of the sample temparature on the sonochemical luminescence.

Luminol reaction by pulse burst waves Part I (Measurement of steady-state luminescence)

In an experiment using pulse burst waves, a higher duty ratio ($R = T_{on}/T_{on} + T_{off}$) results in a higher total energy if irradiated for the same time length, so the efficiency must be treated as a luminescence intensity per unit energy input. In the steady-state luminescence measurement, the measurement time T_0 is 50 seconds (or a sampling frequency of 20 Hz) and constant. Letting N_0 be the total luminescence intensity during that time length, the luminescence intensity, *N*, per the time ($T_{on} + T_{off}$) of one cycle of the burst is represented by the following equation.

$$N = N_0 \left(T_{on} + T_{off}\right)/T_0 \qquad (1)$$

The luminescence intensity, $\langle n \rangle_{on}$, per the time T_{on} during which ultrasound is emitted can be expressed as:

$$\langle n \rangle_{on} = N/T_{on} \qquad (2)$$

This $\langle n \rangle_{on}$ must be normalized by the luminescence intensity n_c when irradiated with a corresponding continuous wave before a comparison can be made. Figure 4 shows the measurement results of luminescence intensity for T_{on} of 1.0 to 8.0 milliseconds.

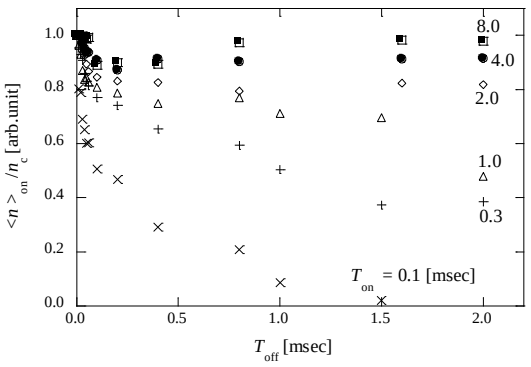


Figure 4: Relative yield of luminescence of luminol as a function of the interval duration time for various pulse duration time.

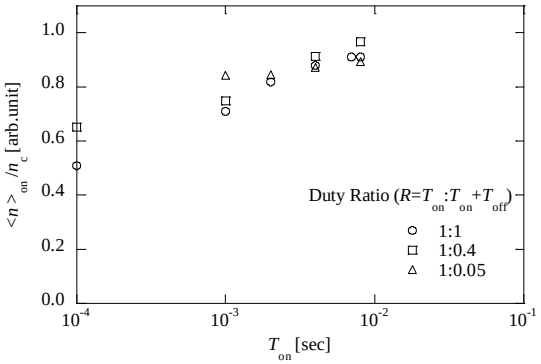


Figure 5: Relative yield of luminescence of luminol as a function of the pulse length for various pulse on/off ratios.

The measurement was performed in the range of T_{off} from zero (C.W.) to 2.0 milliseconds. Whatever T_{on} is, the luminescence decreases rapidly as T_{off} increases from 0 to 0.1 milliseconds. After that, the luminescence becomes steady; however, for short T_{on}, it decreases with increasing T_{off}. For the especially short ON time of T_{on} = 0.1, there is no steady state and the luminescence continues to decrease, though a point of inflection is seen near 0.1 milliseconds. Out of these results, data was taken that could be used for plotting in the same way as Figure 8 of Reference [3] by Henglein et al., which was the plotted in Figure 5. The experiments by Henglein et al. and us differ in ultrasound frequency, element size and other ultrasound irradiation conditions as well as luminol concentration, sample volume and other sample conditions. Despite this, our results for ($R = 1:1$) are similar to the results for ($R = 1:1$) by Henglein et al., and it was found that the values of $\langle n \rangle_{on}/n_c$ lie between 0.7 and 0.9 and they are about the same. Seeing the results for larger *R* of 1:0.4 and 1:0.05 than in the experiment by Henglein et al., the luminescence efficiency was not improved strikingly over the results for 1:1. It can be said from this result that, in an experiment using pulse burst waves, it is impossible to form active bubbles and hold their activity with a high degree of efficiency compared to an experiment using continuous waves. As to the cause of this, we support the prediction

