STERILIZATION BY PLASMA PRODUCED AT REB OUTLET INTO ATMOSPHERE

E.M. Babich[2], V.A. Kiselev[1], A.F. Linnik[1], V.I. Maslov[1], N.I. Onishchenko[1], N.I. Sklyar[2], V.V. Uskov[1]
[1]NSC Kharkov Institute of Physics and Technology, Kharkov, Ukraine;
[2]Institute of microbiology and immunology of Mechnikov of Ukrainian AMS, Kharkov, Ukraine

It has been shown, that at REB (4 MeV, 0.5 A, 2 μs, 3 Hz) outlet into atmosphere besides of sterilization straightly by REB electrons, more intense sterilization can be achieved by produced plasma. The sterilization process is determinated by influence of plasma particles, radicals, soft X-ray and UV radiation, and ozone.
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1. INTRODUCTION

At present for cold sterilization and decontamination of the medicinal vegetative raw material and other biological objects instead of toxic gases using the various physical methods are applied more and more: radiation technology (by the relativistic electronic beam (REB) and X-ray radiation [1-3]), processing by plasma of the electric discharge [4, 5] and ozonization. Each of these methods has the certain merits and shortages. Sometimes the combination of these methods results in essential positive results [6]. In particular, REB allows to sterilize the closed objects or medical products of the large thickness (about path length of relativistic electrons in irradiated substance), with larger efficiency, than the gas discharge or ozonization, influences on capsular bacteria and spores (especially at spore density above 10^7 spores/cm^2) [4].

In this paper it has been shown that along with sterilization by relativistic electrons sterilization by plasma produced at REB injection into atmosphere takes place.

2. EXPERIMENTAL ARRANGEMENT

In experiments electron linear resonant accelerator "Almaz - 2" was used to produce REB with parameters: electrons energy is 4 MeV, beam pulsed current is 0.5 A, the duration of a pulse is 2 μs, modulation frequency of a pulse is 2805 MHz (so the pulse consists of $6\cdot10^3$ short bunches of relativistic electrons), the repetitive frequency is 3 Hz. The electron beam was injected from the accelerator into air of atmospheric pressure through a titanium foil of thickness 50 microns. The beam diameter at accelerator exit was equal 0.8 cm. As a result of the passage through the foil angular divergence of the beam electrons appeared that was approximately equal ≈ 10°.

The density of plasma produced at REB outlet into atmospheric air was measured by the special microwave probe [7]. It was shown that the plasma density coincides with one calculated using relationship $n_p \approx 2{,}8\cdot10^{15} j/\beta^2 P$, where j is beam current density, P is neutral gas pressure (in our case it is one atmosphere) [8]. At the distance l=10-12 cm from the exit foil plasma density is equal $n_p \approx 10^{11}$ cm^{-3} and its diameter is equal 10 cm.

Plasma luminescence was investigated with the help of photomultiplier FEU-29, which sensitivity maximum is in the visible light region of the spectrum. It was revealed that duration of plasma luminescence ($\tau_r \approx 5$ μs) is much longer than the duration of the beam current pulse (τ_p=2 μs).

The spatial distribution of soft X-ray radiation, measured by the pinhole camera and the semi-conductor X-ray probe, coincided with area of the maximum plasma luminescence (Fig. 1). Duration of the pulse of this radiation was approximately equal to $0.5\tau_p$. It was observed in second half of pulse of the beam current. The energy of this radiation ($E_v \leq 1$ keV) was determined by measuring decrease of its intensity after passage metal foils of various thicknesses.

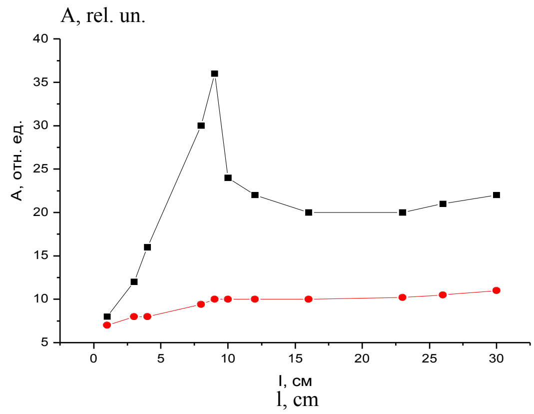

Fig. 1. The spatial distribution of soft X-ray radiations from plasma: 1 - X-ray probe is open, 2 - X-ray probe is covered with a copper foil

