Depreciated Food Treatment by Combined Ionizing and Non-ionizing Radiations

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Abstract : The combined effects of ionizing radiations (electron beams of 5.5 MeV) and non-ionizing radiations (microwaves of 2.45 GHz) on the inactivation of total number of germs (TNG) and moulds in depreciated food such as wheat bran, wheat flour and minced beef are presented. The following types of treatments were performed with electron beams (EB) and microwaves (MW): separate EB irradiation, separate MW irradiation, successive irradiation (EB irradiation followed by MW heating) named (EB+MW), successive irradiation (MW heating followed by EB irradiation) named MW+EB and simultaneous irradiation with EB and MW named S(EB+MW). The first interesting experimental result is that the microorganism's survival fraction diminishes exponentially with EB dose for the separate EB irradiation while for the separate MW irradiation the microorganism's survival fraction exhibits an oscillatory decrease versus MW dose: periods of microorganism inhibition are followed by periods of microorganism stimulation. The second important result is that all combined irradiation procedures such as S(EB+MW), followed by MW+EB and then by EB+MW, cause greater reduction of NTG and mould survival fractions than separate EB irradiation or separate MW irradiation. The S(EB+MW) treatment type produces the maximum reduction of NTG and moulds. The third important result is that the EB dose level could be reduced by a factor in the range of 1.5-2.5 by combined EB and MW irradiation.

Keywords: - Waste food treatment, Electrons, Microwaves, Combined irradiation

1 Introduction

In many cases EB and MW treatments provide distinct advantages over conventional processes in terms of product properties, process time saving, increased process yield and environmental compatibility. A technical and economic comparison between EB irradiation and microwaves treatment for industrial applications is given as a preliminary analysis in [1]. EB [2-10] and MW [11-21] decontamination techniques represent two technically and commercially viable alternatives to thermal or chemical-physical processes that are more and more socially and politically less accepted. Thermal and chemical processes represent, at present time, the most largely diffused technology for biological wastes treatment. Incineration plants utilize liquid or solid fuels in addition to refuses (mean heat value of 3000 kcal/kg) and the final results are ashes and gases: ashes must managed as hazardous wastes, gases (acid gases mixed with organo-chlorine pollutants) require special systems and equipment (post-combustion room, filters, scrubbers, sprayers) before discharge into atmosphere [1].

Ethylene oxide (EtO) is widely used as a sterilant because of its potency in destroying pathogens. The EtO use is of regulatory concern because EtO is: flammable and explosive, probable human carcinogen, a toxic air contaminant, an ozone depleter. Radiation processes have proved to be better than conventional process (mainly EtO) with regards to safety for workers and consumers, disinfection / sterilization reliability and processing capability as well as managing.

A comparative analysis of the application of separate EB irradiation and separate MW heating to the sterilization processing has led to the following main conclusions:

1) Both EB and MW microorganism inactivation methods are based on the radiation ability to alter physical, chemical

and biological properties of materials [4 and 22]. However, there is a fundamental difference between the EB (ionizing radiation) and MW (nonionizing radiation) in their interaction with matter: EB interacts with matter by the same physical mechanisms (basically, Coulomb interaction with electrons in inorganic and organic materials generating excitation and ionization) at any matter state (solid, liquid and gas phase) and matter temperature while MW interacts with matter by different physical action principles as a function of matter state and temperature, MW frequency and polarization, environmental factors (temperature, humidity), samples volume and geometry, MW applicator types etc.