model shown in Figure 1 of Reference [3] by Henglein et al. However, the point of inflection at 0.1 milliseconds that appeared in this experiment cannot be explained by that alone. We thought that a cause of this point of inflection came from the sonochemical reaction vessel. A pulse burst wave consists of repeated pulses and intervals, and it takes time for the ultrasound intensity to reach its maximum from the occurrence of the first pulse. We thought this time lag is a cause. The sound pressure of the pulse burst wave was measured for a time equal to 20 wavelengths (=0.2×10^{-1} milliseconds) and results are shown in Figure 6. It was found from the results that the sound pressure rise at the first T_{on} is sufficiently shorter than 0.1 milliseconds and is about 10 wavelengths (=0.1×10^{-1} milliseconds). Consequently, it can be said that the dull rise of sound pressure is not a cause of the point of inflection appearing at 0.1 milliseconds. Because a chemical reaction by ultrasound takes place with high efficiency in a sound field of standing waves [11], we suspected that the time taken by the sound wave to reciprocate in the sample was in some way related to the 0.1 milliseconds of our experiment. The ultrasound frequency used in the experiment is 1 MHz, so 0.1 milliseconds corresponds to 100 periods. Taking the speed of sound in water at 1500 m/sec, 0.1 milliseconds corresponds to two and half round trips because the liquid level of the reaction vessel is 30 mm. In the experiment, the liquid level was increased by 1.5 and 3 times, and the results are shown in Figure 7. The point of inflection at 0.1 milliseconds did not move even when the liquid level was changed, and all of the levels resulted in about the same value. Consequently, it was found that the rapid efficiency decrease to 0.1 milliseconds is irrelevant to the time taken by ultrasound to fill the cell. Although the cause of the point of inflection appearing at 0.1 milliseconds has not been clarified, it can be said that T_{off} must be set to a far shorter time than 0.1 milliseconds if active bubbles are to be formed and held with as small a sacrifice in energy efficiency as possible.

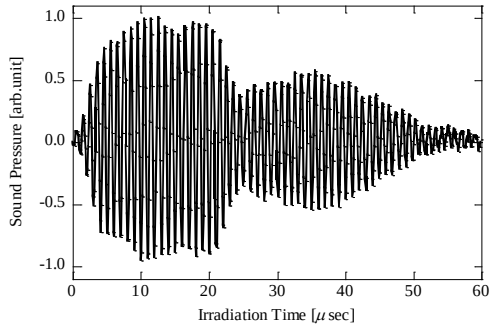


Figure 6: Sound pressure observed by PVDF sensor.

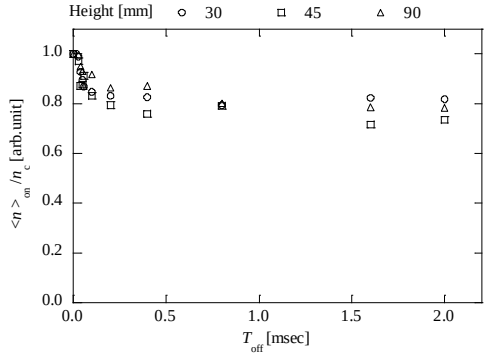


Figure 7: Relative yield of luminescence of luminol as a function of the interval duration.

Luminol reaction by pulse burst waves Part II (Measurement of transient luminescence)

To understand the formation of active bubbles in more detail, we measured transient luminescence. In the transient luminescence measurement, a pulse burst wave was irradiated after a preirradiation for 28 milliseconds as shown in Figure 8. The time from the preirradiation to the start of pulse burst wave irradiation was set to the same time length as T_{off}. Why preirradiation is done is because the luminescent behavior differs according to whether the sample has experienced an ultrasound irradiation or not [3, 6]. In the measurement of transient luminescence, the luminescence sampling rate is high and the luminescence measuring time is short compared to steady-state luminescence, so the initial difference will come out as a large difference. Without preirradiation, an evaluation by transient response measurement cannot be made under the same conditions as steady-state luminescence measurement. Results with T_{off} fixed to 1.6 milliseconds and T_{on} set at 2.0 and 8.0 milliseconds are shown in Figures 9(a) and (b). One will see that luminescence stops at T_{off} and resumes at the next irradiation T_{on}. One will also see that light is emitted during the following irradiations with no less luminescence intensity than by a continuous wave (C.W.) in any case.