In experiments on research of REB interaction with an atmosphere the current of the plasma electrons (a plasma current) was observed both along the direction of the beam movement [9] and perpendicularly to it. The value of plasma current directed perpendicularly to beam movement, and the energy of the plasma electrons were determined with the help of a ring metal collector of diameter 12 cm, inside which the beam passed. The collector located at distance of 10cm from exit foil of the accelerator, registered plasma current, which density is equal 30 μA/cm^2. The energy of these electrons was no more than 12 eV.

3. STERILIZATION EXPERIMENTS

The researches of sterilization efficiency by such plasma were performed together with Mechnikov Institute of microbiology and immunology. In these experiments the glass pans of diameter 100 mm and height 5 mm were arranged so that the pan center was at distance of 10 cm from exit foil of the accelerator. The pan was placed parallel to accelerator axis and its bottom was at distance of 7 cm from this axis so the direct falling of REB electrons on irradiated samples was excluded. (Fig. 2).

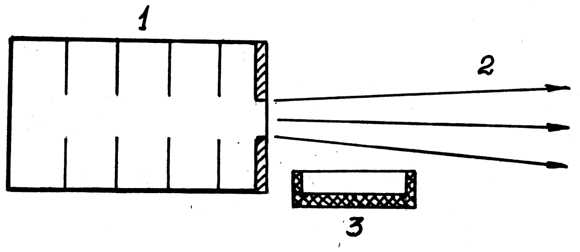

Fig. 2. Scheme of experiment:
1 - accelerator, 2 – electron beam, 3 - pan

On the pans bottom divided into six sectors irradiated bacteria cultures were inoculated on Endo medium. Inoculation was also performed in the control pan and on preliminary irradiated medium without inoculations.

The density of microorganisms in the sectors varied in limits from 10^8 up to 10^2 CFU/ml (CFU – colony - forming units). As irradiated samples cultures E. Coli ATSS #25922, Kebsiella pneumoniae #32, and E. Coli #3 were used. Pans with inoculations were irradiated in two modes: 5 and 10 minutes.

It was revealed that at irradiation during 10 min the growth of bacteria was absent in all sectors (Table 1).

Table 1

Culture E. Coli ATSS #25922 (irradiation during 10 min)		
Control	Irradiated medium	Irradiat. culture
10^7 CFU/ml	10^7 CFU/ml	NG
10^6 CFU/ml	10^6 CFU/ml	NG
10^5 CFU/ml	10^5 CFU/ml	NG
10^4 CFU/ml	10^4 CFU/ml	NG
10^3 CFU/ml	10^3 CFU/ml	NG
10^2 CFU/ml	10^2 CFU/ml	NG

NG means the absence of the bacteria growth (no growth). The irradiation during 5 min resulted in 3-4 orders reduction of microorganism density (Table 2).

Table 2

Culture E. Coli ATSS #25922 (irradiation during 5 min)		
The control	Irradiated medium	Irradiat. culture
10^7 CFU/ml	10^7 CFU/ml	10^5 CFU/ml
10^6 CFU/ml	10^6 CFU/ml	10^4 CFU/ml
10^5 CFU/ml	10^5 CFU/ml	10^3 CFU/ml
10^4 CFU/ml	10^4 CFU/ml	10^2 CFU/ml
10^3 CFU/ml	10^3 CFU/ml	NG
10^2 CFU/ml	10^2 CFU/ml	NG

When the pan was covered with a steel plate of thickness 5 mm the density of microorganisms in the pan did not change in comparison with control after irradiation during 10 mines. It testifies that presence of the hard X-ray radiation, arising at the accelerator operating, for such times does not result in reduction of microorganisms concentration.

The sterilization efficiency was lower at irradiation Kebsiella Pneumoniae #32 (Table 3). Obviously it is connected with that this culture consists of capsular bacteria, which are less sensitive to various sterilizing factors.

