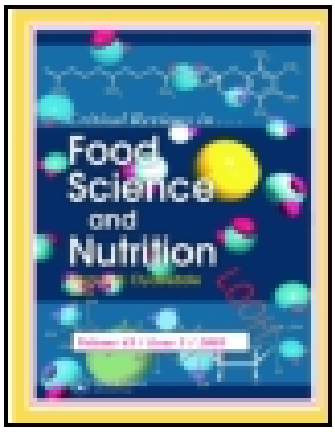

Critical Reviews in Food Science and Nutrition
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/bfsn20

Surface processing: existing and potential applications of ultraviolet light

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Accepted author version posted online: 24 Aug 2013.

To cite this article: Lara Manzocco & Maria Cristina Nicoli (2013): Surface processing: existing and potential applications of ultraviolet light, Critical Reviews in Food Science and Nutrition

To link to this article: http://dx.doi.org/10.1080/10408398.2012.658460

Surface processing: existing and potential applications of ultraviolet light

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Abstract

Solid foods represent optimal matrices for ultraviolet processing with effects well beyond non thermal surface disinfection. UV radiation favours hormetic response in plant tissues and degradation of toxic compound on the product surface. Photoinduced reactions can also provide unexplored possibilities to steer structure and functionality of food biopolymers. The possibility to extensively exploit this technology will depend on availability of robust information about efficacious processing conditions and adequate strategies to completely and homogeneously process food surface.

Key-words: UV-C, photoreaction, hormesis, polymer structure.

THE SURFACE ISSUE

Surface of solid food is the primary location for microbial access and quality depletion during manipulation. For instance, microbial contamination of ready-to-eat foods is mainly distributed on their surface. Enzymatic activities, oxidation and dehydration mainly take place at the product surface, where the loss of biological and mechanical barriers as well as the contact among reactants induce changes in colour, texture, flavour and nutritional properties. Table 1 shows some examples of foods with surface deterioration issues. Not only do they include ready-to-eat food but also raw material intended for further processing or preparation at industrial, catering or home level. Eggs are typically characterised by surface contamination. This issue is not negligible in low moisture products, such as powders and grains whose surface is eventually contaminated by mycotoxins.

Food manipulation increases the risk of deterioration at the product surface but its internal part presents lower quality criticism, being often regarded as sterile. For this reason, non thermal surface disinfection, even though not volumetric, could be sufficient to ensure product safety and shelf life extension. They should be invisible: mild enough to not impair the sensory attributes of the product (Gómez-López, Ragaert, Debevere, and Devlieghere, 2007).

SURFACE PROCESSING

Product surface is the target of a number of different processes performed to delay food spoilage phenomena by removing or inhibiting their causes. Table 2 schematically shows the main surface technologies applied in the food sector, subdivided by the nature of their mechanism of action.

Physical surface processes include traditional water washing or air blowing treatments performed to remove undesired materials that contaminate food surface. Physical protection of food surface is also the basis of packaging technologies, including edible coating strategies. In these cases, packaging is used to isolate the product surface from microorganisms and environmental chemical contaminants, and to protect it from mechanic damage.

Curing and bioprotection can be regarded as biological strategies affecting food surface; the product surface is biologically modified by favouring the development of the natural occurring micro ecology or by inoculating selected microbes with specific performances.

Chemical surface processes are based on the modification of the composition or redox potential at food surface. Dipping or spraying with solutions containing substances with antimicrobial activity (e.g., ethanol, lactate, acetate, sodium benzoate, potassium sorbate, nisin, herbal antibacterial) or contrasting enzymatic and physical deterioration (e.g., citric acid, ascorbic acid,

calcium chloride) represent traditional and efficient surface processes that lead to the modification of the coimposition of food surface. Solutions with high oxidative potential, such as those containing chlorine or hydrogen peroxide, are historically exploited to sanitise food surface. Despite their efficacy, awareness about the toxicity of sanitizers residues is increasing (Hekmati and Bradley, 1979). For this reason, advanced oxidation processes begetting small amounts of persistent chemical species are increasingly demanded. Strategies based on different chemical and physical principles have been developed to deliver reactive oxidant components and free radicals on food surface. Oxidative compounds can be formed in water solution by electrolysis, ultrasound cavitation or photocatalysis, and vehiculated on the product surface by washing solutions (Kokesi et al., 2004; Guentzel et al., 2008). They can also be generated by ozonation, γ-irradition or plasma technologies, and projected onto the target surface (Mahapatra et al., 2005; von Keudell et al., 2010). Ultraviolet radiation and pulsed light technologies are surface oxidation processes combining different actions: (i) direct oxidation of surface molecules through photon impact; (ii) formation of limited amounts of oxidant molecules in the atmosphere contacting the surface; (iii) micro-metric scale thermal effects by electron excitation of food surface components.

Some surface processing technologies are based on concomitant exploitation of both physical and chemical actions. When food is properly isolated from the environment by an adequate physical barrier (e.g., storing cell, package, edible coating), the chemical and redox potential of the atmosphere in contact with the product surface can be modified and eventually controlled, thus significantly increasing food stability. In this case, the modification of the redox potential of

the atmosphere is combined with the inclusion of active molecules in the atmosphere or into the physical barrier (e.g., antimicrobials, antioxidants, volatile absorbers).

The modification of food surface also results from heat treatments, including cooking, baking, or blanching. Thermal modification of cellular biopolymers induces alteration of microbial activity and of tissue physiological metabolism at the product surface. In addition, collapsed, dehydrated or glassy crusts formed upon intense heating are actually edible coatings, which limit moisture migration and control the development of alterative reactions, due to their structure and chemical composition.

ULTRAVIOLET PROCESSING OF SOLID FOOD

Ultraviolet processing of solid food is U.S. FDA-approved (U.S. FDA 2002) and does not require authorisation according to EC novel food regulation. Ultraviolet radiation is easy to use and characterized by favourable costs of equipments, energy and maintenance (Barbosa-Canovas et al., 1998; Miller et al., 1999; Bintsis et al., 2000). It is safe to apply but some simple precautions are necessary to avoid worker exposure to light and evacuate the generated ozone. Moreover, the technology has not been reported to form known toxic or significant non-toxic by products (Keyser et al., 2008).

