Synergistic Effects Obtained by Combined Electron Beam and Microwave Irradiation

D. Martin, G. Craciun, E. Manaila, D. Ighigeanu, C. Oproiu, N. Iacob

National Institute for Laser, Plasma and Radiation Physics, Electron Accelerators Laboratory,
#409 Atomistilor St., P.O.Box-MG36, 077125 Magurele, jud. Ilfov, Romania

Email contact of main author: martin@alpha2.infim.ro

Abstract. A new method based on microwave energy addition to accelerated electron beam energy for biological waste processing is described. The comparative effects obtained by applying separate and combined electron beam (EB) and microwave (MW) irradiation to the microbiological decontamination of the wheat flour, wheat bran and sewage sludge are presented. The research results demonstrated that the combined EB and MW irradiation produces the biggest reduction of microorganisms. The combined irradiation EB + MW decreases the wheat flour NTG by a factor of 7.38 and of 6.58 bigger than separate EB irradiation and separate MW irradiation, respectively. Also, EB + MW procedure decreases the wheat flour moulds by a factor of 3.7 and 2.43 bigger than separate EB irradiation and separate MW irradiation, respectively. The process has been demonstrated in a pilot-scale operation.

1. Introduction

Electron beam (EB) and microwave (MW) treatments are two of the most emerging biological decontamination techniques because in many cases provide distinct advantages over conventional processes in terms of product properties, process time saving, increased process yield and environmental compatibility [1]. Both, EB (ionizing radiation) and MW (non-ionizing radiation) sterilization techniques are based on the radiation ability to alter physical, chemical and biological properties of materials. Irradiation with electron beams was put forth as a very effective method for material biological decontamination because can produce ions, electrons, and free radicals at any temperature in the solid, liquid and gas [2, 3]. EB radiation processes are very effective for sterilization but the required radiation dose is still high. Low irradiation doses are required for the process efficiency and a high dose rate must be used to give large production capacities. Thus, for industrial scale processing, the problem of reducing the absorbed dose level as well as the electron beam cost is especially important. The main idea of this work was to combine the advantages of both, EB irradiation and MW irradiation, i.e. high EB irradiation efficiency and high MW selectivity and volumetric heating for biological waste processing. In this work, the research regarding the investigations on the influence of separate and combined EB and MW irradiation on decontamination of wheat flour, wheat bran and sewage sludge are presented. The first expected and obtained result was the decrease of the required EB absorbed dose level by additional use of MW irradiation. Thus, the ionizing radiation costs could be much reduced and the application of low intensity EB accelerators will become very economically attractive in the field of biological waste processing. EB and MW application for biological waste processing is developed by the project "Study of the combined electron beam and microwave irradiation effects on some foods and therapeutic products", in the framework of the "Bio-Technologies Program" (BIOTECH in Romanian), within the National Program for Research, Development and Innovation, supported by the Education and Research Ministry.

