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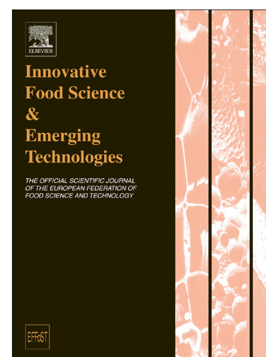

Principles and mechanisms of ultra violet light emitting diode technology for food industry applications

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PRINCIPLES AND MECHANISMS OF ULTRA VIOLET LIGHT EMITTING DIODE TECHNOLOGY FOR FOOD INDUSTRY APPLICATIONS

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Abstract

The application of ultraviolet (UV) light to water, food contact surfaces, medical equipment and liquid foods for microbial inactivation is widely employed. To date, UV disinfection sources employed are primarily low-pressure and medium-pressure mercury lamps, emitting monochromatic and polychromatic light respectively. Despite the widespread use of mercury lamps, there are multiple drawbacks associated with their use including; high energy consumption, large size which limits reactor design, high heat emission and the presence of mercury. Light emitting diodes (LEDs) have potential for use as highly efficient UV decontamination sources. Recent advances in semiconductor development have resulted in UV-LEDs becoming more widely available. UV-LEDs emit monochromatic light, which enables customised UV-LED disinfection systems at specific wavelengths to be developed. The application of UV-LEDs for disinfection purposes has been studied in recent years, particularly with respect to water disinfections systems. In this review, studies relating to UV-LED food applications are discussed including chemical changes induced in foods, as a result of UV treatment, together with advantages and limitations of the technology.

1. Introduction

Various novel technologies including high pressure processing or high hydrostatic processing, pulsed electric field, cold plasma processing and ultrasound have been investigated as alternatives to thermal pasteurisation for a range of food applications (Chizoba Ekezie, Sun, & Cheng, 2017; Daher, Le Gourrierec, & Pérez-Lamela, 2017; Soni, Oey, Silcock, & Bremer, 2016; Wang, et al., 2018). These technologies have been shown to have potential to achieve desired food safety objectives while extending the shelf life of foods, with minimal impacts on food quality attributes. However, there are restrictions to the use of some of these technologies in relation to solid foods applications. More specifically, in relation to high pressure processing, there is a requirement for > 40% free water within the food matrix for adequate microbial inactivation to be achieved, which excludes a large range of food products (Muntean, et al., 2016).

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Among the array of novel technologies investigated to date, light based technologies have been shown to have strong potential for selected food and non-food applications (D'Souza, Yuk, Khoo, & Zhou, 2015; Song, Mohseni, & Taghipour, 2016). Among these, ultraviolet light (UV) technology has been investigated extensively and has been used at commercial level for niche applications. The application of UV light is well established for disinfection of food preparation surfaces and for air/water treatment (Koutchma, Forney, & Moraru, 2009). UV is electromagnetic radiation within the 10 to 400 nm wavelength spectrum, in between X-ray and visible wavelengths. It is often referred to as non-ionising radiation; however, the shortest wavelengths emit some ionisation (Sandle, 2013) The UV spectrum may be sub-divided based on various wavelengths and applications as shown in Figure 1. Three wavelength sub-divisions are widely used in the scientific literature namely UVA (315 – 400 nm), UVB (280 – 315 nm) and UVC (<280 nm) (Soni, et al., 2016). Wavelengths below 200 nm are classified as vacuum ultraviolet. Each UV wavelength range has specific effects on biological materials. UVC, which is known as the germicidal wavelength, can damage the DNA of pathogenic and spoilage microorganisms primarily because DNA absorbs is a maximum in this range (J.R. Bolton & Cotton, 2008). UVA radiation mainly inactivates microorganisms by causing oxidative disturbance to the other biomolecules including proteins and lipids by the production of reactive oxygen species (Brem, Guven, & Karran, 2017).

In recent years the potential of UV for food safety purposes and pathogen inactivation has been actively explored (Gómez-López, Koutchma, & Linden, 2012). UV radiation can eliminate the need for dangerous chemicals in surface disinfection applications in the food industry, while reducing the exposure of consumers to foodborne illnesses. However, many of the lighting technologies used today, including high-pressure mercury and xenon lamps, have limited control of UV or infrared (IR) light (D'Souza, et al., 2015). Moreover, while mercury lamps are widely used commercially, their use in the food industry poses some risk to consumers due to the potential exposure of food products to toxic mercury (Hamamoto, et al., 2007). In addition to this, the accumulation of mercury waste is undesirable as it can have damaging effects on environment or human health if not disposed of correctly. Light Emitting Diodes (LEDs) are an alternative source of ultraviolet light suitable for food industry applications. The cost of LEDs, which have a compact and robust design, has recently decreased substantially due to technological advances. Output power is increasingly efficient. Additionally the average life span of LEDs in comparison to UV lamps used in industry today is much longer (D'Souza, et al., 2015; Hamamoto, et al., 2007). Moreover, heat emissions from LEDs are far lower than from UV lamps, thus they are more suitable for food treatment applications. Also since LEDs do not need a warm-up time and thus consume less energy (Akgün & Ünlütürk, 2017).