Ultraviolet radiation in the electromagnetic spectrum from 200 to 280 nm can be generated by low-pressure mercury vapour fluorescent lamps with transparent and colourless quartz tubes. These lamps emit 95% of their energy in the 254 nm range (UV-C). More recently, high intensity lamps have been developed with enhanced potential for UV inactivation of bacteria (Ngadi, et al., 2003). They have made UV treatments relatively inexpensive and readily available.

Classification of ultraviolet radiation sources according to emission spectrum and predominant wavelengths was recently reported by Falguera et al. (2011a).

Ultraviolet processing requires exposure of the entire food surface to the radiation emitted by the UV lamps. Researchers mainly operate using static but versatile systems. Low pressure mercury lamps are placed above flat foods (e.g., ham, cheese, bread slices) or flowing powders. In the case of non-flat foods (e.g., bread, sausages), the geometry of lamp arrangement depends on product size and shape.

Ultraviolet radiation units can be subdivided depending on the presence of a food support during the treatment (Fig. 1). Flat products are placed on a support below the lamps. After a certain time, product is turned to allow exposure of both sides (Fig. 1A). They can also be allocated on a supporting net or a film, which minimise blockage of UV radiation (Fig. 1B). In this case, product turning is not required. Additional lamps or mirrors can be placed on the product sides to increase exposure efficiency (Fig. 1C). When no food support is present, the product flows near the lamps in a vessel containing water. Lamps are inserted in waterproof quartz tubes, and the product is eventually mixed by air blower units (Fig. 1D). The water stream can be replaced by an air flow that allows the formation of a fluidised bed of food particles. Commercial equipments are based on the same concepts, but the process is made continuous substituting the static food support (Fig. 1A, B, C) with conveyor belts, or changing the vessels (Fig. 1D) into tunnels that allow food movement through the ultraviolet radiation unit.

The intensity of UV radiation is expressed as irradiance, Wm^{-2}. Irradiance is generally modified using lamps with different power or, more often, changing the distance between lamp and product surface. The longer the distance, the lower the light irradiance. Irradiance in a given

point of the product surface can be easily measured using UV radio meters that are increasingly common. Treatments are generally compared considering their light dose (J m^{-2}) that accounts for both radiation intensity and exposure time. It must be observed that the same dose can be obtained under completely different processing conditions. For instance, a dose of 1 kJ m^{-2} can be equally obtained by 1 min exposure to 1000 W m^{-2} UV light or by 60 min exposure to 17 W m^{-2} UV light. Even though the UV dose is the same, the time-scale of the phenomena affected by irradiation could significantly affect the overall efficiency of the process.

Unpacked food with high moisture is often treated in ultraviolet radiation units without temperature and moisture control. Commonly applied doses do not significantly modify the overall temperature of the product. However, long treatments performed in small chambers can develop heat, increasing the air temperature in the chamber. In this case, moisture loss and temperature increase of the product could be not negligible.

Product dehydration and after exposure contamination can be avoided by packing food in materials not shielding UV radiation. Light transmittance measures performed in our lab showed that most packaging materials are not permeable to ultraviolet radiation (Table 3). Some polymers are able to transmit most of the incident UV radiation. However, they lose this capacity when sealed. Sealing may thus provide critical areas that shield UV radiation and decrease the actual dose delivered on the product surface. UV light permeability data of packaging materials, and methodologies to increase it, are either scarce or incomplete. Exploitation of ultraviolet radiation to process packed food will strongly depend on the availability of technological solutions to produce UV transparent materials with adequate mechanical and gas barrier properties.

SURFACE DISINFECTION BY ULTRAVIOLET RADIATION

The antimicrobial effects of UV light are well known to be primarily mediated through absorption of highly conjugated double-bond systems in biomolecules (Koller, 1965; Smith and Hanawalt, 1969; Miller et al., 1999). UV light is able to damage microbial DNA, causing cross-linking between neighbouring thymine and cytosine in the same DNA strand (Rames et al., 1997). DNA transcription and replication are thus blocked, compromising cellular functions and eventually leading to cell death (Sastry et al., 2000). Inactivation of viruses is also due to damage of the genomic material but, at high doses, UV radiation can affect the capsid proteins (Sommer et al., 2001).

Disinfection by ultraviolet radiation has the undoubted advantage of not requiring heat or chemicals (Devine et al., 2001). For this reason, UV treatments are widely used for sterilisation purposes of equipments, devices, packaging and many other non food surfaces intended to come into contact with food or for medical use (Bintsis et al., 2000; Guerrero-Beltrán and Barbosa-Cánovas, 2004). By ensuring accurate enlightening of the surface, more than 5 log reductions in microbial counts are observed on stainless steel and plastic materials exposed to UV light doses between 2 and 4 kJ m^{-2} (Kim et al., 2002; Sommers et al., 2009). Long life low pressure mercury lamps are generally used to these purposes. Air-cooled UV cassettes emitting an intense radiation can be installed in filling equipments as well as for the disinfection of conveyor belts, transport containers and working surfaces.

Due to its poor penetration depth, UV radiation is traditionally applied to disinfect transparent fluids (Bintsis et al., 2000). It is commonly used to disinfect water or air intended to come into

contact with food. UV processing has also been proposed to extend shelf life of sugar syrups, apple cider and fruit juices (Nakayama and Shinya, 1981; Stother, 1999; Tran and Farid, 2004; Choi and Nielsen, 2005). However, the presence in the fluid of soluble solids, absorbing and scattering light, reduces the penetration of the radiation and thus the efficacy of the process (Guerrero-Beltrán and Barbosa-Cánovas, 2004; Ye et al., 2007). In the case of apple pulp, the penetration depth, defined as the depth at which the light irradiance inside the material (I) falls to $1/e$ of the intensity of the incident light at its surface (I_o), resulted 0.2 mm (Manzocco et al., 2011b). This evidence shows that light exposure of solid foods affects only a thin surface layer of the product, while a negligible light dose is able to reach its internal part.