2. Methods

Both, EB and MW disinfection/sterilization processing are based on the radiation ability to alter biological properties of microorganisms especially due to the water presence in the living cells. Water is known to be a component of every biological system and a constituent present in most chemical processes. Due to the presence of water, both electron beam irradiation and microwave irradiation can much enhance the microorganism death rate. The EB processing uses the Coulomb interaction of the accelerated electrons with atoms or molecules of irradiated matter. By this interaction ions, thermalized electrons, excited states and radicals are formed. Thus, the water irradiation by the EB produces radicals such as e^-_{aq}, OH^*, H^*, H_2^*, $H_2O_2^*$, $OH^-_{aq}{}^*$, H_2O^* and O_2^{-*}. The free radicals react with cell membranes, enzymes and nucleic acids to destroy microorganisms. The fact that the interaction by the radicals is effective to a wide range of micoorganisms is one of the advantages of the ionizing irradiation [2, 3]. The various products formed during radiolysis of water may, in this way, influence directly or indirectly the chemical processes and biological effects occurring in the individual compounds dissolved in water. The MW processing is a relatively new technology that provides new approaches to improve the decontamination process compared with classical methods [4-9]. The frequency range of MW (300 MHz - 300 GHz) corresponds to quantum energies ($W=h\nu$, where h is Planck's constant and ν is the radiation frequency), which are small ($1.2\ \mu\text{eV} \le W \le 1.2\ \text{meV}$) as compared to that for ionizing radiation. Hence, MW cannot interact with atoms by generating transitions between principal energy levels, e.g. between a base state and an excited state [10]. Instead of this, microwaves couple to transitions within the hyperfine structure of the dynamical state. Hyperfine splitting of the principal energy levels may be due to the interaction of magnetic moments of the electron shell and of the nucleus [10]. The effect of MW is explained by their heating property on the polar or polarizable molecules of biological systems. Most reports suggest that for various microorganisms, the death rate is enhanced by MW heating more than by conventional heating and the more intense the microwave electric field, the more is the death rate enhancement [4-9]. Also, due to the presence of water, which absorbs MW energy very strongly due its exceptional polarizability, it is possible to pump vibrational modes of DNA leading to unwinding and strand separation. The final comparison is between the two heating methods: conventional and MW. The use of conventional heating requires a significantly longer time than MW heating. Also, with MW heating and its concurrent deep penetration, superior results can be obtained. In addition, it seems that the cancroids residue induction is less by MW heating that by classical heating. A final comparative analysis of the application of EB irradiation and MW heating to the material decontamination processing has led to the following main conclusion: EB processes are very effective for material decontamination and the feature of inducing decontamination at room temperature brings unique advantages of EB over MW processing. Regarding to food biological decontamination, opponents argue that research not proved the safety of ionizing irradiation but the cumulative evidence from over four decades of research, carried out in laboratories in the United States, Europe and other countries worldwide, indicates that irradiated food is safe to eat. The EB (up to 10 MeV) irradiated food is not radioactive, there is no evidence of toxic substances resulting from irradiation and there is no evidence or reason to expect that irradiation produces more virulent pathogens among those that survive to irradiation treatment. The radiolitic products creation is not limited to the irradiation process: cooking, canning and pasteurization also produce radiolytic products. At the prescribed dosage levels, irradiation produces small amounts of such compounds. However, fore more public acceptance, any dose level reduction in the ionizing radiation food processes is better. MW processes are less effective for food decontamination than EB processes but the cost of MW systems is considerably smaller than ionizing radiation systems. The research has shown that some microorganisms exhibit more sensibility to EB irradiation and other to MW exposure. Thus, by combined EB and MW irradiation could be possible to extend the kind range of microorganisms to be inactivated. The main idea of this work is to combine the advantages of both, EB and MW, for the material microbiological decontamination, i.e. the EB high efficiency and MW high selectivity and volumetric heating, in order to assure higher material microbiological safety, to extend the kind range of microorganisms to be inactivated, to reduce the required EB absorbed dose level and irradiation time to a minimum and to decrease the decontamination process costs. Also, the combined effects of MW heating with EB irradiation could decrease the average microwave power level while keeping the temperature increase as low as possible.