The objective of this review is to discuss the principles, applications and limitations of UV-LEDs for food applications including surface decontamination and for both liquid and solid food safety. The potential chemical changes induced will also be discussed. To the best of the authors' knowledge, this is the first review to discuss application of UV-LEDs for food applications.

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2. Principles of UV-LED

2.1. Fabrications

LEDs are two-terminal semiconductor devices, which emit light when a specified voltage is applied across the two terminals. Different semiconductor materials emit different colours (wavelengths) of light; the emission wavelengths are dependent on a special property of the semiconductor called "band gap" (Table 1). Compound semiconductor materials, also known as III-V materials (because they are made up of group III and group V elements from the periodic table) are generally direct band gap materials and therefore efficient in the conversion of electrical current into light (This phenomenon is also known as electroluminescence). These III-V materials form the backbone of modern LED technology. Positively charged carriers (holes) in the *p*-type layer (layer which mainly contains "holes") are driven towards the active (junction) layer where they recombine with electrons which are driven by the same voltage from the *n*-type layer (layer which mainly contains "electrons") in the opposite direction towards the junction (Figure 2). When the electron and hole meet they recombine under emission of a photon which carries away the energy that is released upon the electron and hole recombination. The higher the band gap of the material, the higher the energy of the emitted photon, and the shorter the wavelength of the emitted light. In practical devices there is usually a very thin layer of a material with a lower band gap at the semiconductor junction. This thin quantum well speeds up the electron and hole recombination process and makes it more efficient. The band gap of the quantum well then sets the emission wavelength. By varying the III-V composition of the quantum well the emission wavelength can be tuned.

Aluminium nitride / aluminium gallium nitride (AlN/AlGaN) based materials are the materials of choice for UV emitting devices (210-400 nm). To reach the shorter wavelengths, the level of aluminium incorporation into the AlGaN material must be increased and this generally results in lower efficiency, higher forward voltage values and poorer lifetime expectancy. LEDs are characterised by their electrical and optical properties. The forward and reverse current-voltage characteristics are the key electrical characteristics, while emission wavelength, total optical power as a function of bias current (L-I characteristics), and emission profile (as a function of emission angle) are the basic optical characteristics.

2.2 Factors affecting UV food processing

Some intrinsic parameters (e.g. absorption coefficient of food matrix) and extrinsic parameters (e.g. reactor design) that affect UV food processing applications are discussed below.

2.2.1 Intrinsic parameters

Different food matrices will absorb light differently; therefore they will have different absorption coefficients. UV light will only penetrate up to several millimetres depending on the optical properties of the food (Choudhary & Bandla, 2012) Guerrero-Beltran and Barbosa-Canovas (2006) have stated that the colour or turbidity of a liquid influences the optical absorption coefficient of the liquid. The penetration capacity of the UV light reduces as the absorption coefficient increases. As the penetration capacity decreases, the inactivation rate (K_{max}) will decrease as the treatment has not reached the full depth of the medium/food. To maximise UV processing efficiency foods should be exposed in thin layers.

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2.2.2. Extrinsic parameters

Significant factors to consider in relation to reactor configuration and design are the source of UV light, type of light exposure (continuous/ pulsed), the distance of sample from light source, reactor type (continuous/ flow through reactor) and environmental conditions within the reactor (temperature/light or dark).

2.3. Dosimetry and characterisation of UV-LED

Many studies investigating the application of UV light have used the terms UV 'dose' or 'fluence'. The term 'dose' should be avoided as a synonym of fluence because dose refers, in other contexts, to absorbed energy, but only a small fraction of all incident UV light is absorbed by microorganisms (Bolton James and Linden Karl 2003). Other commonly used terms are; irradiance (mW/cm^2) and fluence rate (mW/cm^2). Irradiance refers to the radiant power of all wavelengths incident from all upward directions on a small element of surface containing the point under consideration divided by the area of the element and fluence rate refers to total radiant power incident from all directions onto a small sphere divided by the cross sectional area of that sphere (J. R. Bolton, Mayor-Smith, & Linden, 2015). J. R. Bolton, et al. (2015) outlined the recommended nomenclature from the Photochemistry Commission of the International Union of Pure and Applied Chemistry in a recent review. It was therein proposed to replace the conventional power and energy-based concepts with concepts based on photons. Photon fluence and photon fluence rate are recommended for measuring energy applied to samples. It is crucial that standardised dosimetry methods and nomenclature are adopted for photobiological reactions and food safety applications of UV light to facilitate process scale up and industrial applications.