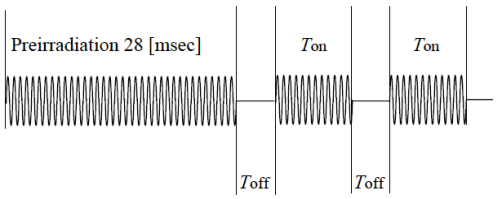
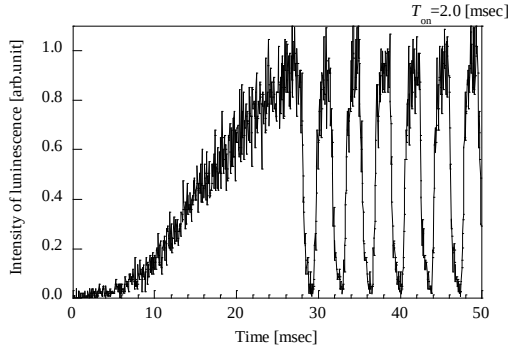


Figure 8: Sound signal for the measurement of the transient luminescence.

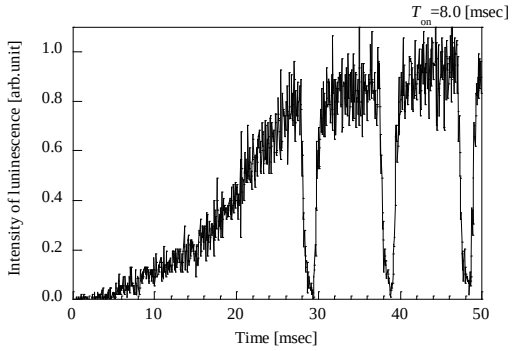
A measurement of transient luminescence was made with T_{on} set at 2, 4 and 8 milliseconds and T_{off} set at 0.4 and 0.8 milliseconds and the change of luminescence over time during one duration of T_{on} is shown in Figures 10(a) and (b). The results for T_{off} = 1.6 milliseconds are taken from part of Figure 9 and plotted here again. Based on these results, we predict how the luminescence that has decreased during T_{off} changes by the next T_{on}, as shown in Figure 11. From this model, the change of luminescence over time during T_{on} can be represented by Equation 3.

$$\frac{n(t)}{n_c} = 1 - \exp\left(-\frac{t - t_0}{\tau}\right) \qquad (3)$$

where $n(t)$ represents the change of luminescence over time, n_c is the value when the luminescence has attained a steady state, τ is the characteristic time of the luminescence to attain the steady state (time of activation), and t_0 is the time at which the luminescence starts. The continuous and broken lines in Figure 10 are the calculation results of curve fitting by this equation.

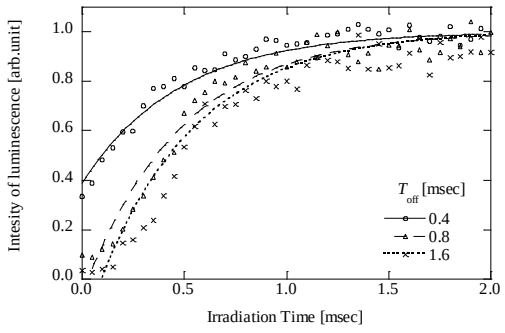


(a)

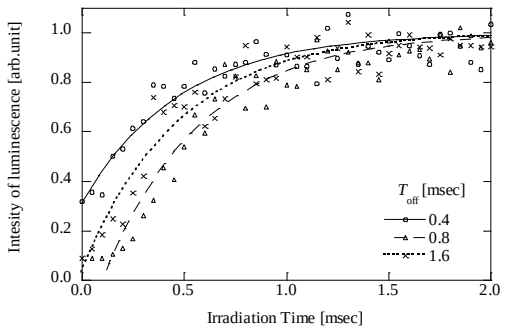


(b)

Figure 9: Relative intensity of luminescence of luminol as a function of the time for various pulse duration time (a) pluse duration time Ton= 2.0 msec and (b) 8.0 msec.



(a)



(b)

Figure 10: Time development of the luminescence during one pulse duration time for the various interval duration time (a) pulse duration time = 2.0 msec and (b) 8.0msec

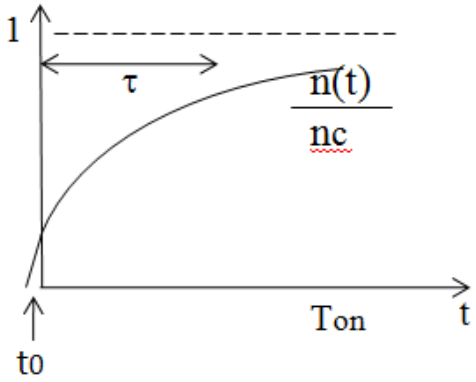


Figure 11: Intensity of luminescence during sonication.