Pulsed light is a modified and claimed improved version of delivering ultraviolet radiation to bodies (Gómez-López et al., 2007). The treatment is based on short xenon lamp flashes of an intense broad spectrum, rich in UV light (Guerrero-Beltràn, and Barbosa-Cànovas, 2004, 2006; Gómez-López et al., 2007). High energy doses allow the intensity of the light penetrating opaque materials to be increased, thus extending the potential of application from transparent liquids to those containing particles. If high penetration intensity is required to inactivate microorganisms in a fluid mass, it can be an unnecessary feature when treating foods that exert surface contamination issues. This is particularly critical for products required to maintain their original fresh-like appearance. Application of pulsed light to food surfaces, beside being expensive and requiring authorisation according to the EU novel food regulation, is actually limited by the intense photothermal effect that can result in surface temperatures up to 120 °C (Jun et al., 2003). In addition, off-odours are often developed upon pulsed light treatments, even if they may subsequently fade (Gómez-López et al., 2005).

Many research groups have provided evidence about the efficacy of UV radiation on a number of different products but difficulties arise when comparing data. This is due to differences in UV treatment units and incomplete communication of processing parameters. Table 4 summarises the main literature data relevant to the efficacy of UV radiation in inactivating microorganisms on the surface of different foods. Although different food matrices were studied, data showed in Table 4 indicate that the surface germicidal effect of UV light seems independent on the light dose applied. Despite a minimum light dose is required (0.4 kJ m^{-2}), there is not a clear relation between dose and inactivation effect. To analyse the effect of surface properties on the efficacy of UV treatments, data shown in Table 4 were elaborated reporting the median value as well as minimum and maximum values of logarithmic reductions achieved for the different products (Fig. 2). Logarithmic reductions higher than 2 are generally observed for smooth skinned fruits (i.e., apple, tomato, pepper) and occasionally for shell eggs. Lower germicidal effects are achieved for products with rough surfaces. In order for UV light to kill microorganisms, photon impact on microbial cells must occur. Therefore, microorganisms will survive if they are shadowed by any body between them and the light source. To this regard, top dirty particles (e.g., faeces on eggshells, soil residues on fruit and vegetables) represent protective layers for the microbes. In general terms, low germicidal effect of ultraviolet radiation on food surface can be attributed to the combination of factors associated to both microorganism and food characteristics (Table 5). Impact of UV light on microrganisms can be difficult because of their physical location. They can be internalized at a thickness which is not exposed to a large enough light dose. They may also be protected from light by other cells, as occurs when biofilms are formed. In the case of chicken breast, 10-40 kJ m^{-2} UV light exposure was associated to a

decontamination effect lower than that observed in another trial at 1-3 kJ m^{-2}. This was attributed to the possible formation of protective biofilms due to the prolonged time span between inoculum and treatment (Sommers et al., 2010). Additional factors limiting the efficacy of light are due to food shadowing effects. Not only may food pieces shadow each other but also macro-microscopic surface irregularities can lead to local shadowing. Food surface topography plays an important role in the efficiency of UV disinfection (Woodling and Moraru, 2005; Allende et al., 2006). For instance, tissue wounding or product physical damage caused by food manipulation can offer protective places for microorganisms to survive the treatment and subsequently grow. Stermer et al. (1987) found that the bactericidal effect of UV light was less effective on rough meat surface because bacteria were partially shielded from the radiation. Similarly, Lopez-Rubira et al. (2005) attributed to surface topography unclear effects of UV radiation on microbial population of pomegranate arils. Critical factors also involve light transmission properties of food. Its scattering properties affect the penetration depth of the radiation and the light dose that actually reaches microorganisms situated in surface irregularities. Dark foods absorb higher amounts of light, decreasing the energy available for microbial inactivation. The effect of food composition is also not negligible since organic solutes and macromolecules cause strong UV attenuation effects.

EFFECT OF ULTRAVIOLET RADIATION ON INTACT PLANT TISSUES

Hormesis can be defined as a stimulation of beneficial plant responses by low or sublethal doses of a chemical inducer or a physical stressor (Luckey, 1980). In this conditions, the plant is submitted to an eustress, which is considered a positive form of stress. On the contrary, distress

commonly implies negative physiological implications for the plant due to adaptation mechanisms to the stressor. Although ultraviolet irradiation is usually considered an environmental distress causing physiological damage to the plant tissues, low levels of UV energy have been reported to exert eustress effects, reducing postharvest decay in a number of fresh fruit and vegetables (Stevens et al., 1996). It is noteworthy that only fragmentary indications are available about the UV dose discriminating eustress from distress. Literature data (Liu et al., 1993; Lu et al., 1987; Stevens et al., 1990; Wade et al., 1993; Baka et al., 1999; Marahaj et al., 1999; Ait-Barka et al., 2000a, 2000b; González-Aguilar et al., 2001, 2007; El Ghaouth et al., 2003; González-Aguilar et al., 2004; Allende et al., 2006; Costa et al., 2006; Lemoine et al., 2007) associate hormetic responses to UV doses ranging from low values (0.03 J m^{-2}) up to levels difficulty exceeding 5-7 J m^{-2} (Fig. 3). From a technological point of view, both eustress and distress effects of ultraviolet radiation could be exploited to obtain plant tissues responses that can help to increase the product quality and nutritional value or to prolong its fresh-like appearance.

UV eustress

UV eustress has been proposed as an efficient technology to delay ripening and senescence of intact fruit and vegetables, extending their shelf life. In the case of tomato, climacteric was delayed, retarding development of red colour and softening (Liu et al., 1993; Marahaj et al., 1999; Ait-Barka et al., 2000a, 2000b). Slower senescence rates were observed in onions (Lu et al., 1987), potatoes (Stevens et al., 1990), broccoli (Costa et al., 2006; Lemoine et al., 2007), strawberries (Baka et al., 1999), peaches (El Ghaouth et al., 2003; González-Aguilar et al., 2004) and mango (González-Aguilar et al., 2001;2007) among others.