Table 3

Culture K. Pneumoniae #32 (irradiation during 10 min)		
The control	Irradiat. medium	Irradiat. culture
10^8 CFU/ml	10^8 CFU/ml	10^4 CFU/ml
10^7 CFU/ml	10^7 CFU/ml	10^3 CFU/ml
10^6 CFU/ml	10^6CFU/ml	10^2 CFU/ml
10^5 CFU/ml	10^5 CFU/ml	10 CFU/ml
10^4 CFU/ml	10^4 CFU/ml	NG
10^3 CFU/ml	10^3 CFU/ml	NG

If the pan was covered with lavsan film of thickness 20 microns the difference between densities of microorganisms in the open pan and covered one was about two orders (Table 4). In this case, in our opinion, sterilization occurs under influence of soft X-ray radiation from plasma as influence of the charged particles of plasma and radicals produced in plasma in result of plasma-chemical processes are excluded.

Table 4

Culture E. Coli #3 (irradiation during 10 min)		
The control	covered pan	open pan
10^7 CFU/ml	10^4 CFU/ml	10^2 CFU/ml
10^6 CFU/ml	10^3 CFU/ml	10 CFU/ml
10^5 CFU/ml	10^2 CFU/ml	NG
10^4 CFU/ml	10 CFU/ml	NG
10^3 CFU/ml	NG	NG
10^2 CFU/ml	NG	NG

There is an open question on presence and influence of the ultra-violet (UV) radiation that has been observed in gas discharge plasma [4]. However carried out experiments have shown that in such lavsan film UV-radiation is strongly absorbed (the intensity of UV - radiation with wavelength of λ=250 microns is 6 times weakened). When the pan was covered with aluminium foil of thickness 200 microns sterilization efficiency was the same one as in the case the lavsan film.

For research of influence of the microwave radiation, arising at REB interaction with plasma the pan was covered with teflon plate of thickness 10 mm. Such plate is transparent for the microwave radiation but it excludes all other kinds of influence. Sterilization was absent.

4. DISCUSSION

At REB injection into atmosphere plasma is produced as a result of collisional ionization of gas molecules by beam electrons and as a result of cascade processes. Outside of area of the beam propagation the gas ionization is carried out by beam electrons scattered on large angles as well as by high energy secondary electrons. Each relativistic electron (with energy 1-4 MeV), with taking into account cascade processes, creates on 1 cm of its way in atmosphere 60 secondary electrons [10].

The estimation of produced plasma density in dependence on value of the beam current density can be derived from the equation [11]:

0

$$\frac{\partial n_e(z)}{\partial t} = \frac{I_b}{e A_p(z) E_{ei}} \left(\frac{dE}{dZ}\right) - \alpha n_e n_i - k_{np} n_{¿} 2 \qquad (1)$$

where I_b is beam current, A_p is area of the plasma channel produced by electron beam, $\frac{dE}{dZ}$ is rate of ionizing beam losses , E_{ei} is the energy of electron - ion pair production.

The first member in the right part of (1) gives the number of electrons produced by electron beam. The second and third members determine plasma electron losses, which are caused by dissipative recombination with a constant α and by attachment to oxygen with a constant k_{np}. n_i is the ion density which in our case is equal to electron density n_e.

Taking values $\frac{dE}{dZ}$ and E_{ei} from [12], and values α and k_{np} from [13], one can derive the attainable stationary plasma density. At the distance of 10 cm from the exit foil $n_e \approx 3\times10^{11}$ cm^{-3}. The beam density in this cross-section is $n_b \approx 5\times10^{8}$ cm^{-3}. This value of plasma density agrees with experimental data.

At REB propagation in atmosphere in the beam region and near to it nitrogen oxides are produced as a result of chemical reactions of the oscillatory excited molecules and atoms of nitrogen and oxygen [14]:

$$O + N_2^* \rightarrow NO + N; \quad N + O_2^* \rightarrow NO^* + O;$$
$$NO^* + O_2^* \rightarrow NO_2 + O.$$

The presence of NO_2 in our case the experiment testifies, in which at distance of 5 cm aside from the beam axis and at distance of 10 cm from the exit foil a glass pan with 10 cm^3 distilled water was settled. After 10 minutes of the accelerator operation, in the pan the 3% solution of nitric acid was obtained, which presence proved to be true by reaction of "brown ring". Besides a large amount of ozone (up to 0.1 mg/m^3) observed at REB outlet into atmosphere testifies dissociation of oxygen molecules.