The values of τ are as shown in Figure 12, respectively. For t_0, the results shown in Figure 13 were obtained. τ and t_0 take small values whatever T_{on} is if T_{off} is short, such as 0.4 milliseconds, and the loss of ultrasound energy is small. For T_{off} of 0.8 and 1.6, the value of τ changes little and the mean of τ including the results for T_{off} is 4.8×10^{-1} milliseconds.

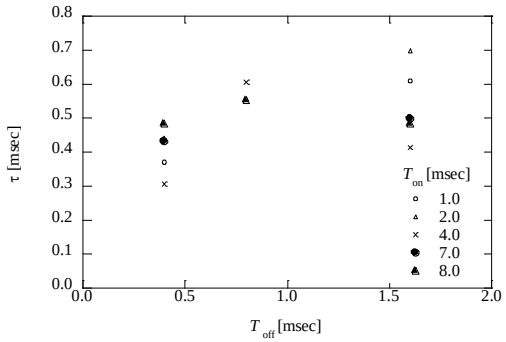


Figure 12: Depnednce of the activation time on the interval duration time for various pulse duration time.

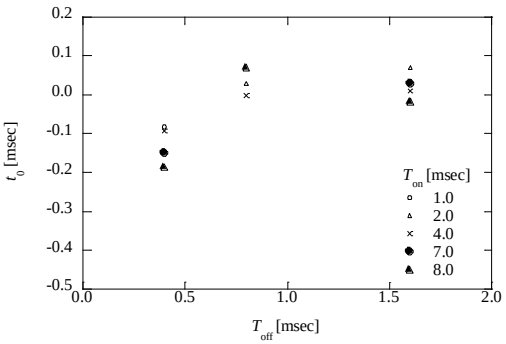


Figure 13: Dependence of the time of the start point of luminescence on the interval duration time for various pulse duration time.

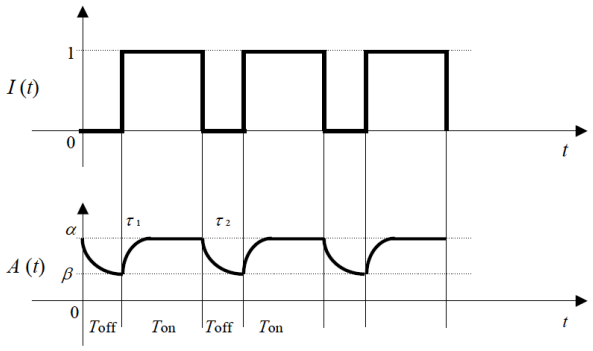


Figure 14: Sound intensity and activation of the bubble.

CREATION AND EVALUATION OF MODEL

To be able to predict the change of luminescence intensity over time from T_{on} and T_{off}, a model was created for the change of luminescence intensity over time by pulse burst waves (figure 14). Letting $A(t)$ be the degree of activation of bubbles, $A(t)$ during ultrasonic irradiation is represented by Equation 4 and $A(t)$ during T_{off} is represented by Equation 5.

$$A(t) = 1 - (1-\alpha)\exp\left(-\frac{t}{\tau_1}\right) \tag{4}$$

$$A(t) = \beta\exp\left(-\frac{t+T_{off}}{\tau_2}\right) \tag{5}$$

where α is the value when the degree of activation is at its minimum and β is the maximum value of the degree of activation. τ_1 is the characteristic time when the degree of activation increases (time of activation) and the same value as τ in Equation 3 is used. τ_2 is the characteristic time when the degree of activation decreases (time of deactivation). Letting $L(t)$ be the number of luminescent luminol molecules, the change of $L(t)$ can be represented by Equation 6.

$$\frac{dL(t)}{dt} = I(t)A(t) - \frac{1}{\tau_2}L(t) \tag{6}$$

where τ_2 represents the time of luminescence to vanish. On the right side, $I(t)$ $A(t)$ is the term to form luminescent luminol molecules and $L(t)$ / τ_2 represents the number of luminol molecules that have become inactive after emitting light. $L(t)$ is expressed as Equation 7.

$$L(t) = \int_{-\infty}^{t} I(t')A(t')\exp\left(-\frac{t+t'}{\tau_2}\right)dt' \tag{7}$$

Luminescence intensity *n(t)* can be expressed as Equation 8.

$$n(t) = \frac{1}{\tau_2}L(t) = \frac{1}{\tau_2}\int_{-\infty}^{t} I(t')A(t')\exp\left(-\frac{t+t'}{\tau_2}\right)dt \tag{8}$$

Calculating the luminescence intensity between time t =0 and time t by Equation 8 gives Equation 9.