These effects were attributed not only to the germicidal activity of UV radiation, which reduces pathogen propagation on product surface, but also to induced plant tissue resistance to microbial spoilage. The latter is probably mediated by the modification of physiological processes, such as respiration, germination, growth and development. Induced formation of phytoalexins (Bridge and Klarman, 1973; Devlin and Gustine, 1992; Lamikanra, and Richard, 2004), activation of genes encoding pathogenesis-related proteins (Green and Fluhr, 1995) and lipid peroxidation markers (González-Aguilar et al., 2007) have been reported. Common plant responses to hormic UV treatments include: (i) increased levels of phenilalanine ammonia-lyase activity that enhances the biosynthesis of phenols toxic to pathogens; (ii) reduced ethylene production, and (iii) development of antisenescent putrescine exerting opposite physiological effects to ethylene (Stevens et al., 1998; Maharaj et al., 1999).

The eustress effects of ultraviolet radiation can also be exploited to steer the nutritional and sensory properties of fruit and vegetables. For instance, the increase in polyphenols and flavanoids upon UV irradiation has been claimed as a possible strategy to enhance the antioxidant activity of fruit and vegetables (Lemoine et al., 2007; Alothman et al., 2009; Artés-Hernández et al., 2010). Additional exploitation of UV irradiation has been proposed to increase nutritional value of mushrooms. UV irradiation can actually convert ergosterol, the most abundant phytosterol in mushrooms, to a variety of photoirradiation products, including previtamin D_2 that undergoes spontaneous thermal rearrangement to vitamin D_2 (Jasinghe and Perera, 2006; Teichmann et al., 2007).

Effects of UV exposure on volatile aroma profile in tropical fruits were also demonstrated. UV radiation increased the intensity of melon cubes flavour (Manzocco et al., 2011a). Melon

terpenoids, such as geranylacetone, β-cyclocitral and β-ionone, can significantly increase in direct response to UV light stress (Lamikanra et al., 2002; Beaulieu, 2007). By contrast, esters may decrease only following prolonged UV-light exposure of thinly sliced laminar tissues. Low energy UV radiation could contribute to enhance the overall terpenoid concentration but more intense treatments could result in excessive endogenous terpenoid concentrations, and thus in off-flavour development (Beaulieu, 2007).

UV distress

UV treatments at distress conditions are usually associated to severe cell damage, which rapidly turns into a depletion of the fresh-like appearance of the product. These effects are expected in both intact plant tissues and in fresh-cut produce, although through different mechanisms. UV abiotic distress promoted visible damage in intact fruits such as peaches, grapefruit and citrus (Stevens et al., 1996; Rodov et al., 1992). In lettuce, 7.1 kJ m^{-2} UV irradiation induced tissue softening and increased browning during storage (Allende et al., 2006). A dose of 24.4 kJ m^{-2} provoked ripening and caused abnormal browning, manifested as sun-scalding of the tomato fruit's surface (Maharaj et al., 1999). Damage generally appears after *circa* 24 hours from irradiation as a superficial bronzing due to deposition of a red-brown pigment in epidermal and adjacent hypodermal cells (Wade et al., 1993). The development of browning in intact plant tissues is not closely related to the evolution of oxidative enzymes but to the decrease of chlorophylls content and occurrence of noticeable modifications at the ultra-cellular levels (González-Barrio et al., 2005; Hosseini Sarghein et al., 2011; Najeeb et al., 2011).

EFFECT OF ULTRAVIOLET RADIATION ON WOUNDED PLANT TISSUES

A certain level of physiological response to sublethal ultraviolet stress can also be observed in wounded plant tissues (i.e., fresh-cut produce). It is likely that the mechanisms governing the response of wounded tissues to ultraviolet radiation could be considerably different from those observed in intact fruit and vegetables. Understanding the mechanisms of wounded tissue response to ultraviolet radiation is required to exploit possible eustress mechanisms in fresh-cut plant tissues. However, at the moment, little information is available.

In fresh-cut fruit and vegetables exposed to high UV doses, the occurrence of cell damage is partially attributable to the breakage of cellular membranes that causes a loss of cell compartmentalisation (Wang et al., 1993; Gómez et al., 2010; Manzocco et al., 2011b). Intense irradiation of cut apple caused membrane rupture, decrease in intracellular volume and loss of cell turgidity (Fig. 4). As a result, enzyme-substrate contact increased with acceleration of enzyme catalysed reactions. Gómez et al. (2010) showed that apple slices exposed to doses in the range from 5.6 to 14.1 kJ m^{-2} turned darker than the control. Additional consequence of cell membrane modification is the leakage of intracellular water which tends to quickly evaporate following local thermal effects due to UV electron excitation of food surface components (Manzocco et al., 2011b).

Exploitation of UV distress to increase fresh-like appearance of cut fruit and vegetables

The intense damage induced by ultraviolet radiation at distress doses can be successfully exploited to increase the fresh-like appearance of cut fruit and vegetables. Ultraviolet radiation can promote dehydration of a thin surface layer of the wounded plant tissue, begetting a protective edible film that hinders juice leakage and dehydration during refrigerated storage

(Manzocco et al., 2011a; 2011b). It is also likely that surface dehydration could reasonably account for local modification of water availability, inhibiting microbial growth and enzymatic activity during storage. These effects would be strictly dependent on the applied dose. Fig. 5 schematically represents the effect of UV dose on the occurrence of different events conditioning browning rate during storage of fresh-cut apple. At low UV doses, polyphenoloxidase photooxidation can be regarded as the prevalent event. At high doses, extensive cell degradation occurs with reactant release and intense browning. In this condition, it is not excluded that phenols could be directly photooxidised independently on enzyme catalysis. Intermediate UV doses can exert a synergic effect on browning rate, promoting both enzyme inactivation and surface dehydration. In the case of fresh cut apple, the dose associated to minimum browning during storage was found to be around 1.2 kJ m^{-2}. It is likely that optimal light doses for surface dehydration are critically affected by tissue structure and moisture level. However, no information is available in literature.

EFFECT OF UV RADIATION ON BIOMOLECULES

A number of different molecules exerting nutritional (e.g., fatty acids, vitamin A, riboflavin,) or sensory effects (e.g., carotenoids, betanine, melanoidis) as well as showing toxicity (e.g., mycotoxins, acrylamide, pesticides) present absorption spectra with peaks in the UV radiation range. From a theoretical point of view, irradiation at these wavelengths, especially when performed in the presence of molecular oxygen, is able to activate photoisomerisation and oxidative reactions, potentially leading to the molecule degradation.

