Application of Laser Induced Micro-Blasts for Liquid Disinfection

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Abstract. The present method is based on the bactericidal effect of the microblasts induced by various sources (laser breakdown , electrohydraulic effect...). Using the elaborated conception of physical phenomena providing liquid disinfection it is possible to determine optimal conditions of water treatment. The problem of optimization is solved using methods of mathematical modelling and special experiments. The essence of the method is in complex using of two phenomena. The first one is forming a shock wave that can be induced by laser breakdown. The second effect is forming large cavitation zones which provides disinfection of a treated liquid too. The detailed analysis of wide scale intensity pressure waves induced by different phisycal phenomena affecting various kinds of bacteria, viruses. phages and other microorganisms is carried out. Systematization of these data made it possible to expose that the pressure wave close to the shock wave should be considered as the main destruction factor. Besides, the value of the excess pressure P can be a proper criterion of the microorganism destruction. Easy mathematical formulas to connect the rate of disinfection of the treated water and the rate of the impact are obtained. Such an approach allowed to get the criteria of viability for different kinds of microorganisms. Practical application of the present work is linked with problems of the drinking water disinfection and disinfection of the treated wastewater.

1. Introduction

Ecological and sanitary conditions of water reservoirs and sources of drinking water in the North-West region of Russia are estimated as critical. In particular, a Neva - Neva Bay water system gets not enough treated industrial and municipal sewages in amounts exceeding its self-purifying ability. At present the most popular are biological and reagent means of wastewater treatment with its chlorination. But this way can not provide the effective heavy metal compound purification. Besides, chlorination leads in appearance of harmful chlorine-organic compounds.

Chlorine and ozone liquid disinfection can lead in forming harmful mutagen chlorine-organic compounds (trigalogenmetans) and aldegides. That is why specialists all over the world elaborate new reagentless methods of disinfection.

Analysis of the characteristics of different methods of liquid disinfection shows that UV-radiation, electro-hydraulic effect (EHE) and laser breakdown (LB) are the most perspective physical methods. Expenditure of energy for UV lamps is 0.02-0.1, for EHE is 0.1-10 and for LB 0.03-0.1 Kwt*hour/m^3 respectively. Exploitation costs for UV- radiation and LB are close to chlorination method and are at least 10 times less compared ozonation method.

A micro-blast in liquid is characterized by a local discharge of energy. An energy $E < 5KJ$, an energy density $E/V >$ KJ/cm^3 and relationship $a_0 \Delta t / R_0 \leq 1$ between an energy discharge time Δt and character size of discharge region $R_0 \cong \sqrt[3]{V}$ can be considered as criteria of a micro-blast (a_0 is sound velocity in water). A micro-blast is accompanied by forming a bubble with high temperature and a pressure wave that can convert into a shock. A pressure wave is characterized by a definite rise time till a maximum of pressure ΔP, a shock is characterized by an instantaneous rise of pressure till a front value ΔP_f.

A peak pressure in a shock ΔP_f near the source is about 10^3-10^7MPa for laser breakdown (Rady [1975]), 10^3MPa for chemical explosive (Baum et al. [1980]) and 10^2 MPa for electrical breakdown (Naugolnyh et al. [1975]). Far from the source a shock turns into an acoustic impulse ($\Delta P_f < 1$MPa.)

2. Features of pressure wave impact on microorganisms

In spite of a consideradle amount of publications devoted to exposure which factor among accompanying EHE (pressure wave, cavitation, UV radiation) is prevailing (Zhuk [1978], Edebo et al. [1969], Allen et al.

[1966], Brandt et al. [1962]) the answer has not been found yet. That is why within this study experiments with shocks induced by chemical explosive matter TNT (weight G=1g, energy E=4.2KJ) were carried out.

Samples of waste water in polyethylene round bags were set in water at different distances according to different levels of forcing. The volume of samples was from 0.75 to 3 liters. In this case a zone with pressure about 10MPa that can lead in substantial microorgamisms damage is situated at distance about 1 m from the source. In these conditions such factors as UV- radiation, electro-magnetic and heat impulse can be eliminated.

Results of experiments with TNT are presented in Fig.1, where ΔP_f is a front peak pressure, Nm and Nc are relative changes of a microbe number and Coli-index respectively according to Sanitary Station analysis. A rather big value of presure (10MPa) providing a decrease of microorganisms up to 100-1000 fold the amount can be explained by shock wave front dispersion at polyethylene film (thickness 0.2 mm) and a presence of a thin air layer.