$$n_1(t') = \tau_2\left[1-\exp\left(\frac{-t}{\tau_2}\right)\right] - (1-\alpha)\frac{\tau_1\tau_2}{\tau_1-\tau_2}\left[\exp\left(-\frac{t}{\tau_1}\right) - \exp\left(-\frac{t}{\tau_2}\right)\right] \tag{9}$$

where α is the value expressed as Equation 10.

$$\alpha = \frac{1-\exp\left(\frac{T_{on}}{\tau_1}\right)}{\exp\left(\frac{T_{off}}{\tau_2}\right) - \exp\left(-\frac{T_{on}}{\tau_1}\right)} \tag{10}$$

Then, calculating the luminescence intensity between $t = t_0$ and $t = t_1 + T_{on}$ gives Equation 11.

$$n_2(t') = \exp\left(-\frac{t-t_1+T_{on}}{\tau_2}\right)\left[\tau_2\left(1-\exp\left(-\frac{T_{on}}{\tau_2}\right)\right) - \tau_1\tau_2(1-\alpha)\frac{\exp\left(-\frac{T_{on}}{\tau_1}\right) - \exp\left(-\frac{T_{on}}{\tau_2}\right)}{\tau_1-\tau_2}\right] \tag{11}$$

The luminescence intensity to this point of time can be expressed in a similar way, and adding the results gives Equation 12.

$$\sum_{m=1}^{\infty}\int_0^{\infty} n_m(t')dt = \frac{\exp\left(-\frac{t}{\tau_2}\right)\exp\left(-\frac{T_{off}}{\tau_2}\right)}{1-\exp\left(-\frac{T_{on}+T_{off}}{\tau_2}\right)}\left[\tau_2\left(1-\exp\left(-\frac{T_{on}}{\tau_2}\right)\right) - \tau_1\tau_2(1-\alpha)\frac{\exp\left(-\frac{T_{on}}{\tau_1}\right) - \exp\left(-\frac{T_{on}}{\tau_2}\right)}{\tau_1-\tau_2}\right] \tag{12}$$

Consequently, $n(t)$ can be expressed as Equation 13.

$$n(t) = n_1(t) + \frac{\exp\left(-\frac{t+T_{off}}{\tau_2}\right)}{1-\exp\left(-\frac{T_{on}+T_{off}}{\tau_2}\right)}n(T_{on}) \tag{13}$$

The luminescence intensity is expressed as the sum of the luminescence intensity during [illegible] and luminescence intensity during T_{off}. That is, the luminescence intensity can be expressed as Equation 14 during T_{on} and as Equation 15 during T_{off}, respectively.

$$\frac{n(t)}{n_c} = \frac{n_1(t)}{\tau_2} + \frac{\exp\left(-\frac{t+T_{off}}{\tau_2}\right)}{1-\exp\left(-\frac{T_{on}+T_{off}}{\tau_2}\right)}\frac{n_1(T_{on})}{\tau_2} \qquad 0 < t < T_{on} \tag{14}$$

$$\frac{n(t)}{n_c} = \frac{\exp\left(-\frac{t+T_{off}}{\tau_2}\right)}{1-\exp\left(-\frac{T_{on}+T_{off}}{\tau_2}\right)} \frac{n_1(T_{on})}{\tau_2} \qquad -T_{off} < t < 0 \tag{15}$$

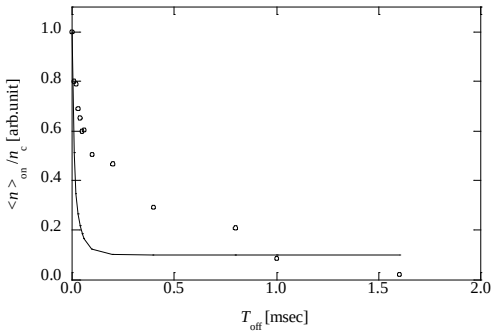
Because the luminescence intensity $< n >_{on} / n_c$ is the integral from - T_{off} to T_{on}, it can be expressed as Equation 16.

$$\frac{\langle n \rangle_{on}}{n_c} = \frac{1}{T_{on}} \int_{-T_{off}}^{T_{on}} \frac{n(t)}{n_c} dt \tag{16}$$