In the case of liquid food, oxidative reactions induced by UV irradiation have been reported in a number of different products, conditioning their nutritional value and causing oxidation of lipids, colour fading or development of off-flavours (Attoe and von Elbe, 1981; Beckbolet, 1990; Rafsgaard et al., 1993; Ramírez et al., 2001; Whited et al., 2002; Tran and Farid, 2004; Ibarz et al., 2005; Manzocco et al., 2008; Falguera et al., 2011b).

When dealing with solid foods, oxidation of molecules with nutritional or sensory relevance is not a critical issue. Their eventual decrease in a thin surface layer does not affect their concentration in the overall product. For instance, no changes in colour of carotenoid containing solid food (i.e., melon cubes) were detected, although these pigments are well known to be photosensitive (Pesek and Warthensen, 1987; Manzocco et al., 2011a). By contrast, different undesired molecules, which can be potentially degraded by UV irradiation, are mainly located on the product surface. This suggests that this technology could be used for solid food surface decontamination. As an example, aflatoxins, which tend to accumulate on the surface following microbial growth, have been shown to degrade by UV radiation in both peanuts (Shantha and Sreenivasa-Murthy, 1981) and rice (Nkama et al., 1987; Nkama and Muller, 1988). An accurate review of the effects of UV irradiation on mycotoxins has been recently produced by Falguera et al. (2011b). UV radiation is also known to degrade melanoidins (Kwak et al., 2004), and could be exploited to decompose or polymerise toxic Maillard reaction products such as acrylamide. The latter mainly forms on product surface due to the high temperature reached in these areas during intense heat treatments (Taeymans et al., 2004). Similarly, pesticide residues, such as pyridine and thiabendazole, have also been reported to decompose upon exposure to UV irradiation (Ibartz et al., 1985; Panadés et al., 1997). Pesticides are seldom applied by spraying

and tend to accumulate on the product surface, making UV treatments potentially applicable for decontamination.

Effect of ultraviolet radiation on food biopolymers

Organic polymers are well known to undergo chemical modification when they are exposed to light. Polymer photoreaction is far from being a novel technology. Ancient Egyptians and Babylonians used to photoreact linen during mummification, and waterproof papyrus boats by polymerisation of asphalt oil (Decker and Bendaikha, 1998; Wondraczek et al., 2011).

The common feature of photoresponsive polymers is the presence of a chromophore incorporated in the macromolecular matrix (Wondraczek et al., 2011). Fig. 6 schematically shows the main photoinduced polymer reaction. At first, the electromagnetic energy is captured by the chromophore and converted to a chemical signal due to photoisomerisation. The latter allows the chemical signal to be transferred to the functional part of the polymer that controls its overall properties. Absorption of electromagnetic energy and relevant electronic transitions are thus responsible for a series of modification in the polymeric material. As a consequence, conformation and size of the polymer can deeply change, potentially leading to modifications in its functional properties and biological activity. For instance, unfolding or dissociation of proteins could beget systems with well-defined water absorption capacity, interface behaviour, rheological and mechanical properties as well as with steered biological activity (e.g., enzymatic activity, digestibility, allergen reactivity). According to the molecular changes occurred, relevant modifications in the architectural organisation of the polymeric units could be expected. These modifications would affect the optical and mechanical properties of the material they are embedded in. Polymer crosslinking or grafting with specific functional groups could allow

formation of tridimensional networks. These novel architecture would stabilise supramolecular structures and immobilize biomolecules with the desired nutritional or health protecting role.

Most research carried out on photoinduced reactions is relevant to synthetic polymers as, in this sector, the number of exploitable chromophores is limitless. This makes UV curing one of the most efficient processes to quickly produce polymeric materials with the desired properties. For instance, coatings, adhesives and printing plates are based on polymer photoreactions. However, food biopolymers such as proteins and polysaccharides can also undergo photoinduced reactions.

Effect of ultraviolet radiation on proteins

Proteins are major targets for photoinduced modifications due to the abundance of endogenous chromophores within their structure. Both amino acid side-chains (e.g., thriptophan, tyrosine, phenylalanine, cysteine) and bound prosthetic groups (e.g., flavins, heme) may act as efficient chromophores. Proteins have the additional ability to bind exogenous chromophores, and rapidly react with other excited state species (Davies and Truscott, 2001). Based on these considerations, protein photo-oxidation is generally reported to occur via two major routes: (i) direct photo-oxidation arising from the absorption of radiation by the protein structure or bound chromophore; (ii) indirect protein oxidation mediated by singlet oxygen generated by energy transfer by either protein bound, or other chromophores (Davies and Truscott, 2001). The result would thus be a modification in the protein properties due to side-chain oxidation, backbone fragmentation and/or formation of cross-links and aggregates (Davies, 2003). For instance, UV irradiation is applied to photocrosslink collagen to modify the biodegradation rate, and the tensile properties of collagen-based surgery implants (Tomihata et al., 1994). Application of UV irradiation has also been proposed in the food sector. UV induced crosslinking was observed in

gluten, zein, and albumin films (Rhim et al., 1999). Kuan et al. (2011) also showed that photocrosslinking of egg white protein and sodium caseinate improved emulsifying and foaming properties. In addition, dry granules of UV irradiated fish gelatin exhibited improved gel strength, marked reduction in viscosity, and significant changes in melting temperatures (Bhat and Karim, 2009).