2) 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. The free radicals produced by EB react with cell membranes, enzymes and nucleic acids to destroy microorganisms. However, EB required dose is high. Thus, for industrial scale processing, the problem of reducing the absorbed dose level is especially important. Low irradiation doses are required for the process efficiency and a high dose rate must be used to give large production capacities. Also, for more public acceptance, any dose level reduction of the ionizing radiation processes is desired.
3) MW processes are less effective for biological waste decontamination than EB processes but the cost of MW systems is considerably smaller than ionizing radiation systems. Otherwise, MW heating offers a lot of advantages as compared with conventional heating such as less start up time, faster heating, selectivity, energy efficiency (most of the electromagnetic energy is converted into heat) and the reduction of processing time up to three orders of magnitude [16]. Conventional heating modes usually used in sterilization, disinfection or sanitation lead to surface heating, whereas MW penetration within the material leads to a core heating or a volumetric heating because of wave length which has the same order of magnitude as the material dimensions [16]. In this case fast temperature rises take place while the heat is conducted from the core to the surface of the material contrary to the conventional heating (surface heating) where the heat goes from the surface to the core. The frequency range of microwave (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 ($1.2\ \mu eV \leq W \leq 1.2\ meV$) as compared to that for ionizing radiation [23]. Hence microwaves cannot interact with atoms by generating transitions between principal energy levels, e.g. between a base state and an excited state. Instead of this, microwaves may couple to transitions within the hyperfine structure of the dynamical state [23]. Hyperfine splitting of the principal energy levels may be due to the interaction of magnetic moments of the electron shell and of the nucleus [23]. This mechanism could induce a resonant absorption of MW energy and in this way it is possible that MW to pump vibrational modes of DNA leading to unwinding and strand separation. However, the existence and origin of such “non thermal” effects is still a matter of controversial discussions [24-28]. The effect of microwaves is currently 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 microwave heating more than by conventional heating and the more intense the microwave electric field the more is the death rate enhancement [13-15]. However, the exact nature of the MW sterilizing effect is not well understood and no theory currently exists to explain it. The advantages of the use of microwaves and microwave systems in a wide range of areas, including disinfection / sterilization processing can be summarized as follows [11-21]: rapid energy transfer; volumetric and selective heating; very high heating rate; convenient and clean heating; fast switch on and off; clean environment, free from products of combustion; compact equipment; the microwave system cools very rapidly when the field is switched off.
4) 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 EB irradiation and MW irradiation can much enhance the microorganism’s death rate. Thus, the irradiation by the EB of water produces a lot

of radicals. The fact that the interaction by these radicals is effective to a wide range of pollutants is the one of advantages of the EB irradiation. 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. Also, because to the presence of water, which absorbs MW energy very strongly due its exceptional polarizability, MW processing is very effective for microorganism inactivation.

5) The reported research results have shown that some microorganisms exhibit more sensibility to EB irradiation and other to MW exposure. This suggests that, by combined EB and MW irradiation could be possible to extend the kind of microorganisms to be inactivated.

In view of the above conclusions, we decided to develop an innovative waste food processing based on the sterilizing effects of both, EB irradiation and MW heating. The main goal of this work was to combine the advantages of both, EB and MW, i.e. the EB high efficiency and MW high selectivity associated with volumetric heating, in order to assure higher material microbiological safety, to extend the kind of microorganisms to be inactivated, to reduce the required absorbed dose level and irradiation time and thus to decrease the sterilization process costs. This application is based on our laboratory results concerning the effects of combined EB and MW on different kind of bacteria in cultures [29]. This paper presents the comparative results obtained by applying separate EB irradiation, separate MW heating and combined (successive and simultaneous) EB irradiation and MW heating to the reduction of total number of germs (TNG) and moulds in natural products such as waste wheat bran, waste wheat flour and waste minced beef. In view of the argument that the separate EB processing as well as the separate MW processing can strongly support the treatment of industrial waste streams, it is expected that combined EB and MW processing to become also a technically and commercially viable alternative to thermal or chemical-physical processes.

2 Experimental Installations and Procedures

A small-pilot installation, named SPI-EB+MW, was built for the comparative studies of waste food decontamination using separate and combined (successive and simultaneous) EB irradiation and MW heating (Fig. 1 and Fig. 2).

The SPI-EB+MW consists mainly of the following units:

- An EB source (ALID-7 electron linear accelerator of 5.5 MeV and 670 W maximum output power);
- A multimode rectangular cavity, MRC, of 0.612 m x 0.612 m x 0.367 m inner dimensions, in which are injected both EB and MW;
- A conveyor which moves the vessels with samples through MRC.

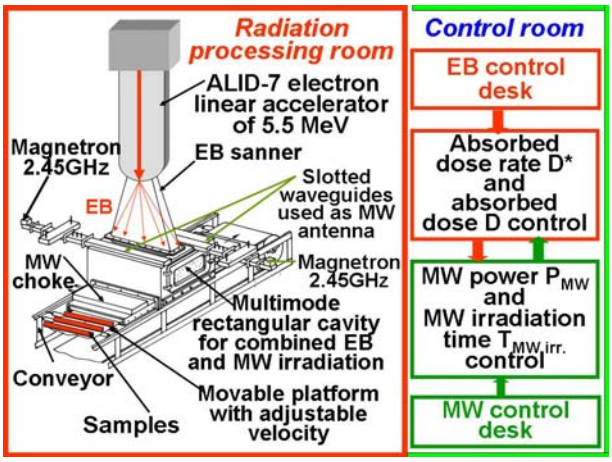

Fig. 1. The schematic drawing of the small-pilot installation for combined EB and MW irradiation SPI-EB+MW