A wide variety of methodologies are reported for determining UV light applied include bioassays, mathematical models and chemical actinometry, and quasi-collimated beam apparatus (Bolton, Mayor-Smith et al. 2015). Furthermore, as LEDs are available at numerous wavelengths, fluence modelling across various wavelengths is required. Fluence response has been described as the measurement of microbial inactivation as a function of UV fluence (Beck, Wright, Hargy, Larason, & Linden, 2015). Other significant areas requiring further investigation are UV resistance and extrinsic (processing conditions, food characteristics) parameters across numerous microorganisms for efficient microbial control (Gayán, Condón, & Álvarez, 2014).

2.4. Microbial inactivation mechanisms

UV light microbial inactivation occurs through various mechanisms that are dependent on the wavelengths applied in treatment, and can be achieved directly by absorption of the incident light on microbial cell DNA initiating lesion formation (depending on wavelength exposure), or indirectly due to generation of reactive oxygen species by the interaction of radiation with cellular chromophores acting as photosensitisers (Brem, et al., 2017; Gayán, et al., 2014). The key mechanism by which UV radiation inactivates microorganisms is largely by formation of lesions which interfere with DNA replication. The UVC wavelength spectrum, in particular the 260-265 nm range is the peak absorption range for DNA in cells, and thus it is the most lethal range for microorganisms (Gayán, et al., 2014). The absorption of UVC radiation induces the formation of DNA photoproducts, namely cyclobutane pyrimidine dimers (CPDs) and pyrimidine 6-4 pyrimidone (6-4PP) photoproducts, which inhibit transcription and replication. The UVA range is not as efficiently absorbed by native

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DNA and therefore does not induce severe damage by dimer formation. However this range may still produce secondary photoreactions of existing DNA photoproducts or damage DNA via indirect photosensitisation reactions (Hamamoto, et al., 2007; Sinha & Hader, 2002).The indirect germicidal properties of UV radiation are related to membrane damage, growth delay or DNA damage by the production or increase in reactive oxygen species such as superoxide anion radicals ($O_2^{\cdot-}$), hydroxyl radicals ($OH^{\cdot}$), hydrogen peroxide (H_2O_2) and singlet oxygen ($^1O_2$) (Hamamoto, et al., 2007).

The three main pathways in which microorganisms' DNA is repaired are; reverse damage repair, excision repair (pre-cell replication) and tolerating damage pathways (activated once replication has started or immediately afterward) (Gayán, et al., 2014).

3. Applications of LED-UV systems

The application of UV light is well established for disinfection of food preparation surfaces, and for air and water treatment (Koutchma, et al., 2009). However, due to undesirable characteristics of current commercial ultraviolet radiation sources (low- and medium-pressure mercury lamps) such as overheating, high energy costs, the risk of mercury contamination to food or food surfaces and limited control over wavelength emission, alternative sources including LEDs have been investigated. UV-LEDs have shown promise for food safety applications using various wavelengths, including water disinfection, surface decontamination and liquid/solid food applications. The following section will review recent food applications reported in the literature.

3.1. Water disinfection

Drinking water safety is a significant issue worldwide, and particularly in developing counties and rural areas (Song, et al., 2016). To facilitate the growing global population and associated increase in pollution, it is imperative that safe, cost-effective, energy efficient and durable UV water disinfection systems are further developed to tackle harmful microorganisms found in drinking water. Current UV water disinfection systems consist of low and medium pressure mercury lamps emitting monochromatic and polychromatic light, respectively. Many studies have demonstrated the disinfection capabilities of UV-LEDs in water samples. However comparison of results between studies remains difficult due to lack of uniformity in research methodology (Song, et al., 2016).