3. Apparatus

For comparative studies of EB and MW irradiation an original installation which permits separate EB irradiation, separate MW irradiation and combined (successive or simultaneous) EB and MW irradiation was designed. Fig. 1 and Fig. 2 show schematic drawing and photograph of this installation, respectively. This installation consists mainly of the following units: an EB source (ALID-7 electron linear accelerator of 5.5 MeV and 670 W, built in Romania, National Institute for Lasers, Plasma and Radiation Physics, Electron Accelerator Laboratory-Bucharest); two microwave injection systems of 2.45 GHz and 850 W maximum output power; a multimode rectangular cavity of 612 mm x 612 mm x 367 mm inner dimensions, in which are injected both EB and MW; a conveyor which moves the vessels with samples. The scanned EB is introduced perpendicularly to the cavity upper end plate through a 100 μm thick aluminum foil while MW power is coupled by two slotted waveguide systems used as MW radiating antennas. For the applications performed with this installation we used the pulse modulated MW of 2.45 GHz in order to keep the MW power at high peak level and to control average power to the level which overcome the samples temperature over-rise, especially in the food decontamination case. Trains of high voltage pulses followed by inhibited high voltage pulses are periodically applied on the magnetron anode yielding a corresponding variation of the average magnetron output power, with programmed steps, maintaining the advantage of peak microwave output power operation [11]. The ALID-7 accelerator is of travelling-wave type, operating at a wavelength of 10 cm. It is driven by a tunable S-band magnetron (EEV M 5125 type), delivering 2 MW of power in 4 μs pulses. The electrons are injected from a diode type gun at a voltage of 80 kV and travel through the accelerating structure, a disk-loaded tube operating in the $\pi/2$ mode. The first portion of the accelerating structures is a variable phase velocity buncher and the remainder has a uniform section. The optimum values of the EB peak current I_{EB} and EB energy E_{EB} to produce maximum output power P_{EB} for a fixed pulse duration τ_{EB} and repetition frequency f_{EB} are as follows: E_{EB} = 5.5 MeV; I_{EB} =130 mA; P_{EB} = 670 W (f_{EB} = 250 Hz, τ_{EB} = 3.75 μs). The radiation effects are related to the amount of energy deposited into a certain mass of matter. The dose is the total energy deposited by radiation per unit mass of the medium. It is expressed in Gray (1 Gy = 1 Joule $\cdot$ kg^{-1}). The following types of irradiation procedure were performed: separate EB irradiation; separate MW irradiation; successive irradiation: first MW irradiation and then EB irradiation, (MW+EB); successive irradiation: first EB irradiation and then MW irradiation, (EB+MW); simultaneous irradiation with EB and MW, S(EB+MW).

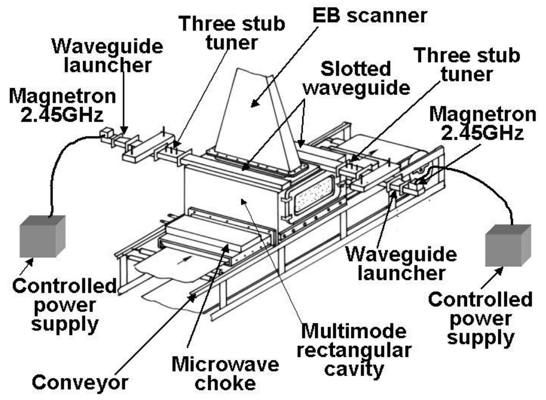

FIG. 1. Schematic drawing of the installation for combined EB and MW irradiation with conveyor