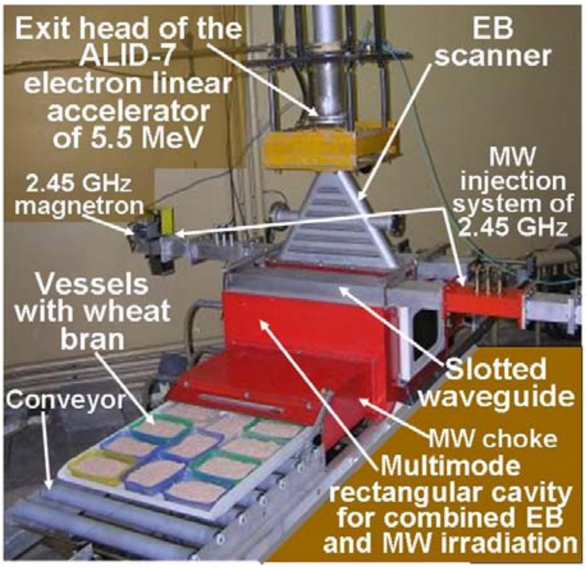

Fig. 2. Photograph of the small-pilot installation for combined EB and MW irradiation SPI-EB+MW

The ALID-7 electron linear accelerator was built in the Electron Accelerator Laboratory, National Institute for Lasers, Plasma and Radiation Physics, Bucharest, Romania [30]. It is of the travelling-wave type, driven by 2-MW peak power tunable EEV M5125-type magnetrons operating in S-band. 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 accelerator ALID-7 is an industrial equipment which was initially designed and optimized to produce maximum X-ray output for nondestructive testing. Due to ever growing ecological problems we are facing, this accelerator is at present used for research to small pilot scale level application of the combined EB and MW irradiation for food decontamination, polyelectrolyte production, SO_2, NOx and VOCs removal and other.

The scanned EB is introduced perpendicularly to the MRC upper end plate through a 100 μm thick aluminum foil while MW power is coupled through the same MRC upper end plate by two slotted waveguides used as MW radiating antennas. The slotted waveguide system provides good microwave energy transfer and uniformity over a large area [31]. Each slotted waveguide, consisting of several inclined series slots cut in the broad wall of a WR430 standard rectangular waveguide propagating the dominant mode, have the advantage of simplicity of structure, manufacturing ease and adaptability of configuration to meet radiation pattern requirements [31]. The WR430 waveguide was selected because it has shortest guide wavelength (λ_g) at the used frequency (2.45 GHz) which provides that the radiating slots to be placed as close as possible. All slots are spaced $\lambda_g/2$ away from adjacent slots in the same waveguide. The MW power of 2.45GHz is fed into one end of the slotted waveguide and the other slotted waveguide end is connected to a movable short circuit for impedance matching. The conventional operation of 2.45 GHz oven magnetron supplied by an L.C. single-phase-half-wave doubler (LHCHWD) was properly modified in order to permit the use of a manually-controlled or PC-controlled electronic variator for the MW power adjustment from zero to 850 W for each slotted waveguide [32].

In the present work, we have used a sampling method involving an electron collection monitor and its associated instrumentation for monitoring absorbed dose rate and accumulated absorbed dose during the irradiation process. This monitor which intercepts only a fraction of the scanned electron beam gives a relative value of the absorbed dose rate: it has been first calibrated by several chemical systems (such as the Ceric dosimeter) placed at the position of the samples to be irradiated. The thickness of the irradiated samples was set to be in accordance with the EB useful penetration, i.e. the sample densities (0.3 g/cm^3 for waste wheat bran, 0.5 g/cm^3 for waste wheat flour and 1 g/cm^3 for waste minced) and EB energy of 5.5 MeV, used in experiments with SPI-EB+MW. The samples were irradiated in plastic boxes (that are current used in MW ovens) put in sealed plastic bags.

The EB effect is expressed by the absorbed dose (D) in Gray or J kg^{-1} and absorbed dose rate (D*) in Gy s^{-1} or J kg^{-1} s^{-1}. The MW effect is expressed by SAR (Specific Absorption Rat) in W kg^{-1}, which is equivalent to D* and SA (Specific Absorption) in J kg^{-1}, which is equivalent to D [33]. For the combined EB and MW irradiation (successive and simultaneous), the EB absorbed dose rate was set to a certain level in order to accumulate, during the irradiation process, a certain EB absorbed dose (0.5 kGy, 1 kGy, 2 kGy, 5 kGy and so on). For MW irradiation we used the SAR values to prevent during irradiation time the rise of the final sample temperature below 40-45^{o}C: 1.375 kW kg^{-1} for minced beef and wheat flour and 2.2 kW kg^{-1} for wheat bran. The proper correlation between D* and D characterizing EB irradiation and SAR and SA characterizing MW heating is a very difficult procedure because the SAR and SA values depend strongly on the following factors: 1)sample physical and chemical properties; 2) sample temperature, humidity, quantity and geometry; 3) environmental factors (temperature, humidity); MW applicator geometry. Due to these considerations, the SAR and SA values must be determined prior the food processing by combined EB irradiation and MW heating.