Rattanakul and Oguma (2018) investigated multiple LEDs (265nm, 280 nm, 300nm) as potential water decontamination devices using various microbial species (*Pseudomonas aeruginosa*, *Legionella pneumophila*, *Escherichia coli*, *Bacillus subtilis* spores, bacteriophage Qβ) to determine the most efficient treatment wavelength taking both inactivation efficiency and energy consumption into account. It was determined that the 280 nm LED was most efficient as both high inactivation rate (3-log_{10}) and low energy consumption (<1.04 kWh/m^3) were observed, thus highlighting the potential use of 280 nm LEDs in water treatment. Oguma, Kita, Sakai, Murakami, and Takizawa (2013) reported higher time-based efficiency for 280 nm LEDs. Both 265 nm and 280 nm LEDs achieved 4-log_{10} CFU/ml reduction of *E. coli* with reported fluences (RF) of 10.8 and 13.8 mJ/cm^2, respectively in a batch reactor. A lower inactivation of 0.6-log_{10} CFU/ml was reported using LEDs at 310 nm at RF of

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56.9mJ/cm^2 – indicating that this wavelength alone may not be sufficient for *E.coli* inactivation purposes. Lui, Roser, Corkish, Ashbolt, and Stuetz (2016) also investigated LEDs at 310 nm and no significant disinfection was found. A study carried out by Chatterley and Linden (2010) evaluated the potential of UV-LEDs (265 nm) for water disinfection in comparison with low-pressure mercury lamps. Based on results in this study, it was concluded that the low pressure mercury lamps and LEDs were not significantly different for the inactivation of *E.coli* indicating the potential replacement of mercury lamps with energy efficient LEDs, which is consistent with later studies (Beck, et al., 2017; Sholtes, Lowe, Walters, Sobsey, Linden, & Casanova, 2016).The inactivation of three microorganisms (MS2 coliphage, T7 bacteriophage and *E. coli*) using UV-LEDs (255 nm, 275 nm) and low-pressure mercury lamps was reported by Bowker, Sain, Shatalov, and Ducoste (2011). It was found that low-pressure mercury lamps achieved higher *E. coli* and MS-2 inactivation than 255 nm and 275 nm LEDs, and a similar T7 bacteriophage inactivation to 275 nm LEDs. It was also concluded that the 275 nm LEDs produced more efficient T7 bacteriophage and *E. coli* inactivation than 255 nm LEDs, while both 255 nm and 275 nm LEDs produced comparable microbial inactivation for MS-2. G.-Q. Li, Wang, Huo, Lu, and Hu (2017) compared UV-LEDs (265 and 280 nm) against low pressure mercury lamps. They reported that 265 nm LEDs were more effective for *E. coli* inactivation than 280 and LP lamps. In addition to comparative inactivation studies, the reactivation levels were also compared. Photoreactivation and dark repair both occurred after LED treatments. However, the levels of photoreactivation and dark repair after 280 nm LEDs treatment were significantly lower than those observed for 265 nm and LP mercury treatments.

Aoyagi, et al. (2011) investigated the efficiency of UVC LEDs (255 nm and 280 nm) for inactivation of bacterial viruses (φX174, Qβ, MS2 *E. coli* bacteriophages). While it was observed that 255 nm LEDs exhibited higher inactivation efficiency, the 280 nm LEDs were recommended, as the manufacture of high-power 280 nm LEDs is less complex than 255 nm LEDs. Würtele, et al. (2011) was one of the few studies to investigate the inactivation capabilities of UV-LEDs at 269 nm. They concluded that the 269 nm LEDs had a greater germicidal efficiency but that the spore inactivation caused by the 282 nm LEDs was significantly higher than that caused by the 269 nm LEDs (for the same time and power output), due to the higher photon output at the same current (20 mA). Lui, et al. (2016) reported that UVC LEDs at 270 nm inactivated *E. coli* and *E. faecalis*, achieving a 5-$\log_{10}$ CFU/ml reduction in 3 min and 18 min respectively. UVA LEDs (365 nm, 385 nm, 405 nm) were also investigated, however a far longer duration (3 hr) was required to achieve 5-$\log_{10}$ reductions at these wavelengths. UVA LEDs (365 nm) were also investigated in a study by Hamamoto, et al. (2007). After 75 min UVA treatment, 5- $\log_{10}$ CFU/ml reductions were reported for most microorganisms tested (*Vibrio parahaemolyticus, enteropathogenic Escherichia coli, Staphylococcus aureus and Escherichia coli* DH5α).

(Chevremont, Farnet, Coulomb, & Boudenne, 2012) investigated the effect of both independent and coupled UVA and UVC LEDs on microbial pollution in wastewater. They reported that all coupled wavelengths tested were significantly more effective than those used separately for faecal *enterococci*, total coliforms and faecal coliforms. Moreover, the most effective combinations of different LED wavelengths were combinations 280nm/365nm and 280nm/405nm – both UV-A/UVC combinations – which is consistent with a previous study

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investigating similar combinations of coupled wavelengths for inactivation of strains of *E. coli* and E. faecalis (Chevremont, Farnet, Sergent, Coulomb, & Boudenne, 2012). Coupled wavelength applications may exhibit some synergistic effects due to the complementing properties of each UV range. A study carried out by Beck, et al. (2017) also investigated the potential of dual-wavelength synergy, using 260 nm and 280 nm (both UVC range) LEDs and water samples inoculated with *E. coli*, MS2 coliphage, Adenovirus 2 and *Bacillus pumilus* spores. While inactivation was observed at both LED wavelengths, no synergistic effects were observed from combining 260 nm and 280 nm LED compared to the sum of individual wavelengths acting independently.