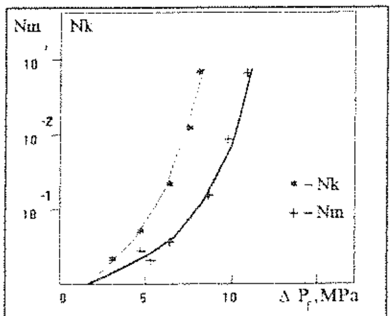

Fig.1. Microbe number Nm and coli-index Nk of experiments with TNT confirm a bactericidal effect dependence on peak pressure ΔP_f.

LB and EHE have small energy discharge compared TNT explosive (0.1-100J) and a zone with the same pressure values is located at distance 0.03-0.3 mm where UV radiation and other accompanying factors are absent due to a strong absorption.

It is well known that microorganisms can survive in conditions of steady pressure up to 100MPa (Sytnik [1982]). Nevertheless low frequency pulsating pressure with frequency 50 Hz and amplitude 6-7MPa during 5-10 seconds has an open bactericidal effect (Landbeck et al. [1963]). Shock wave is the limit case of non-steady regime of pressure wave. It is obviously that such a regime with steep growth of pressure can lead in optimal bactericidal effect.

As an appropriate physical model for a bacteria cell an elastic spherical shell can be suggested. In (Guly G., [1987]) a problem of shock wave diffraction for such a model is considered. It is proved that diffraction processes have a character time about time that shock wave front needs to go a distance of 2-3 diameters of sphere. Further impact on the object proceeds in quasi-stationary regime. So only an initial part of the impulse is bactericidal. A character size of bacteria is 1-10 micron. That is why a mechanical destruction of bacteria can be achieved by using shock waves. Duration of shock wave depends on shock energy and changes from 1 to 20μs when energy is within 0.1-1000J. Space duration of the shock wave in these conditions is 1.5-30mm i.e. up to 1000 times as bigger as character size of the microorganism.

Hence for a shock wave only a front pressure amplitude ΔP_f can serve as a criterion. For a wave with a definite time of growth till maximum of its amplitude a criterion will be a definite integral index depending on maximum of pressure amplitude and gradient of pressure versus time. It is evident that the following condition must be valid: the objects affected by a wave with a "spread" front and pressure ΔP_m will be affected by a shock wave with the same peak pressure $\Delta P_f = \Delta P_m$. Particular values of ΔP_f for different microorganisms can be determined only in experiments.

Well known experimental data on blast disinfection are mainly related to EHE and can not be directly used to determine criterion value of ΔP_m. The cause is in using repeated treatment of large volumes of water. To solve this problem it is necessary to elaborate a model of repeated treatment. Such a model allows to use results of the experiments for criterion determination.

Let us consider a volume V_0 of liquid containing the only type of microorganisms with concentration C_0. The following assumptions are adopted :

- all microorganisms have the same viability to mechanical factors of micro-blast;
- a disinfected volume of liquid after a single treatment has a spherical form;
- a peak pressure is a criterion value;
- after each treatment a volume of liquid is well mixed;

Then concentration of microorganisms can be found from the equation:

$$C/C_0 = (1 - V/V_0)^n$$

where V - disinfected volume of liquid with criterion peak pressure Pm; C - concentration of microorganisms after n micro-blasts.

As it is seen from Fig.2, in variables $\ln C_0/C = n * \lg(1 - V_0/V)$ this relationship grafically represents a straight line. Values of V for each type of microorganisms can be determined using the presented relationship and experimental data on repeated treatment for different number of treatments. It also allows to verify the adopted physical model. Such an approach was applied to analyze results of experiments. Data on these experiments are presented in Table 1. Results of the analysed experiments are shown at Fig.2. A good correspondence of calculations and experiments with different types of microorganisms and different conditions shows that the presented approach is adequate to reality.

3. Theoretical model of LB in liquid

Comparing the characteristics of shocks induced by different sources (laser and electrical breakdown, chemical explosive) is very actual. The main problems connected with this comparison are the following:

- to determine a part of initial energy used to form a wave with required parameters;
- to prove that it is possible to imitate parameters of a micro-blast by another type of microblast.

A simple mathematical model for parameters of pressure waves induced by LB is presented below. The model is aimed at evaluation of source energy convertion into mechanical energy of shock wave and uses results of Witham [1977] . Dispersion and dissipation effects are neglected.