3 Results and Discussions

The following types of irradiation modes were performed:

- Separate EB irradiation;
- Separate MW irradiation;
- Successive irradiation (EB irradiation followed by MW heating) named EB+MW;
- Successive irradiation (MW heating followed by EB irradiation) MW+EB;
- Simultaneous irradiation with EB and MW named S(EB+MW).

Figs. 3-11 present the results obtained by the above mentioned irradiation modes upon survival fraction (number of colony forming units of irradiated samples per number of colony forming units of control samples) of TNG (total number of germs) and moulds in samples of wheat bran, wheat flour and minced beef. The effectiveness of separate and combined EB and MW irradiation was determined by counting the total number of viable colonies after each treatment type as well as in controls (non-irradiated samples). 10 g from each exposed sample (as well as control sample) was suspended in 90 ml sterile, distilled water in order to obtain initial dilution (10^{-1}) that was again serial diluted (rate 10) in distilled water ($10^{-2}…10^{-10}$). One ml from each dilution was then transferred to the Petri dishes (two Petri dishes from each dilution) and then added nutrient broth (solution of 52 g of Brain Heart Infusion Agar powder dissolved in 1000 ml distilled water). The Petri dishes were incubated at 37°C for 24 hours. After incubation the number of colony forming units (CFU) was counted and averaged to determine total viable colonies per gram (CFU g^{-1}).

The first interesting experimental result is that the microorganism's survival fraction diminishes exponentially with EB dose for the separate EB irradiation while for the separate MW irradiation the microorganism's survival fraction exhibits an oscillatory decrease versus MW dose: periods of microorganism inhibition are followed by periods of microorganism stimulation. A representative example is given in Fig. 3 and Fig. 4 for TNG survival fraction of waste wheat bran samples irradiated by EB and MW, respectively. The microorganism's behavior at specific MW exposure conditions challenges conventional assumptions that the magnitude of a response increases with increases "dose" (specific absorption SA in the case of MW treatment) [34]. The microorganisms exhibit at MW exposure an abnormal behavior that cannot be readily identified as compared with their behavior at ionizing radiations. Fig. 5 shows that at higher SAR level (5 kW kg^{-1} compared with 2.2 kW kg^{-1}) the oscillatory decrease shape of the TNG survival fraction is kept but the amplitudes of the maximum and minimum peaks are much diminished. Also, the succession of TNG inhibition and stimulation peaks is modified.

The second very important result is that all combined irradiation procedures, such as S(EB+MW) followed by MW+EB and then by EB+MW, cause greater reduction of NTG and mould survival fractions than separate EB irradiation or separate MW irradiation. The S(EB+MW) treatment type produces the maximum reduction of NTG and moulds. The effects of different irradiation mode upon TNG and moulds survival fraction are given in Figs.6-11 where each column represents an average on 4 distinct measurements made after 4 distinct experiments performed in the same conditions for each type of irradiation mode: MW, EB, EB+MW, MW+EB and S(EB+MW). As shown in Figs. 6-11, the S(EB+MW) simultaneous irradiation resulted in the highest microorganism's reduction, followed by the MW+EB successive irradiation, EB+MW successive irradiation, EB separate irradiation and finally MW separate irradiation. The following results are representative:

- S(5 kGy + 110W/80 s) simultaneous irradiation with EB and MW decreases TNG in wheat bran by a factor of 65.4 and 11000 bigger than EB separate irradiation of 5 kGy and MW separate irradiation of 110 W/80 s, respectively (Fig. 6). Also, S(5 kGy+110 W/80 s) simultaneous irradiation with EB and MW decreases the moulds in wheat bran by a factor of 12 and 428.6 bigger than EB separate irradiation of 5 kGy and MW separate irradiation of 110 W/80 s, respectively (Fig. 7);
- S(1 kGy + 110W/80 s) simultaneous irradiation with EB and MW decreases TNG in wheat flour by a factor of 7.5 and 20.83 bigger than EB separate irradiation of 1 kGy and MW separate irradiation of 110 W/80 s, respectively (Fig. 8). Also, S(1 kGy+110 W/80 s) simultaneous irradiation decreases the moulds in wheat flour by a factor of 6.75 and 6.1 bigger than EB separate irradiation of 1 kGy and MW separate irradiation of 110 W/80 s, respectively (Fig. 9);
- S(2 kGy+110 W/50 s) simultaneous irradiation with EB and MW decreases TNG in minced beef by a factor of 6.29 and 108.5 bigger than EB separate irradiation of 2 kGy and MW separate irradiation of 110 W/50 s, respectively (Fig. 10). Also, S(2 kGy+110 W/50 s) simultaneous irradiation with EB and MW decreases the moulds in minced beef by a factor of 3.175 and 32.5 bigger than EB separate irradiation of 2 kGy and MW separate irradiation of 110 W/50 s, respectively (Fig. 11);
- S(5 kGy+110 W/50 s) simultaneous irradiation with EB and MW decreases TNG in minced beef by a factor of 7.2 and 620.5 bigger than separate EB irradiation of 5 kGy and MW irradiation of 110 W/50 s, respectively (Fig.10). Also, S(5 kGy+110 W/50 s) simultaneous irradiation with EB and MW decreases the moulds in

minced beef by a factor of 4.33 and 86.7 bigger than EB separate irradiation of 5 kGy and MW separate irradiation of 110 W/50 s, respectively (Fig. 11);

The third important result is that the EB dose level could be reduced by a factor in the range of 1.5-2.5 by combined EB and MW irradiation. The following results are representative:

- S(5 kGy+MW) simultaneous irradiation has the same effect on the TNG (Fig. 6) and moulds (Fig. 7) in wheat bran as EB separate irradiation of about 10 kGy;
- S(1 kGy+MW) simultaneous irradiation has the same effect on the TNG in wheat flour as EB separate irradiation of about 1.5 kGy (Fig. 8). Also, S(1 kGy+MW) simultaneous irradiation has the same effect on the moulds in wheat flour as EB separate irradiation of about 2.5 kGy (Fig. 9);
- S(2 kGy+MW) simultaneous irradiation has the same effect on the TNG and moulds in minced beef as EB separate irradiation of about 5 kGy (Figs. 10 and 11). Also, S(5 kGy+MW) simultaneous irradiation has the same effect on the TNG and moulds in minced beef as EB separate irradiation of about 10 kGy (Figs. 10 and 11).

Fig. 12 and Fig. 13 give some demonstrative examples regarding the effects of combined EB and MW irradiation on several quality indicators for wheat bran (protein, moisture) and wheat flour (protein, moisture, gluten quality index, gluten deformation, wet gluten, ash) controlled by our national regulations. As shown in Fig. 12 and Fig. 13, combined EB and MW irradiation (especially simultaneous EB and MW irradiation) produces small changes of tested quality indicators at the used dose levels. Table 1 presents the variation of TNG and moulds in control samples and samples of waste wheat bran irradiated with S(5 kGy + 2.2 kW/kg x 80 s) in the first month and then tested monthly during 7 months to determine the effectiveness of S(EB+MW) sterilization. The results presented in Table 1 demonstrate the efficacy of waste food processing based on the simultaneous EB irradiation and MW heating.