5. CONCLUSION

Thus, on the basis of the carried out experiments it is possible to draw the conclusion that sterilization and decontamination of various biological objects with using REB, propagating in atmosphere is more effective comparatively to sterilization and decontamination in vacuum. The plasma produced at REB propagation in atmospheric air and radiations from it are additional sterilizing factors. Thus the main influence is exerted by the radicals, produced as a result of chemical reactions of oscillatory excited molecules of nitrogen and oxygen, the plasma charged particles and soft X-ray radiation from the area of REB interaction with the produced plasma. The microwave radiation from plasma does not practically influence on sterilization efficiency. It needs the further researches the presence of ultra-violet radiation from area of interaction and its influence on sterilization efficiency. The experiments have shown that sterilization efficiency by only plasma at presence of capsular bacteria considerably falls. Therefore in such cases it is preferable to use joint processing by an electron beam and plasma.

REFERENCES

1. A.K. Pikaev // *HVE.* 1993, v.27, N2, p. 44-53.
2. B.M. Peretrante, M.C.Hsiko, J.N.Bardsleg // *Plasma Sources Sci. Technol.* 2001, v.10, N1, p. 82-91.
3. S.Yu. Sokovnin, Yu.A. Kotov, G.A. Mesyaz, S.N.Runin // *Ecologiya.* 1996, N3, p. 222-224.
4. I.A.Soloshenko, V.V.Tsiolko, V.A.Khomich et al. // *Fizika Plazmy*. 2000, v.26, N9, p. 845-853.
5. V.Yu.Bazhenov, A.V.Ryabtsev, I.A.Soloshenko et al. // *Ukr. Jour. Phys*. 2003, v. 48, p. 27-31.
6. E.M.Babich, V.S.Gladkov, E.A.Kornilov, V.A.Kiselev et al. // *Ecotechnologiyi i resursosberezheniye.* 2001, N6, p. 40-43.
7. R.Jonsen // *Rev. Sci. Instr.* 1988, N3, p. 119-125 (in Russian).
8. R.P.Kikvidze, A.A.Rukhadze, N.M.Minaev et al. // *Fizika Plazmy*. 1984, v.10, N5, p. 976-981.
9. V.A.Kiselev, A.F.Linnik, V.I.Maslov, I.N.Onishchenko, V.V.Uskov // *Problems of Atomic Science and Technology.* 2002, N5, p. 101-103.
10. Yu.A.Budagov, G.I.Merzon, B.Sitar. *Ionizing measurements in physics of high energies.* M.: “Energoatomizdat”, 1988, p.36.
11. A.S.Fisher, R.H.Pentel, F.Feinstein et al. // *J. Appl. Phys.* 1988, v.64, N2, p. 572-578.
12. L. Olsen // *Phys Rev*. 1974, v.9, N12, p. 2631-2635.
13. Mac-Daniel. *Processes of collisions in ionizing gas*. M.: “Mir”, 1997.
14. V.D.Rusanov, A.A.Fridman // *DAN SSSR*. 1976, v.231, N5, p. 1109-1112.

СТЕРИЛИЗАЦИЯ ПЛАЗМОЙ, ОБРАЗОВАННОЙ ПРИ ВЫВОДЕ РЭП В АТМОСФЕРУ

Е.М. Бабич, В.А. Киселев, А.Ф. Линник, В.И. Маслов, Н.И. Онищенко, Н.И. Скляр, В.В. Усков

Показано, что при выводе РЭП (4 МэВ, 0.5 А, 2 мкс, 3 Гц,) в атмосферу, кроме стерилизации непосредственно электронами РЭП, более интенсивная стерилизация достигается образованной плазмой. Стерилизация определяется воздействием плазменных частиц, радикалов, мягкого рентгеновского и ультрафиолетового излучений, а так же озона.

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Показано, що при виводі РЕП (4 МеВ, 0.5 А, 2 мкс, 3 Гц,) в атмосферу, крім стерилізації електронами РЕП, більш інтенсивна стерилізація досягається створеною плазмою. Стерилізація визначається дією плазмових часток, радикалів, м’якого рентгенівського та ультрафіолетового випромінювання, а також озону.