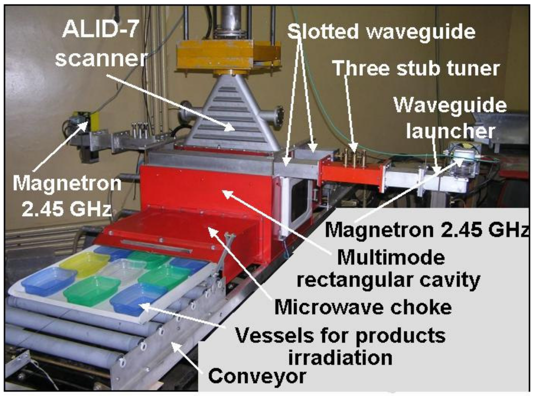

FIG. 2. Photograph of the installation for combined EB and MW irradiation

3. Results

The research comprised the investigations on the influence of separate and combined EB and MW irradiation of the following samples: sewage sludge of 10^{-4} m^3, wheat flour of 0.08 kg and wheat bran of 0.05 kg. The samples were irradiated in plastic boxes (that are current used in MW ovens) put in sealed plastic bags. The results are presented in Figs. 3-16.
Fig. 3 and Fig. 4 present the results obtained by separate EB irradiation, separate MW irradiation and simultaneous irradiation with EB and MW, S(EB+MW), upon the total number of germs (TNG) and total number of coliform bacteria (TNCB) in sewage sludge samples of 10^{-4} m^3 performed from a food industry wastewater treatment station (vegetable oil plant). The results shown in Fig. 3 demonstrate that the properly applied microwave irradiation (proper microwave power level and proper irradiation time) has the same effects upon TNG as EB irradiation. Thus, 1 minute MW irradiation of 670 W has the same effects as 0.5 kGy EB irradiation. Also, as shown in Fig. 3 and Fig. 4, 3 minutes MW irradiation of 670 W has the same effect upon TNG and TNCB as 4 kGy EB irradiation. Both, Fig. 3 and Fig. 4 demonstrate that the simultaneous EB and MW irradiation produces the biggest reduction of TNG and TNCB. Thus, 1-minute irradiation of simultaneous EB of 0.5 kGy and MW of 670 W has the same effect upon TNG (Fig. 3) and TNCB (Fig. 4) as separate EB irradiation of 4 kGy. Also, 3 minutes irradiation time of simultaneous EB of 2 kGy and MW of 670 W has the same effect upon TNG as separate EB irradiation of 10 kGy (Fig. 3).
Fig. 5 and Fig. 7 present the effect of separate electron beam irradiation upon TNG and moulds of wheat flour, respectively. Also, Fig. 9 and Fig. 11 present the effect of separate electron beam irradiation upon TNG and moulds of wheat bran, respectively. Figs.5, 7, 9 and 11 show that, for separate EB irradiation, the survival fraction of TNG and moulds diminishes exponentially with EB absorbed dose. Fig. 6 and Fig. 8 present the effect of separate microwave irradiation time upon TNG and moulds of wheat flour, respectively. Also, Fig. 10 and Fig. 12 present the effect of separate microwave irradiation time upon TNG and moulds of wheat bran, respectively. Figs. 6, 8 and 10 show that, for separate MW irradiation, the survival fraction of TNG and moulds exhibits an oscillatory decrease versus MW irradiation time: periods of TNG inhibition and moulds stimulation are followed by periods of TNG stimulation and moulds inhibitions. This is a new result that was proved by our experimental investigations for many microorganism types [12]. Figs. 13, 14 and Figs. 15, 16 give the comparative results concerning the effects of different irradiation modes upon TNG and moulds for samples of wheat flour and wheat bran, respectively. The results regarding

combined EB and MW irradiation, demonstrate that both irradiation procedures, EB+MW (successive irradiation, first EB and then MW) and MW+EB (successive irradiation, first MW and then EB), cause greater lethal effects upon TNG and moulds than separate EB irradiation or separate MW irradiation. Thus, the MW+EB irradiation procedure decreases TNG of wheat flour by a factor of 7.38 and 6.58 bigger than separate EB irradiation and separate MW irradiation, respectively. Also, MW+EB procedure decreases the moulds of wheat flour by a factor of 3.7 and 2.43 bigger than separate EB irradiation and separate MW irradiation, respectively. The MW+EB irradiation procedure decreases the TNG of wheat bran by a factor of 17.8 and 37.9 bigger than separate EB irradiation and separate MW irradiation, respectively. Also, MW+EB procedure decreases the moulds of wheat bran by a factor of 4.2 and 12.2 bigger than separate EB irradiation and separate MW irradiation, respectively. The EB required absorbed dose with MW+EB irradiation of wheat flour is decreased by a factor of 5 for TNG and by a factor of 2 for moulds compared to separate EB irradiation. The EB required absorbed dose with MW+EB irradiation of wheat bran is decreased by a factor of 2 for both, TNG and moulds, compared to separate EB irradiation. The EB+MW procedure seems to be less effective than MW+EB with one exception: upon wheat bran moulds, as seen in Fig. 16. However, the EB+MW procedure is more effective then separate EB or MW irradiation as shown in Figs. 13-16.