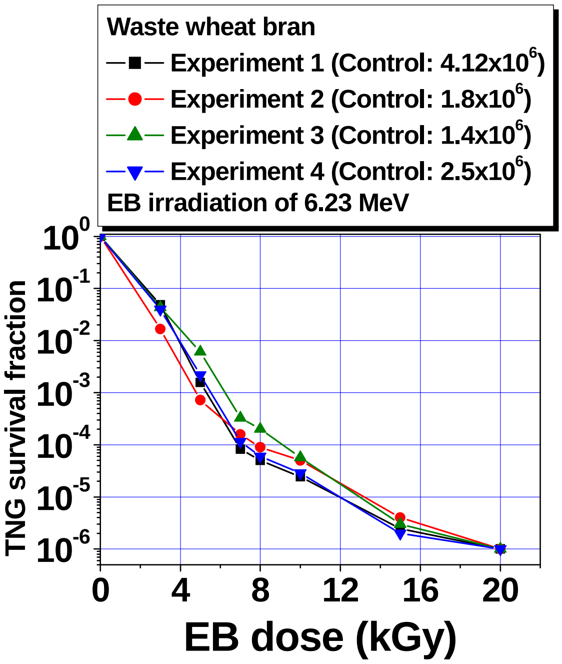

Fig. 3. TNG survival fraction versus EB dose in waste wheat bran

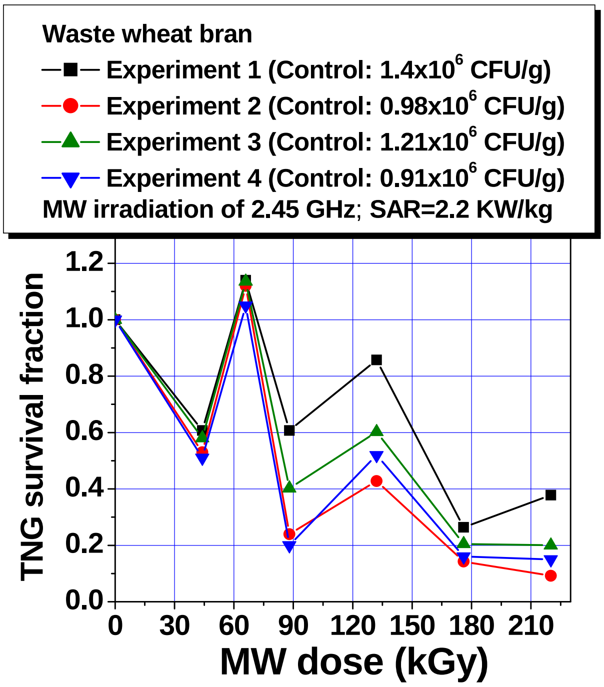

Fig. 4. TNG survival fraction versus MW dose in waste wheat bran

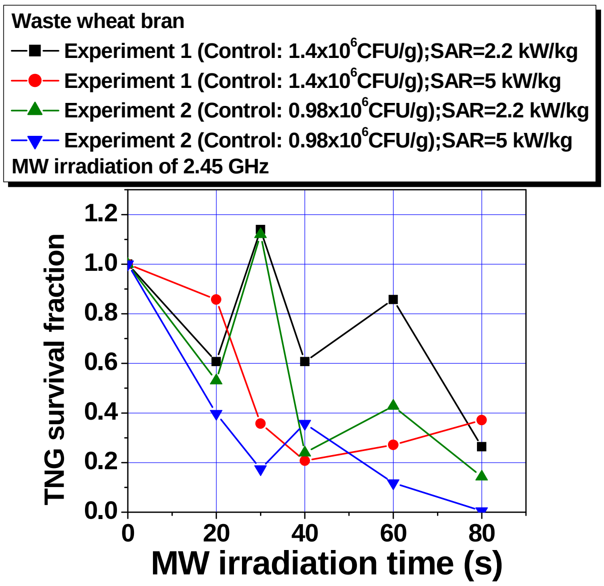

Fig. 5: TNG survival fraction versus SAR and MW irradiation time in waste wheat bran

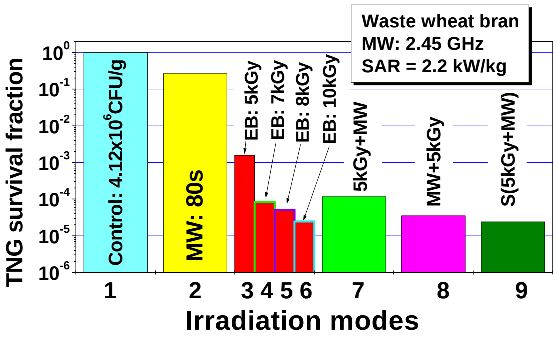

Fig. 6. The effect of different irradiation modes upon the TNG survival fraction in waste wheat bran samples

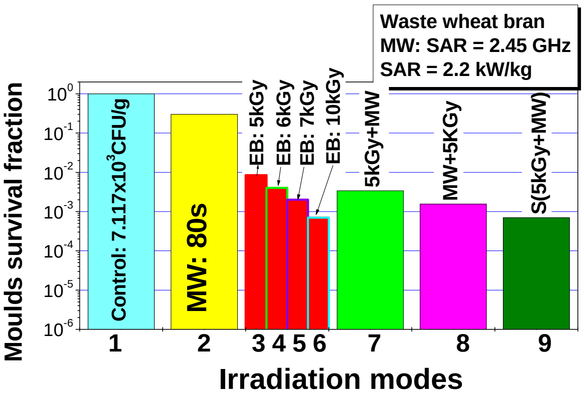

Fig. 7. The effect of different irradiation modes upon the moulds survival fraction in waste wheat bran samples