Table 1

Type of micro-organism	Reference	Sign	V/V0	P_m MPa
E - coli	Brandt, 1962	Δ	0.995	3.1
-	-	□	0.995	3.8
-	Allen, 1966	Δ	0.7	3.0
-	Gilliland, 1967	□	0.75	1.9
-	Singh, 1969	○	0.75	3.5
Phages				
T-2	Singh, 1969		0.978	2.8
T-3	-		0.978	2.8
T-4	-	+	0.978	2.8
T-5	-	○	0.990	2.8
CX-174	-	Δ	0.975	2.8
Enterovirus	-	∇		
80/61 Echo	-	□	0.978	2.9
Pig grippe	-	+	0.99	2.9
Bac. subtilis				
Spores	Gilliland, 1967	∇	0.65	2.0
-	Allen, 1966	∇	0.45	3.3

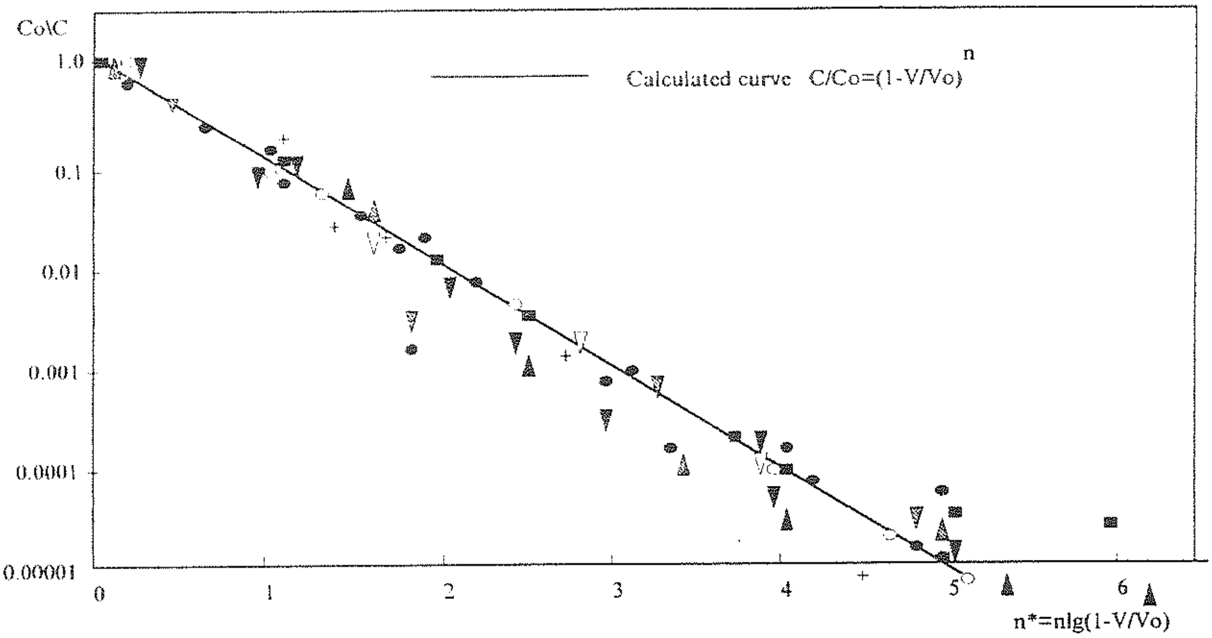

Fig.2 Results of analysis of experimental data .

Let R_0 be a character size of a laser impulse absorption region (R_0=1mm), impulse duration τ=100 ns, impulse energy E=100J, temperature T=10000 K. Then absorption zone is "thermal small" (Rady [1975]) and interaction with surrounding media is limited by processes of pressure and velocity balance at its boundary.

Let the form of the heated volume be a sphere. The process will be described by the following governing equations:

$$\rho_t + u\rho_r + \rho(u_r + 2u/r) = 0$$

$$u_t + uu_r + 1/\rho P_r = 0 \qquad (1)$$

$$P_t + uP_r + a(\rho_t + u\rho_r) = g(t),$$

where r is the radial coordinate, t - time, ρ- density, P - pressure, u - mass velocity, a - sound velocity, g(t) - mass source of thermal energy due to light radiation absorption.