3.2. Surface decontamination

Major foodborne pathogens that cause illness and are of major concern include *Bacillus cereus*, *Escherichia coli*, *Salmonella typhimurium*, *Listeria monocytogenes and Campylobacter jejuni*. When hygiene practices and control points in the food manufacturing industry fail, consumers may become exposed to these harmful bacteria. Cross contamination of food products *via* food contact surfaces can also pose food safety challenges. Recently UV-LEDs have been shown to be effective against various pathogenic micro-organisms pertinent to food (D'Souza, et al., 2015). This section reviews the reported studies on the inactivation of food borne pathogens using LED-UV irradiation and highlights the potential of LED-UV systems for surface decontamination.

Shin, Kim, Kim, and Kang (2016) applied UVC radiation (275 nm) to known foodborne pathogens (*E. coli* O157:H7,*Salmonella enterica* serovar *Typhimurium*, and *L. monocytogenes)* on solid culture media. Inactivation of 6, 6 and 5-$\log_{10}$ CFU/ml reductions for *E. coli*, *S. typhimurium* and *L. monocytogenes*, respectively, were observed after 5 min exposure to UVC light with RF of 1.67 $mJcm^2$. In this study, *L. monocytogenes* was the most UV resistant bacteria, and this is consistent with other studies which reported significant resistance of *L. monocytogenes* to UV light (Gabriel & Nakano, 2009; Lu, Li, & Liu, 2011; Schenk, Raffellini, Guerrero, Blanco, & Alzamora, 2011). It was postulated that the reason for the added resistance in *L. monocytogenes* was due to the thick peptidoglycan wall that surrounds the cytoplasmic membrane in Gram positive bacteria. Bak, Ladefoged, Tvede, Begovic, and Gregersen (2010) investigated the efficacy of UVC (265 nm) LEDs in disinfecting *Pseudomonas aeruginosa* biofilm contaminated tube lumens. An inactivation of 4-$\log_{10}$ CFU was observed in this study which demonstrates the strong potential of this technology for surface decontamination. Contrary to these findings, another study observed no significant inactivation of *P. aeruginosa* biofilm (immature and mature) with UVC (266 nm) treatment. However, it was found that UVB (296 nm) LEDs achieved a 3-$\log_{10}$ reduction in mature biofilms (Argyraki, Markvart, Bjorndal, Bjarnsholt, & Petersen, 2017). J. Li, Hirota, Yumoto, Matsuo, Miyake, and Ichikawa (2010a) investigated the application of pulsed UVA-LED radiation at 365 nm with a reported intensity of 0.28mW/cm^2 on biofilms that were formed *in-vitro*. It was reported in this study that after 5 min of radiation over 90% of the viable microorganisms in biofilms were inactivated.

3.3. Solid food applications

A limited number of studies have investigated the potential of LED-UV systems for inactivation of food pathogens on solid foods. Foods studied investigated include meat products (Haughton, Grau, Lyng, Cronin, Fanning, & Whyte, 2012), dairy products (Kim, Kim, & Kang, 2015) and fruits and vegetables (Aihara, et al., 2014).

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Haughton, et al. (2012) investigated the exposure of skinless chicken fillets inoculated with *Campylobacter jejuni* to near UV LEDs (395±5 nm), $\log_{10}$ reductions of 2.21 and 2.62 were reported for exposure times of 1 and 5 min respectively. Potential applications of this technology within the meat industry including decontamination of carcasses during air chilling were highlighted as *C. jejuni* is prevalent in meat products. A study carried out by Kim, et al. (2015) inoculated cheese slices with well-known food pathogens (*E. coli*, *Salmonella*, *Listeria*). Reductions of 2.2-4.8-$\log_{10}$ after treatment with multiple LEDs (266 nm – 279 nm) at RF of 3 mJ/cm^2 for all three pathogens were reported, with negligible generation of injured cells and no effect on cheese quality. Another study, carried out by Aihara, et al. (2014), reported a 3-$\log_{10}$ reduction in *E. coli* DH5α after 90 min treatment with UVA (365 nm), minimal changes in tissue weight and vitamin C or nitrite/nitrate content were observed. While these results are promising further studies investigating inactivation capabilities of LEDs on solid foods should be carried out to determine suitable treatments and investigate possible effects of UV-LEDs on food quality.