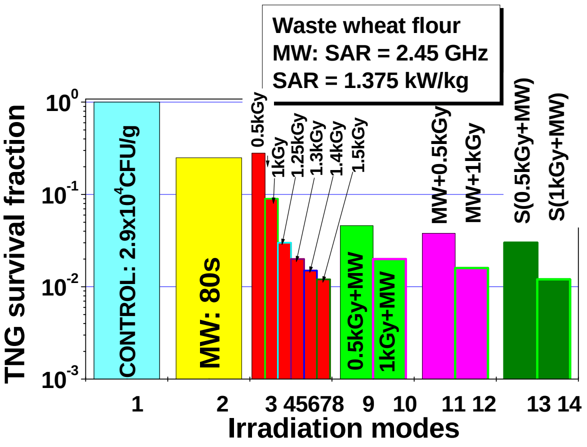

Fig. 8. The effect of different irradiation modes upon the TNG survival fraction in waste wheat flour samples

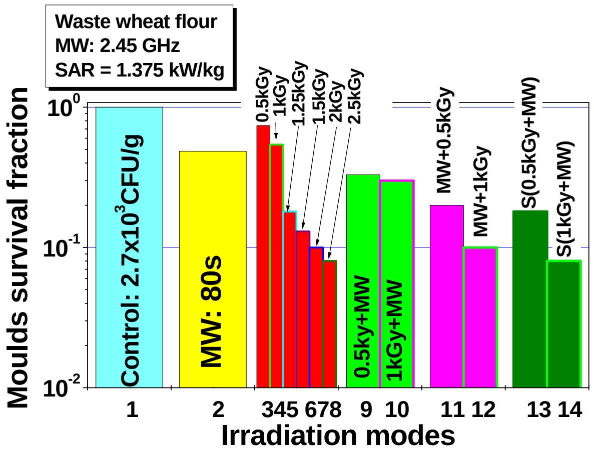

Fig. 9. The effect of different irradiation modes upon the moulds survival fraction in waste wheat flour

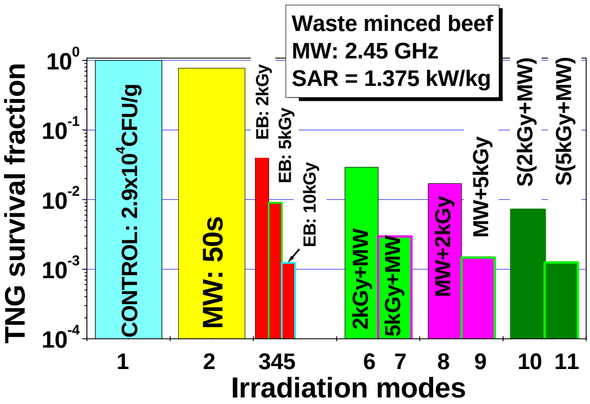

Fig. 10. The effect of different irradiation modes upon the TNG survival fraction in waste minced beef

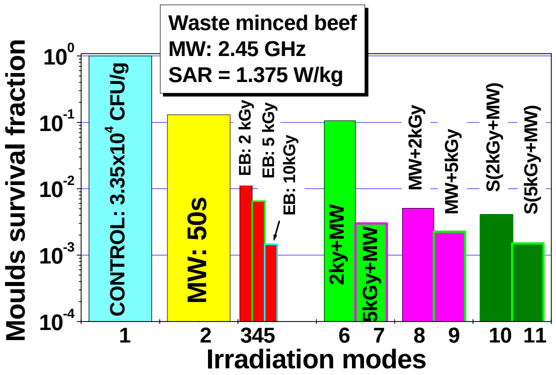

Fig. 11. The effects of different irradiation modes upon the moulds survival fraction in waste minced beef

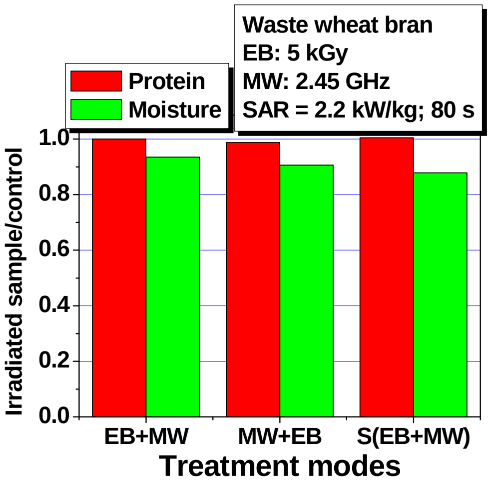

Fig. 12. The effects of combined EB and MW irradiation on wheat bran protein and moisture