- *Enzymes*

The biologic activity of proteins actually depends on their structure that can be modified following photo-oxidation promoted by ultraviolet light exposure (von Tappeiner, 1903; Rhim, Gennadios, Fu, Weller, and Hanna, 1999; Edwards and Silva, 2001). The loss of polyphenoloxidase activity upon UV exposure was attributed to aggregation phenomena, while cleavage into fragments without catalytic activity was mainly observed after UV irradiation of pectin lyase (Manzocco et al., 2009a; 2009b). Sensitivity of enzymatic proteins to UV irradiation is strongly dependent on their nature. Under the same conditions, pectin lyase inactivation upon UV exposure was quicker achieved as compared to that of polyphenoloxidase. The latter was however more sensitive to the increase in irradiance dose. Not only each enzyme exerts its own UV sensitivity, but also isoenzymes can exert different susceptibility to UV exposure. For instance, UV resistant forms of pectolytic enzymes were identified in apple (Manzocco et al., 2009a). The dose of UV irradiation exerts a critical role. Low intensity UV treatments can modify the conformation of the active site of the enzyme leading to an overall activity increase. By contrast, higher intensity treatments would be associated to non-reversible structural changes resulting in enzyme inactivation.

The complex effects of UV irradiation on enzymes give probably reason for the controversial information available in literature, mostly referring to enzymes present in liquid matrices. Pectin methyl esterase in orange juice was indicated to be not affected by UV treatment, whilst polyphenoloxidase in mango nectar and apple juice as well as alkaline phosphatase were reported to be inactivated (Keyser et al., 2008; Tran and Farid, 2004, Guerrero-Beltrán and Barbosa-Cánovas, 2006; Manzocco et al., 2009b). Even less information is available on the effect of UV irradiation on enzymes present in solid food. UV light exposure has been demonstrated to exert contradictory effects on the activity of different enzymes in fresh-cut melon (Lamikanra et al., 2005). In this case, authors interest was focussed on systemic response to wound stimulus in the presence of UV light. Enzymatic activity was thus analysed on the extract of the entire fruit piece, thus hiding the eventual local effect of irradiation on the surface. Experiments carried out in our lab confirmed the capability of ultraviolet radiation to inactivate enzymes on fresh-cut fruit surfaces. The activities of pectolytic and polyphenoloxidase extracted from a thin (0.1 mm) surface layer of UV treated apple slices were actually reduced by more than 60% (Table 6).

When enzymatic activity responsible for fruit quality depletion is mainly localised on the wounded surface (e.g., enzymatic browning), UV treatments could find application to selectively inactivate browning enzymes on the product surface without affecting the interior tissue, and guarantying the required fresh appearance. Based on the significant fluctuations in UV light response of the different enzymes, further research is needed to determine proper operative parameters to selectively promote the inactivation of the target enzymes, minimising undesired physiological effects.

- *Allergens*

Thanks to its potential ability to alter the structure-related biological properties, UV radiation could be valuable to modify the antigenic response of protein allergens. However, little data is available in literature. Liquid peanut butter and soybean extracts were shown to decrease their IgE binding capacity by exposure to pulsed light (Chung et al., 2008; Yang et al., 2010). The treatment led to an increase in sample temperature. Discrimination between the photooxidative effect and the thermal one was not clear. Manzocco and Nicoli (2012) showed that a ten-fold decrease of immunoreactivity of egg white protein was obtained by its exposure to *circa* 12 kJ m^{-2} UV radiation at room temperature. The loss of immunoreactivity was attributed to denaturation phenomena with formation of protein fragments partially retaining the original epitopes. Further research is needed to explore the possibility to use UV radiation to control/reduce immunoreactivity of ingredients or minimise cross contamination in production plants.

- *Effect of ultraviolet radiation on proteins under crowded environmental conditions*

The effect of ultraviolet radiation on protein structure and functions is generally studied, mainly for practical reasons, in simple buffer systems with low concentration of protein to avoid aggregation phenomena. These are idealized conditions, far from highly crowded environments typical of food surfaces. The sensitivity of proteins to ultraviolet radiation was found to be strongly affected by their concentration (Manzocco and Nicoli, 2012). In the case of egg white proteins, photoreactivity was detected only below a critical concentration that was associated to macromolecular crowding. By contrast, proteins were fully resistant to ultraviolet radiation in crowded environments. In other words, in highly concentrated systems, proteins lose their photosensitivity since reactions are under macromolecular crowding control (Minton, 1983;

Zimmermann and Trach, 1991). It can be hypothesised that macromolecular crowding could be selectively exploited to steer the photosensitivity of proteins by proper food formulation strategies. These could include both the modification of protein concentration and the addition of specific crowding agents. When the aim is structure modification (i.e., unfolding, cleavage, cross-linking) to achieve peculiar biological and functional properties, proteins should be light treated in the presence of negligible crowding effects. If the goal is microbe killing and protein structure preservation, they should be enlightened in crowded environments (Fig. 7). Nevertheless, at our knowledge, no further information about the role of macromolecular crowding on light-induced modification of protein structure is available in literature.

Effect of ultraviolet radiation on carbohydrates

Pure glucans, such as cellulose or amylose, are poor absorbers of UV radiation. Cellulosic pulps and starch slurries may however contain different chromophores. The latter are formed upon thermal, acid, basic or oxidative treatments performed during polysaccharide production and hydrolysis. Non enzymatic browning products could represent efficient chromophores, increasing the photoresponse of polysaccharides. Sensitisers such as sodium benzoate, pigments or vitamins, can activate the polymer and promote photocrosslinking and/or depolymerisation. Crosslinking and depolymerisation of photoactivated starch molecules seem associated to irradiation in the presence of oxygen (Fiedorowicz and Tomasik, 1999). In these conditions, crosslinking mainly involves the amylose fraction while depolymerisation affects both amylose and amylopectin (Fiedorowicz and Tomasik, 1999; Vatanasuchart et al., 2005). Peat and coworkers showed in 1948 that extensive photo-oxidation in the presence of oxygen can result in

complete conversion of amylose to CO_2. Under nitrogen, unidentified final products highly resistant to subsequent oxidation are formed.

Starch photocrosslink is particularly interesting for production of renewable biopolymers to replace petrochemical-based plastics and to develop edible coatings. Photocrosslink of starch blends has been associated to improved mechanic performance, increased pasting temperatures, peak viscosity and setback (Follain et al., 2005). Swelling and solubility appeared to be retarded due to restrained granule hydration (Lee et al., 2005). Photocrosslink also delayed retrogradation of wheat starch films, preventing packing of double helices of amylopectin, and hindering the formation of crystalline clusters (Delvill et al., 2003).