Initial parameters in a motionless liquid at $t=0$ are the following: $P=P_0$, $\rho=\rho_0$,$a=a_0$. Mass velocity u is neglected. At the boundary of the expanding sphere $r=Rg(t)$ pressure and mass velocity are continuous.

The system of equations must also include state equations of liquid $P=P(\rho,T)$ and ionizated steam $P= Pg(\rho,T)$, where T is absolute temperature. The energy at the cross section of the sphere is supposed to convert into a heat in a moment. Value g(t) is determined by the relation:

$$g(t)=3/4\, \alpha\, W(t) / Rg, \ =1/(\rho_0 cv)P_T /\rho=\rho_0, T=T_0, \tag{2}$$

where W(t) is density of energy flux at the boundary of an energy discharge zone; cv - specific heat capacity, coefficient α describes properties of medium. For water according to Kuznetsov state equation (Guly [1987])

$$\alpha= 0{,}1 \mathrm{MPa} / \mathrm{sm}^3 \ \mathrm{s} \ \mathrm{W}. \tag{3}$$

The problem can be simplified if liquid movement is isentropic (Cole [1949]). For water this assumption is valid for pressure up to 1000-2000 MPa. In this case the system (1) comes to:

$$\begin{aligned} &2/(\kappa-1)\, a_t + u a_r + a u_r + au/r = 0 \\ &u_t + u u_r + 2/(\kappa-1)\, a a_r = 0 \end{aligned} \tag{4}$$

where a is defined by $a^2 = dP/d\rho$ and P depends on ρ according to Yakovlev et al. [1972] :

$$(P +B) / (P_0 + B) = (\rho /\rho_0)^{\kappa} \tag{5}$$

where B,κ - constants. For water B=306.5 Mpa, κ=7.15, P_0 , ρ_0 - initial pressure and density.

Hence,

$$(P +B) / (P_0 + B) = (a /a_0)^{2\kappa/(\kappa-1)} \tag{6}$$

Initial conditions for (4) are: $a= a_0$, $u = 0$. For water a_0 = 1500 m/ s. Boundary conditions are the following: at a sphere $r = Rg(t)$, $a=a_0 + ag(t)$, where Δ ag(t)- excess of local sound velocity at the boundary of expanding heated region .

When $r \to \infty$, $a \to a_0$, $u\to 0$.

Introducing a non-linearity (Witham [1977]) the following solution can be derived:

$$\begin{aligned} &a = a_0 + a_0 (\tau)Rg(\tau)/r \\ &u = 2/(\kappa-1) [\Delta ag(\tau)Rg(\tau)/r + a_0 Rg(\tau)/r^2 \int_0^{\tau} \Delta ag(\tau)\, d\tau], \end{aligned} \tag{7}$$

where a parameter τ is determined by the equation:

$$a(t - \tau)/ a_0 = r - Rg(\tau) - \beta \ln\{ [(r/Rg(\tau) + \beta)^2 - \gamma)] / [(1 + \beta)^2 - \gamma)] \} - (\gamma + \beta^2) [F(r/Rg(\tau)) - F(1)] /\sqrt{|\gamma|} \},$$

where

$$F(x) = \begin{cases} 1/2 \ln[(x + \beta - \sqrt{|\gamma|}) / (x + \beta + \sqrt{|\gamma|})], & x> 0 \\ \operatorname{arctg}[(x+\beta) / \sqrt{|\gamma|}| , & x< 0 \end{cases} \tag{8}$$

$$\beta=1/2(\kappa+1)/(\kappa-1)\Delta ag(\tau)/a_0 , \gamma=\beta-2/(\kappa-1)/Rg(\tau)\int_0^{\tau}\Delta ag(\tau)d\tau.$$

The obtained solution is not simple. This lack can be corrected by introducing a shock front. Its propagation is described by the equation (Witham [1977]):

$$dt_f / dr = (a^+ + a^- +u^+ + u^-)^{-1} / 2 \tag{9}$$

Initial conditions: $r = r_0$, $t_f = t_0$, r_0, t_0, - coordinate and time of appearance of a shock front, "+" and"-" denote parameters beyond and in front of a shock front.