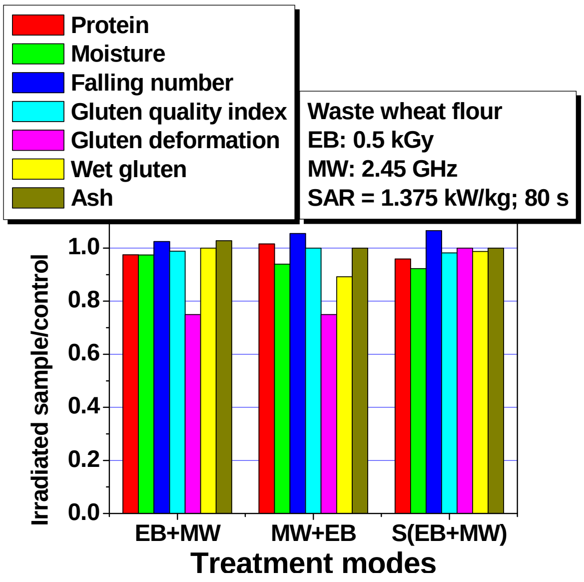

Fig. 13. The effects of combined EB and MW irradiation on wheat flour quality indicators.

Table 1: Samples of waste wheat bran treated with S(5 kGy + 2.2 kW/kg x 80 s) in the first month and then tested monthly during 7 months

Month	NTG (CFU/g)		Moulds (CFU/g)	
	Control	Treated sample	Control	Treated sample
1	2.3×10^6	20	5100	<10
2	2.1×10^6	250	5500	<10
3	1.3×10^6	260	21500	<10
4	3.9×10^5	330	10350	<10
5	7.7×10^5	65	4550	<10
6	8×10^5	55	6000	<10
7	9×10^5	35	5500	<10

4 Conclusions

Simultaneous EB and MW irradiation followed by successive MW and EB irradiation produce the maximum reduction of TNG and moulds in waste wheat bran, waste wheat flour and waste mince beef. In our opinion, it seems that microwave irradiation may modify the microorganisms sensitivity to EB irradiation and thus the application of successive MW+EB and simultaneous MW and EB irradiation lead to greater lethal effects than EB irradiation alone. Also, it seems that the bio-chemical reactions driven by combined EB irradiation and MW heating, complete and sustain each other at a higher rate level especially in the case of S(MW+EB) applied to waste food processing.

The most important conclusion is that the upper limit of EB required absorbed dose, which ensures a complete sterilization effect, could be reduced by a factor at least of two by an additional use of MW energy and to EB irradiation. 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 waste food irradiation followed by composting could be developed to produce disinfected compost and then to recycle this product for agriculture. Recycling means taking a product or material at the end of its useful life and turning it into a usable raw material to make another product [35, 36]. Reducing use of waste materials, designing for reuse and recyclability, is one of the many opportunities for companies to become better environmental stewards of their products [35, 36].

A technical and economic comparison between EB irradiation and microwaves treatment for industrial applications is given as a preliminary analysis in [1]. With reference to EB industrial applications there is a large experience in material processing by electron accelerators: at present time over 1500 electron accelerators are running on a wide spectrum of material (cable, wires, rubber, wood-plastic, etc.) and about over 30 on sterilization process (medical, surgical materials, etc). Also, successful implementation of microwave energy has been demonstrated in numerous industries, including large volume applications such as rubber vulcanization, lumber production, ceramic manufacturing, and food processing [37]. MW energy represent a unique source with the potential of providing a highly flexible technology for a) minimizing generation of selected future wastes, b) reducing existing wastes and immobilizing hazardous components and reclaiming or recycling reusable and sometimes valuable components located in waste products [37]. Microwave heating (dielectric heating) can strongly support the treatment of waste streams, such as sludge processing. The costs for conventional technology vary between USD 40 and 500 per waste tonne [20]. The EB processing costs are estimated to USD 800 -1000 per waste tonne. The dielectric process costs are estimated to be in the range of USD 70-100 per waste tonne [20]. However, the MW process costs are able to sterilize only if the waste is without metallic materials inside: this required the handling of the waste trough several operation not necessary in the case of EB treatment. Finally, the economic analysis shows that both technologies, EB process and MW heating are an interesting alternative to classical treatments. In the case of combined EB and MW process, the required EB dose level and irradiation time is about 2 times reduced in comparison with separate EB process. Under these conditions, the combined EB and MW process costs could be reduced at half from separate EB process costs.

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