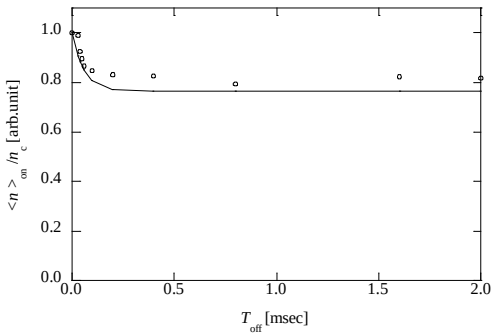
Solving Equation 16 gives Equation 17, which represents the luminescence intensity when irradiated with a pulse burst wave.

$$\frac{\langle n \rangle_{on}}{n_c} = 1 - \frac{\tau_1}{T_{on}} (1-\alpha) \left[1 - \exp\left(-\frac{T_{on}}{\tau_1}\right)\right] \tag{17}$$

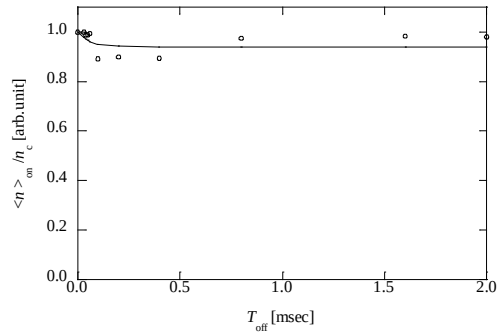
The results of calculation using this equation and the experiment results in Figure 4 are plotted again in Figures 15a to 15c, and the calculated values reproduce the experiment values in most cases. For cases in which T_{on} is short, the model does not reproduce the experiment results, and the reason for that may come from the fact that the degree of deactivation τ_2 during T_{off} differs for different lengths of T_{on} [3]. For T_{off} of 4.0 milliseconds or less and T_{on} of 1.0 millisecond or more in particular, the experiment values are reproduced well, so we think the model to be basically exact.



(a)



(b)



(c)

Fig. 15: Relative yield of luminescence of luminol as a function of the interval duration time for various pulse duration time and estimated results of the model (a) pulse duration time = 0.1 msec (b) 2.0 msec (c) 8.0 msec.

CONCLUSION

In sonochemical luminescence using luminol, the optimum conditions of luminol concentration and sodium carbonate concentration were determined in an experimental way. Using such optimum samples, sonochemical luminescence was produced by a pulse burst wave, and steady-state luminescence and transient response were measured. In the steady response measurement roughly the same results as past reports were obtained, although the conditions were different other than the pulse burst wave. It was found that the luminescent intensity markedly decreases between 0 and 0.1 milliseconds of T_{off}, and it was ascertained that T_{off} must be far shorter than 0.1 milliseconds for luminescence to occur with high efficiency. In the transient response measurement, the change of luminescence over time during T_{on} and T_{off} was measured, and a model was created for the luminescence intensity during each time length. This model reproduces experiment values if T_{off} is 4.0 milliseconds or less and T_{on} is 1.0 millisecond or more.

REFERENCES

[1] S. Umemura, Y. Nagahiko, N. Ryuichiro, *Jpn. J. of Cancer Res.* **84**, pp. 582- 588 (1993)
[2] S. Bennici, A. Gervasini, N. Ravasio, F.Zaccaria, *J. Phys. Chem. B*, **197**, pp. 5168-5176 (2003)
[3] A. Henglein, R. Ulrich and J Lilie, *J. Am. Chem. Soc.* **111**, pp. 1974-1979 (1989)
[4] A. Henglein, Ultrason.: Sonochem. **2**, pp.115-121 (1995)
[5] A. Henglein and M. Guitérrez, *J. Phys. Chem*. **94**, pp.5169-5172 (1990)
[6] A. Henglein and M. Guitérrez, *J. Phys. Chem.* **97**, pp.158-162 (1993)
[7] A. Henglein, D. Herburger, and M. Guitérrez, J.Phys. Chem. Soc. 96, pp. 1126-1130 (1992)
[8] V. A. Shutijov, *Fundamental Physics of Ultrasound*, (Gordon and Breach Science Publichers) (1988)
[9] D. J. Casadonte Jr., M. Flores, C. Petrier, *Ultrason.: Sonochem*. **12**, pp. 147-152 (2005)
[10] H. Yanagida, Y. Masubuchi, K. Minagawa, T. Ogata, J. Takimoto, K. Koyama, *Ultrason.: Sonochem* **5**, pp. 133-139 (1999)
[11] H. Yanagida, Y. Masubuchi, K Minagawa, J.Ttakimoto, K. Koyama, *Jpn. J. Appl. Phys*. **38,** pp. 3103-3104 (1999)