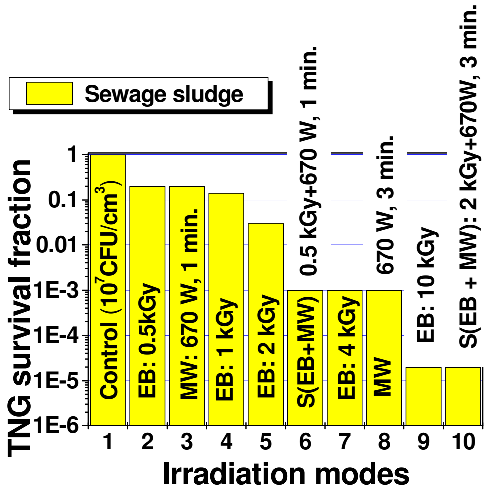

FIG. 3. The effect of different irradiation modes upon TNG

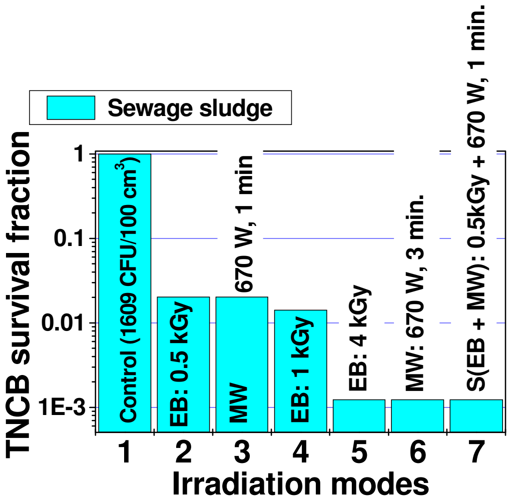

FIG. 4. The effect of different irradiation modes upon TNCB

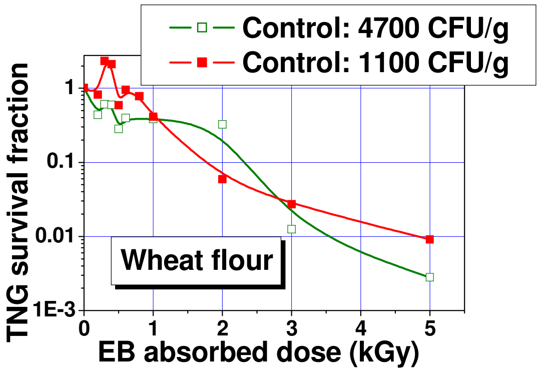

FIG. 5. The effect of separate electron beam irradiation upon TNG of wheat flour

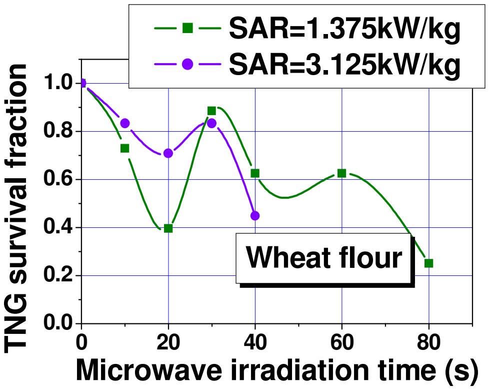

FIG. 6. The effect of separate microwave irradiation upon TNG of wheat flour

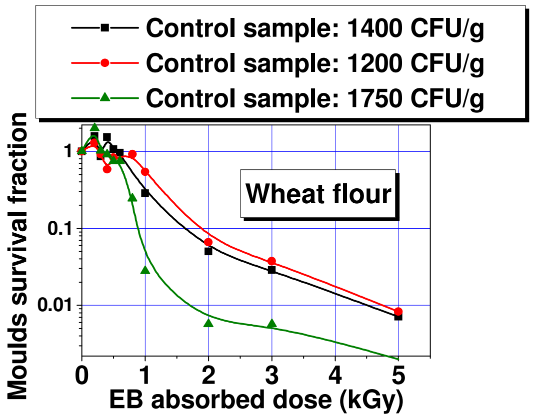

FIG. 7. The effect of separate electron beam irradiation upon moulds of wheat flour