3.4. Liquid food applications

The application of shortwave UV light for liquid food non thermal pasteurisation has been widely studied. (Baysal, 2018). However, the applications of UV-LED for safety purposes are limited. Akgün, et al. (2017) investigated the application of multiple LEDs emitting at selected wavelengths (254, 280, 365, 405 nm) and various combinations of these selected wavelengths in both clear and cloudy apple juices. Results highlighted the efficiency of inactivation at wavelengths emitting at 280 nm and 280/265 nm for *E. coli* with inactivation of 2.0± 0.1 and 2.0±0-4$\log_{10}$ CFU/ml, respectively, in cloudy apple juice. A higher inactivation of 4-$\log_{10}$ was achieved in clear apple juice treated with LEDs emitting at 280 nm. The higher $\log_{10}$ inactivation in clear apple juice was attributed to a higher absorption of UV, as the cloudy apple juice contained colour compounds and suspended solids which may have interfered with light absorption.

4. Chemical changes induced due to UV light

While UV treatment can achieve desired microbial inactivation rates, it can also like any other food processing technology induce chemical changes in foods depending on treatment parameters such as wavelength, exposure time and food matrix. The primary concern with photochemical reactions in food is oxidation of key nutrients (Figure 3). Oxidation can occur due to direct reaction of UV radiation with target nutrients including proteins, lipids and micro nutrients. In the majority of the cases, oxidation of proteins and lipids is caused due to the formation of singlet oxygen ($^1O_2$) during photochemical reactions, initiating a chain reaction (Davies, 2003; Ghnimi, Budilarto, & Kamal-Eldin, 2017). Due to lack of studies carried out utilising LEDs as UV sources, the following section will discuss the effects relating to UV light from a range of sources.

In relation to food quality, lipid oxidation is the most significant oxidation reaction, resulting in many undesirable characteristics, including modification of organoleptic properties and rancidity. In addition to this, in food products lipid oxidation can cause protein oxidation due to close interactions between lipids and proteins (Viljanen, Kylli, Hubbermann, Schwarz, & Heinonen, 2005). Lipid oxidation reaction can be explained by the free radical chain

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mechanism (Ghnimi, et al., 2017). To date limited studies have investigated the oxidative process of lipids in food products treated with UV light (Table 3). The thiobarbituric acid reactive substances (TBARS) method which measures the products of oxidative damage can be employed to determine oxidation in lipids. Additionally, monitoring oxidative stability over time using the oxidative stability index (OSI) can be a useful monitoring tool. H. Chun, Kim, D. Lee, J. Yu, and Song (2010) reported that TBARs values increased slowly during storage, with no significant changes between reported fluences RFs (0.5, 1, 3, and 5 kJ/m^2) of UVC radiation (unfiltered germicidal emitting lamps). Another study by Lazaro, et al. (2014) observed that intensities (0.62 mW/cm^2, 1.13 mW/cm^2 and 1.95mW/cm^2), of UVC (lamp) radiation did not affect the TBARs values, it was also reported that the exposure periods investigated were perhaps not long enough to promote oxidation. Elmnasser, et al. (2008) investigated the effect of pulsed light (xenon lamp) on lipid oxidation and possible interaction between lipids and proteins in whole milk, by means of frontal fluorescence (before and after pulsed UV treatment). No difference between treated and untreated samples was observed. The oxidative effects of pulsed UV (PUV) light in ham slices were investigated by Wambura and Verghese (2011) over a 14 day period by measuring OSI. They observed that all samples decreased in oxidative stability over time, but oxidative stability decreased more rapidly in those samples treated with PUV light. Their study highlights the need for more long term oxidative stability studies on UV treated foods to further investigate this area.

Proteins are major cellular targets for photo-oxidation because of their high abundance, in addition to the presence of endogenous chromophores with their structures (Davies, 2003). During photo-oxidation, the primary structure of protein is affected by the formation of carbonyls and loss of aromatic amino acids (Scheidegger, Pecora, Radici, & Kivatinitz, 2010). The aromatic amino acids (tryptophan, tyrosine, and phenylalanine) all absorb ultraviolet light and have a reported absorption maximum at wavelengths between 280 and 290 nm (Wetlaufer, 1963). Scheidegger, et al. (2010) observed the oxidation of milk proteins in whole and skimmed milk after various UV (mercury lamp) treatment durations (0-24 hr). Protein oxidation was evaluated by the formation of protein carbonyls and dityrosine. In their study, after 1 hr, of exposure time at 254 nm, the appearance of carbonyl moieties was detected. It was also observed that protein carbonyls increased as a function of irradiation time in both whole milk and skimmed milk.