Appearance of a shock corresponds to existence of a caustic depending on a parameter τ . It is can be found by calculating the position of the caustic (Korn [1969]). Functions Δag(τ) and Rg(τ), from (7) are determined by a solution of a problem of an expanding gaz bubble, obtained by Gilmore (Cole [1949]):

$$dEg/dt + Pg dVg/dt =g(t) , \tag{10}$$

$Eg=PgVg/(\chi-1)$ is state equation of an ionizated steam. where Eg(t) - inner energy of the bubble, Pg(t) - pressure in the bubble, Vg(t) - volume of the bubble. Function
g(t) is determined by the following relation:

$$g(t) =\pi K\, W(t)\, R_*^{\,2}(t) \ , \ R=\min(Rg,Rs), \tag{11}$$

where Rs - radius of cross-section of a laser beam at the boundary of a bubble; W(t) - power of a laser impulse; K- coefficient of target light absorbtion; χ - index of an adiabate of a steam-gas mixture.In the zone of the first ionization $\chi= 1.26$ (Zeldovich et al. [1966]).

Representation of g(t) in the form (11) allows to consider focusing of the laser beam at the target and non-focused beam flow.

System (10) is supplemented by equations at the boundary between water and steam:

$$dR_g/dt = u_g(t) \quad , \quad P(t) = P_g(t),$$

where velocity of the boundary of the bubble u is determined on the base of (7) when $u = u_g$, $r = R_g(t)$.

$$u_g(t) = 2/(\kappa-1)\,(\,\Delta a_g(t) + a_0/R_g(t) \int_0^{\tau} \Delta a_g(\tau)\, d\tau\,)$$

So the problem of the spreaded bubble is described by a system of the equations:

$$\begin{aligned} &dR_g/dt = u_g, \quad dY_g/dt = \Delta a_g \\ &dP_g/dt = 3/4/\pi(\chi-1)\, g(t)/ R_g^3 - 3\, \chi P_g u_g/ R_g, \\ &u_g = 2/(\kappa-1)\, \Delta a_g + a_0/R_g, \\ &a_g = a_0\{[(P_g+B)/(P_g+B)]^{2\kappa/(\kappa-1)} - 1\}, \end{aligned} \tag{12}$$

where :

Y_g is a subsidiary function with initial conditions at $t = 0$: $P_g = P_0\, a_0$; $R_g = R_0$; $Y_g = 0$.

Let us consider laser radiation absorption on an arbitrary form target. Liquid flow is not already the flow of one dimension, but it can be described within the limits of the hydraulic approach:

$$\begin{aligned} &2/(\kappa-1)\, a_t + u a_r + a u_r + a u\, S'/S = 0 \\ &u_t + u u_r + 2/(\kappa-1)\, a a_r = 0, \end{aligned} \tag{13}$$

where r is a distance along a fixed flow tube; $S(r)$ - cross-section of this tube. $S' = dS/dr$. System of equations (13) is analogous to (10) and turns into (10) when flow has a spherical symmetry. Initial and boundary conditions for these equations are also analogous.

The following assumptions are adopted:

- spreading of the ionizated region is uniform and can be described by a parameter corresponding to a distance from a target;
- a mean square of ray tube is an integral characteristic of a geometric spread of a pressure wave.

Formally this problem is 1 - dimensional problem. A corresponding system of equations for a spreading ionizated region can be written in the form:

Equations for pressure waves:

$$\begin{aligned} &dR_g/dt = u_g\,, \quad dY_g/dt = \Delta a_g. \\ &dP_g/dt = (\chi-1)\, g(t)/ V_g - \chi P_g u_g V_g'/ V_g \\ &u_g = 2/(\kappa-1)\, \Delta a_g + a_0 S_g'/S_g/2, \\ &a_g = a_0\{[(P_g+B)/(P_g+B)]^{2\kappa/(\kappa-1)} - 1\}, \end{aligned} \tag{14}$$

where V_g and S_g - volume and cross-section of the ionizated region, Y_g - subsidiary function, $V_g' = dV_g/dt$
Initial conditions: at $t=0$ $P_g=P_0$; $R_g= R_0$; $Y_g = 0$.

Equation for the spreading ionizated region:

$$\begin{aligned} &\Delta a = \Delta a_g(\tau)\, \sqrt{S_g(\tau)/ S(r)}, \\ &u = 2/(\kappa-1)\sqrt{S_g(\tau)/ S(r)}\, [\, \Delta a_g(\tau) + a_0 S'(r)/S(r) \int_0^{\tau} a_g(\tau)\, d\tau\,], \end{aligned} \tag{15}$$

where τ is determined by the equation:

$$dr/dt = a_0 + \Delta a_g + u\,, \tag{16}$$

with initial condition at $t = \tau$: $r = R_g(\tau)$.