It has been proposed that a certain level of starch depolymerisation could provide small linear fragments of amylose and amylopectin that facilite the formation of an amorphous matrix structure of starch dough during baking. Good baking expansion of cassava starch was achieved by its exposure to UV radiation. However, no effective starch structure for baking expansion was achieved by prolonging the exposure time (Vatanasuchart et al., 2005). Higher UV doses induced extensive starch depolymerisation (Tomasik and Zaranyika, 1995; Bertolini et al., 2000), reducing paste viscosity and melting enthalpy. Arabic gum was also found to develop better emulsifying properties upon UV exposure (Kuan et al., 2009). Even in this case, short UV treatments were associated to crosslinking while longer exposure produced a viscosity break down. Based on the experimental evidences reported in literature, the changes in viscosity of starch pastes and dough produced by UV treated starch could schematically follow a bell-shaped curve as a function of the applied dose (Fig. 8). Unfortunately, little indication about actual UV

doses applied in these experiments is reported in literature, limiting the possibility to compare different treatments and identify the dose associated to the viscosity maximum.

EFFECT OF ULTRAVIOLET RADIATION ON FOOD STRUCTURE

Foods commonly contain mixtures of proteins and polysaccharides embedded in crowded and complex matrices with abundance of different chromophores from both proteins and minor components. There is plenty of possibility to efficiently photoinduce polymer reactions. The latter could be steered by choosing proper process conditions, and exploited to obtain structures with specific functional or nutritional properties. For instance, food exposure to UV radiation can induce its structural modification. This may result in more compact architectural organisations with not negligible effects on rheological and mechanical properties of food. In addition, the combination between structural modification of food surface components and local thermal effects could induce the formation of compact dehydrated surface films. These protective edible films could contribute to seal the product, modifying its performance during processing, storage and home preparation. It is not excluded that UV structured films could account for local modification of water availability, inhibiting microbial growth and enzymatic activity during storage. They could be exploited to steer weight loss kinetics during storage and eventual rehydration kinetics during processing. However, the potentialities of UV induced modification of food physical properties have been scarcely investigated.

FINAL REMARKS

Ultraviolet radiation is nowadays recognised as an efficient technology allowing microbial decontamination of food to be non thermally achieved. Research relevant to UV treatment of

liquid food has clearly demonstrated that this technology is efficacious on transparent liquids but often fails in ensuring adequate decontamination of opaque fluids. This is probably why UV treatment is considered an "old" emerging technology: nothing new seems to be expected from it. However, analysis of literature data shows the wide potential of UV radiation to modify biomolecule structures and thus their functional and physiological performances. These powerful and peculiar properties together with the poor penetration capacity make UV radiation particularly promising for surface processing of solid foods. As a matter of fact, solid foods are characterised by safety and quality issues considerably different from those associated to liquid foods. Food companies are actually aware of it, giving UV processing serious consideration as a cheap and friendly technology for routine decontamination of solid food. However, UV radiation could be also used for additional purposes which arise from its ability to modify structure and functionality of food biomolecules. The induction of polymerisation reactions or eustress/distress effects, the degradation of toxic compounds or allergens are some examples of the unexplored possibilities provided by UV light technology to improve food safety and quality.

The key-point for turning an "old" emerging technology into a strategic one lies in the possibility to demonstrate that safety and quality improvements achieved by using UV processing can be hardly obtained by other technological tools. However to reach this goal the following research needs are still required:

- better understanding of photo-induced chemical and structural changes of food biomolecules;
- studying the functional, sensory, nutritional and physiological properties of photo-modified food biomolecules;

- deeper understanding of the mechanisms controlling plant eustress/distress in both intact and wounded plant tissues

- exploring the effects of ultraviolet radiation on food supra molecular structure, identifying potential technological applications;

- identifying the relation between ultraviolet radiation dose and intensity of the desired/undesired effects of the plant tissues and food products.

However, the exploitation of food surface UV processing requires technological strategies able to efficiently treat 100% of the food surface. Unfortunately present plant solutions are far from being fully efficacious. To plug these gaps the following technological knowledge and requirements are needed:

- better understanding on the factors controlling UV effects on food surface;
- improvement of industrial UV application skills;
- identification of proper operative conditions which can greatly vary depending on product surface characteristics and UV treatment aims.

The possibility to extensively exploit ultraviolet radiation in the future will depend on the availability of robust information about the boundary discriminating technological conditions leading to disadvantageous quality depletion from those imparting beneficial effects.

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FIGURE CAPTIONS

Figure 1: Ultraviolet processing units for solid food.

Figure 2: Logarithmic reductions achieved by exposure of different products to ultraviolet radiation (symbol: median, bar: minimum-maximum interval)

Figure 3: Plant eustress and distress as a function of ultraviolet radiation dose.

Figure 4: Micrographs of fresh apple slices (A) and apple slices exposed to 144 kJ m^{-2} (B).

Figure 5: Schematic representation of the effect of UV dose on the events conditioning browning rate during storage of fresh-cut apple.

Figure 6: Main photoinduced polymer reaction.

Figure 7: Steering protein photosensitivity by crowding effects.

Fig. 8: Schematic representation of the effect of UV dose on starch paste and dough viscosity.

Table 1: Examples of foods showing surface deterioration issues.

	Surface sensitive food	
	Raw materials	Ready-to-eat food
Plant derivatives	Fruits and vegetables	Fresh-cut fruit and vegetables
	Dried fruit and vegetables	Ready-to-eat vegetables
	Grains	
Meat derivatives	Meat carcasses	Sliced ham/roastbeef
	Meat cuts	Cured meat
		Diced meat cubes

		Sausages
Fish derivatives	Fish	Fresh fish fillets
		Ready-to-eat fish fillets
Cheese derivatives	Cheese	Cheese slices/cubes/sticks
Bakery products		Bread loaves/Sliced bread loaves
		Cakes/Cake slices
		Bread sticks
		Biscuits
		Pancakes
Eggs	Shell eggs	Cooked egg (e.g., boiled eggs with or without shell, fried eggs)
Powders	Spices, flour	

Table 2: Nature and mechanism of action of the main food surface processes.