The effect of UV light on micromolecules has also been reported. A study by Bhat (2016) investigated the impact of ultraviolet radiation on tomato juice quality. Significant increases in phenolic content were observed post treatment (60 min). In a study by Pan and Zu (2012), the vitamin C content in pineapple juice decreased significantly after UV treatment with monochromatic UV light . While vitamin C content decreased in all samples including the control during storage, it was observed that UV treated samples declined more rapidly during storage. Akgün, et al. (2017) investigated the effect of UV light on vitamin C and reported that vitamin C content (2.22 mg/L) in apple juice samples treated with UV-LEDs degraded due to oxidation after long exposure times.

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5. Challenges and limitations

The main challenges to adoption of UV-LEDs in the food industry are the low output power of LEDs, low external quantum efficiency and high cost. The external quantum efficiency (EQE) of AlGa-N-based UV LEDs rapidly decreases with wavelength. Current industry research and development programmes are addressing these issues including low efficiencies which are related to the intrinsic material properties of high-Al content AlGaN such as severely strained epitaxial layers, low n-type and p-type doping efficiencies, and strong TM-polarized light emission (Park, Kim, Cho, & Seong, 2017).

Moreover, the application of UV light to solid food remains problematic as UV light will only penetrate up to several millimetres depending on the optical properties of the food (Choudhary & Bandla, 2012)

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6. Conclusion

Due to the multiple drawbacks of current commercial UV disinfections lamps, UV LED sources are being increasingly investigated. UV LEDs offer multiple practical benefits such as longer life span, compact and robust design, no warm-up time and lower heat emissions in comparison to current commercial UV lamp sources. More significantly, UV LEDs are commercially available at multiple wavelengths and therefore offer the capability to design customised UV reactors. While there is further research required in this area, the studies highlighted in this current review demonstrate the germicidal capabilities of ultraviolet light emitting diodes at various wavelengths in a wide range of applications, thus showing the strong potential of this non-thermal technology in the food industry.

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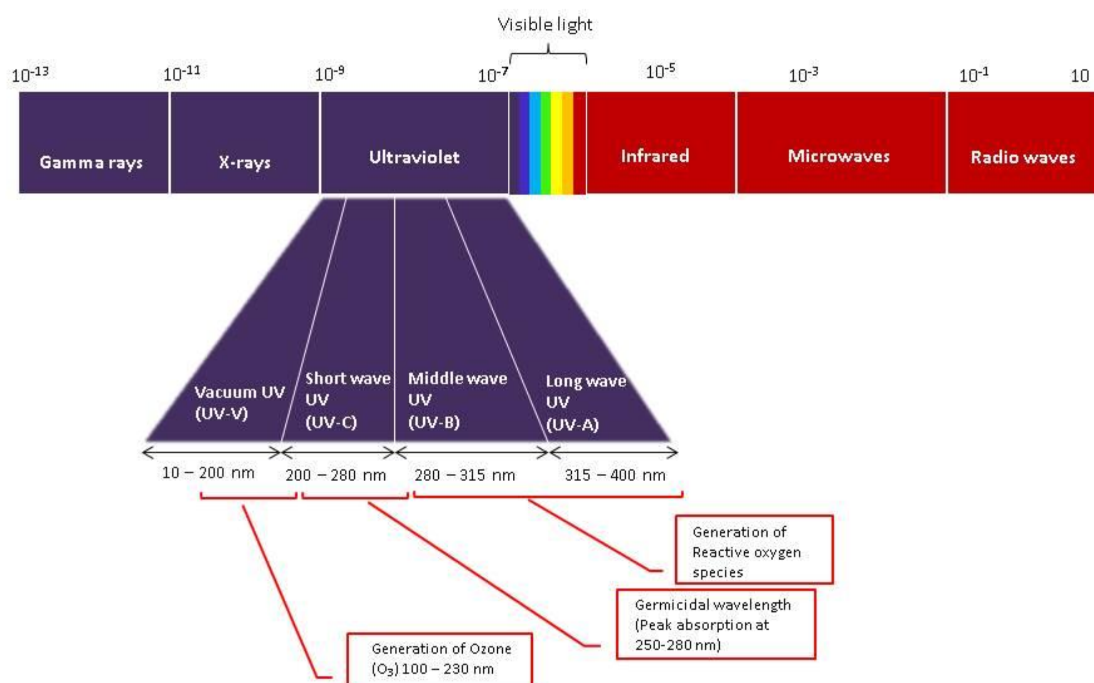

Figure 1: Ultraviolet sub divisions and reported applications.