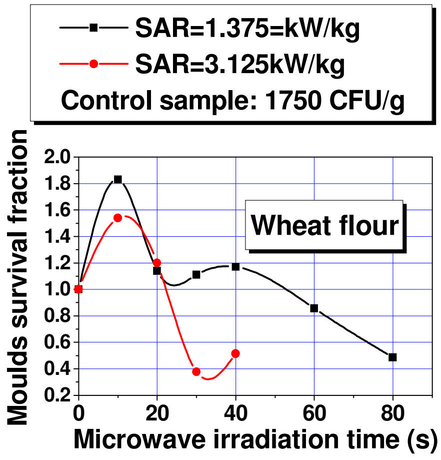

FIG. 8. The effect of separate microwave irradiation upon moulds of wheat flour

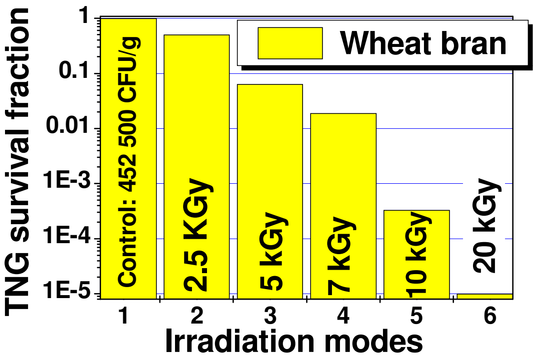

FIG. 9. The effect of EB absorbed dose upon wheat bran TNG

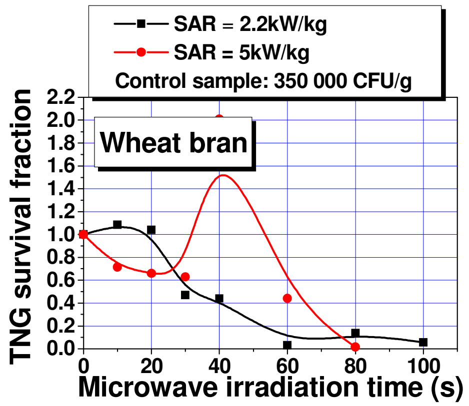

FIG. 10. The effect of MW irradiation time upon heat bran TNG

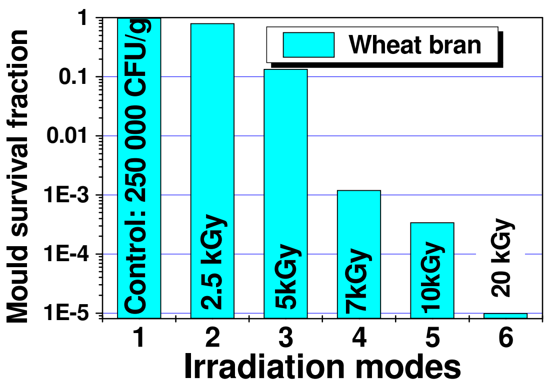

FIG. 11. The effect of EB absorbed dose upon wheat bran moulds

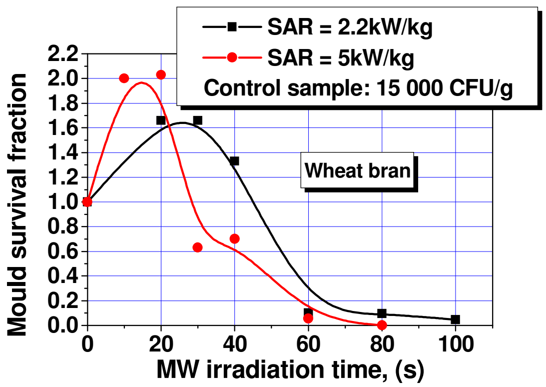

FIG. 12. The effect of MW irradiation time upon wheat bran moulds

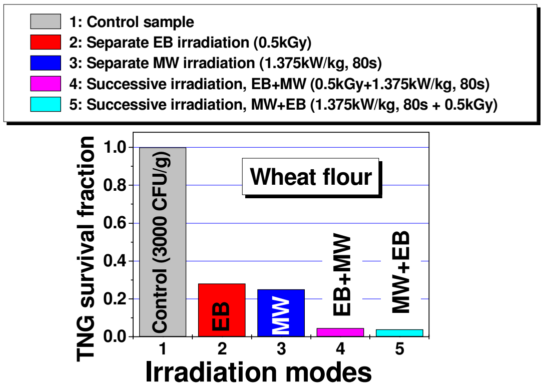

FIG. 13. The effect of different irradiation modes upon wheat flour TNG

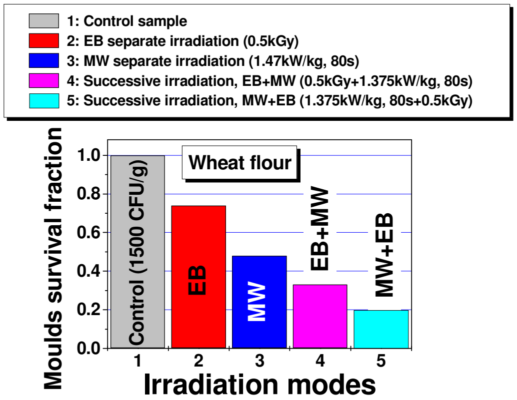

FIG. 14. The effect of different irradiation modes upon wheat flour moulds

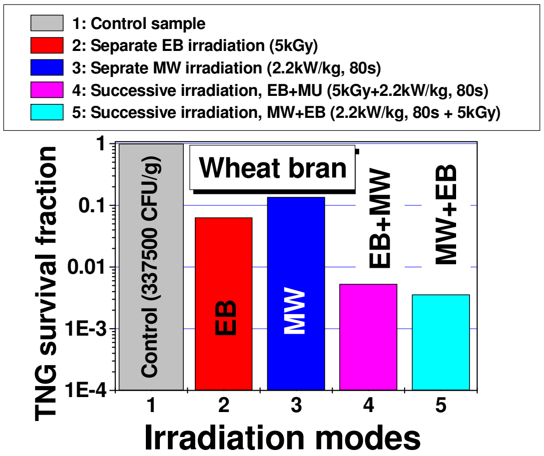

FIG. 15. The effect of different irradiation modes upon TNG of wheat bran

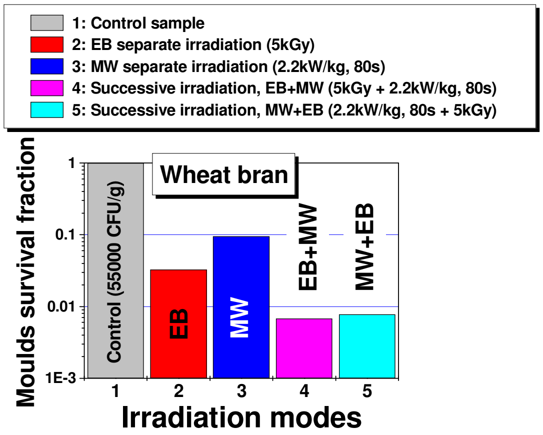

FIG. 16. The effect of different irradiation modes upon moulds of wheat bran

4. Conclusions

Separate MW irradiation, as compared with EB irradiation that diminishes exponentially the viable cells versus EB absorbed dose, always induces an oscillatory decrease of survival fraction of microorganisms versus MW irradiation time: periods of germs inhibition or stimulation are followed by periods of germs stimulation or inhibitions. This is a new result that was proved by our experimental investigations for many microorganism types.

Combined EB and MW irradiation produces the biggest reduction of microorganisms. In our opinion, it seems that microwave irradiation could cause the modification of the microorganisms sensitivity to EB irradiation. Thus, the application of combined EB and MW irradiation lead to greater lethal effects than the EB irradiation alone. Also, the tests demonstrated that irradiation time and the upper limit of EB required absorbed dose, which ensures a good decontamination effect, could be reduced by a factor at least of two by additional use of MW energy to EB energy. The research has shown that some microorganisms exhibit more sensibility to EB irradiation and other to MW exposure. Thus, by combined EB and MW irradiation could be possible to extend the kind range of microorganisms to be inactivated. The most important conclusion is that the combined electron beam and microwave irradiation could become a new disinfection/sterilization method, commercially viable alternative to classical thermal or chemical destruction. Also, ionizing irradiation costs could be much decreased and the application of low intensity radiation sources, which are less expensive, will be extended for the sanitation/sterilization of a wide variety of materials including food items, medical objects, hospital waste, waste water and sewage sludge. The technology of sludge irradiation followed by composting could be developed to produce disinfected compost for agriculture.

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