A value $g(t)$ is calculated analogously to (11). Formulae for variables V_g и S_g, depend on initial form of the region of radiation absorption. The most typical is a case of cylindrical region with height H_c and radius R_c. In this case equations come to the following:

$$\begin{aligned} &V_g = \pi\, [H_c\, (\, R_g+R_c\,)^2 + 2\, R_g R_c^2 + 2R_g^2 R_c + 4/3 R_g^3\,] \\ &S_g = \pi\, [\, 2\, H_c\, (R_g + R_c) + 2\, R_c^2 + 4\, R_g^2 + 2\, R_g R_c\,] \end{aligned} \tag{17}$$

Fig.3 illustrates a comparison of calculations of shocks induced by LB, EHE and TNT-explosive with the same stored energy E_0=20 J. Calculations are made for the distance r= 20 sm. In main the EXE model is similar to LB model and is not presented in the paper. The TNT-explosive model is taken from Cole [1949] . Comparison of calculations shows that a laser breakdown and TNT explosive are the most effective. Electrical breakdown does not form a shock and can not provide an intensive pressure wave. Speaking of a peak pressure the electrical breakdown is equivalent to a laser breakdown with energy 0.1 J.

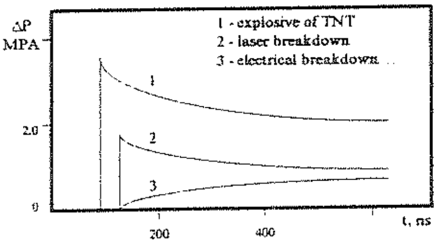

Fig.3 Comparison of calculations by three types of generation with the same stored energy E_0=20 J.

Laser breakdown disinfection experiments were carried out for energy in the impulse about 0.01-0.05 J, duration of impulse 30 ns, wave-length 1.06 micron. Because of small absorption index in liquids for laser wave-lenght a target with strong absoption was used as a source of shock waves. Obtained results allow to pick out parameteres of the target which determine an amplitude in shock wave. These parameteres are absorption index, heat conductivity and size of the target.

Electrical breakdown was used for the following parameteres of discharge electric circiute: voltage 1-5 KV, capacity 0.25-20 microF, induction 1-5 microH.

Samples of liquid were examined in single and multiple treatments. During experiments physical parametres were measured using piezoelectrical and optical methods of registration. Volume of vessels in experiments were from 50 to 500 ml.

Table 2 illustrates results of the single treatment by micro-blast (E=0.03J, volume of the glass vessel 50 and 100 ml) for E-coli. The affected volume was calculated according to the method described above.

Table 2

N^0 of retort	Volume of retort, ml	Amount of colonies before/ after	Target	Affected vol., ml
1a	50	91/87	graphite 6x4x2mm	2.5-1
1б	100	95/91	graphite 6x2x2mm	4.0-1.5
2a	50	107/100	graphite 4x6x4mm	3.5-1.3

Analysis of the obtained results and calculations according to the presented models allows to conclude that a single micro-blast disinfects a volume of liquid if a peak amplitude within a vessel exceeds 3.5MPa. This conclusion is in a good accordance with results of Table 1 comprising data on different publications.

4. Conclusions

Simple mathematical model for shock waves induced by laser breakdown is obtained.

Calculated parameters of shock waves for different sources are compared. It is established that electro-hydraulic breakdown is not effective in forming shock waves, its energy equivalent is about 0.01 - 0.1 % of TNT explosive energy. At the same time the energy equivalent of laser induced micro-blast is about 50 - 100 % of TNT explosive energy.

Experimental study of a bactericidal effect of micro-blast (laser and electrical breakdown, TNT explosive) is carried out. The obtained data are in good concordance with previous investigations and confirm the present method of shock bactericidal effect evaluation. It is proved that the bactericidal effect of pressure wave depends on steep form of its front.

The method of disinfection using power laser impulses is considered as perspective. In particular CO and CO_2 lasers are the most effective for drinking water and sewage disinfection.

Optimal conditions of micro-blast providing a strong bactericidal effect are obtained. A value P_f= 3.5MPa is accepted as a base criterion for liquid disinfection. It corresponds to expenses of light energy of laser about 2 KJ per cubic meter of sewage. An approximate price of lazer induced micro-blast disinfection is estimated as $ 0.025 per cubic meter of liquid.

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