Mechanisms of action	Surface process
Removal of contaminants from food surface	Washing
	Air blowing
Isolation of food surface from the environment	Packaging
	Edible coating
Modification of food surface micro ecology	Curing
	Bioprotection
Modification of composition of food surface	Dipping or spraying with solutions containing antimicrobials or substances

	contrasting enzymatic activity of mechanic damage
Modification of redox potential of food surface	Washing with aqueous solutions containing chlorine or H_2O_2 Advanced oxidation processes Ultraviolet radiation Pulsed light
Modification of composition and redox potential of the atmosphere in contact with food surface	Modified atmosphere packaging Active packaging
Modification of food surface moisture, structure and chemical composition Cellular biopolymer modification	Cooking Baking Blanching Surface pasteurisation Infrared heating

Table 3: Permeability of some plastic materials to UV-C radiation

	Thickness (μm)	UV-C permeability (%)
OPP/PE	60	0
PET/PE	52	0
Polyester	26	0
OPP/CPP	35	0
PP/PP	50	64
BG	25	67
PA/PE	40	80
OPP	40	83

Table 4: Literature data relevant to the efficacy of UV radiation in inactivating microorganisms on the surface of different foods.

Food		Dose (kJ m^{-2})	Microrganism	Log_{10} reduction	Reference
Meat	Chicken breast fillets	1.5	Total aerobic count	2	Stermer et al., 1987

	Chicken breast fillets	0.4-0.9	*L. monocytogenes** *Salmonella typhimurium** *E.coli* O157:H7*	1	Kim et al. 2002
	Chicken breast fillets	3	*L. monocytogenes**	2.0	Lyon et al., 2007
	Chicken breast fillets	10-40	*L. monocytogenes** *Salmonella spp** *Staphilococcus aureus**	0.3-0.4	Sommers et al., 2010
	Ready-to-eat sliced ham	8	*L.monocytogenes** S.Typhimurium* *C.jejuni**	2.6 2.0 1.8	Chun et al., 2009
	Frankfurters	10-40	*L. monocytogenes** *Salmonella spp** *Staphilococcus aureus**	1.5-2.0	Sommers et al., 2009, 2010
	Pork chops	10-40	*L. monocytogenes** *Salmonella spp** *Staphilococcus aureus**	0.4-0.6	Sommers et al., 2010
Fish	Mackarel	50-70	Total microbial count	2-3	Huang and Toledo, 1982
Shell egg		0.4-2.6	*Salmonella Typhimurium**	2.9-4.6	Kuo et al., 1997
		2.2	Total aerobic count	0.8-2	Chavez et al., 2002
		7.5	*Salmonella enterica**	2.5	Rodriguez-Romo and Yousef, 2005
		0.5, 1.9	Total aerobic bacteria	0.9	De Reu et al., 2006
		10-40	*L. monocytogenes** *Salmonella spp** *Staphilococcus aureus**	0.3-1	Sommers et al., 2010
Fruit	Strawberry	4.1	*Botrytis cinerea*	#	Pan et al., 2004
	Apple	ǂ	*Salmonella* spp.* *E.coli* O157:H7*	2.2-3.3	Yaun et al., 2004
	Mango	2.4, 4.9		#	González-Aguilar et

					al., 2007
	Tomato	ǂ	*Salmonella* spp.*	1.8-2.2	Yaun et al., 2004
	Tomato	10-40	*L. monocytogenes** *Salmonella* spp* *Staphilococcus aureus**	3-4	Sommers et al., 2010
	Jalapeno pepper	10-40	*L. monocytogenes** *Salmonella* spp* *Staphilococcus aureus**	3-4	Sommers et al., 2010
Fresh-cut fruit and vegetables	Fresh-cut apple	14.1	*E.coli ** *L.innocua** *S.cereviasiae**	1-1.9	Gómez, Alzamora et al., 2010
	Fresh-cut apple	1.2	Total viable count Enterobacteriaceae	1-2	Manzocco et al., 2011b
	Fresh-cut melon	11.8	Total aerobic count Lactic acd bacteria *Pseudomonas spp*	1-2	Lamikanra et al., 2005
	Fresh-cut melon	1.2	Total viable count Enterobacteriaceae	2.2 2.6	Manzocco et al., 2011a
	Fresh-cut watermelon	4.1	Total viable count	1	Fonseca and Rushing, 2006
	Fresh-cut watermelon	4.8	Enterobacteria	0.7	Artés-Hernández et al., 2010
	Pomegranate arils	9.1	Lactic acid bacteria	2	Lopez-Rubira et al., 2000
	Baby-spinach	24	Psychrotrophic microrganisms Enterbacteria Moulds	1-1.2 1-1.3 0.6-0.7 0.5-0.9	Escalona et al., 2010
	Spinach leaves	4-11	Mesophilic bacteria Psychrophilic bacteria Enterobacteria	1	Artés-Hernández et al., 2009
	Lettuce	ǂ	*Salmonella* spp.* *E.coli* O157:H7*	1.9-2.8	Yaun vet al., 2004
	Lettuce	2.37	Lactic acid bacteria	1.7	Allende et al., 2006
	Lettuce	ǂ	Total aerobic	0.9-1.4	Kim et al., 2009

	bacteria
	Coliform bacteria
	Psychrotrophic bacteria
	Yeasts and moulds

*inoculated

ǂ Irradiance: 15-240 W m^{-2}. Dose is not reported.

Reduced fungal infection, Log_{10} reduction is not reported

Table 5: Microbial and food factors limiting the germicidal effect of ultraviolet radiation on food surface.

Microrganism driven	Microorganism internalisation
	Microrganism shadowing each other
	Biofilm formation
Food driven	Food pieces shadowing each other
	Macroscopic surface irregularities
	Microscopic surface roughness
	Composition
	Colour

Table 6: Polyphenoloxidase and pectate lyase activity in 1.5 cm thick slices exposed to 4 kJ m^{-2} UV irradiation.

Portion analysed	Enzymatic activity (%)	
	Polyphenoloxidase	Pectate lyase

Control	Whole	100	100
UV-irradiated	Whole	100	100
	Slice surface (0.1 mm)	45	38