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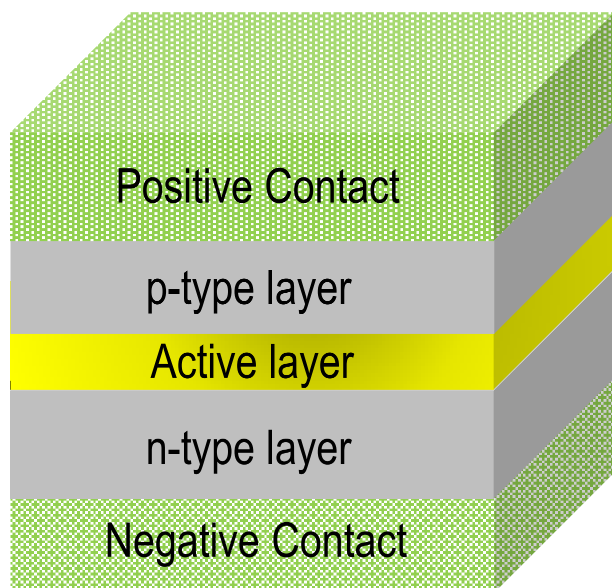

Figure 2: Structure of a standard LED wafer (not to scale).

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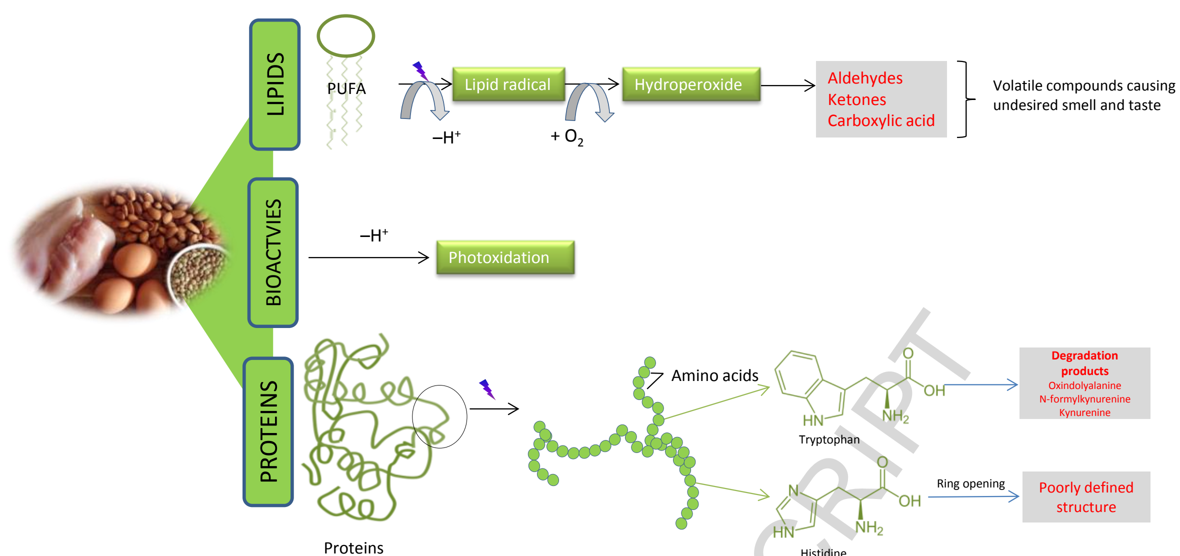

Figure 3: Chemical changes in food caused by UV treatment

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Table 1: Semiconductor materials commonly used across the spectral ranges

Colour/ Wavelength (nm)	Semiconductor (*p, n*)	Quantum well material (active layer)
UV (210-400)	AlGaN	AlGaN
Blue/Green (400-570)	GaN	InGaN
Yellow (570-610)	GaAsP	InGaAlP
Red (610-760)	GaInP AlGaAs	InGaAlP
IR (>760)	GaAs	AlGaAs

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Table 2. Overview of LED-UV studies for food applications

Food	Microorganism	Wavelength (nm)	Reported fluence (mJ/cm^2)	Significant findings	Reference
Water	*E. coli* IFO 3301 *B. subtilis* spores ATCC 6633, Bacteriophage Qβ ATCC 15597 *Legionella pneumophila* *Pseudomonas aeruginosa*.	254 265 280 300	-	280-nm UV-LED proved most effective.	(Rattanakul, et al., 2018)
	E. coli K12 IFO 3301	265 280 310	10.8-56.9	265 and 280 nm achieved 4-log_{10} CFU/ml reduction in a batch reactor.	(Oguma, et al., 2013)
	E. coli K12 ATCC 29425	254 265	-	Inactivation of *E. coli* achieved	(Chatterley, et al., 2010)
	φX174 Qβ MS2 coliphage,	255 280	-	280 nm LEDs recommended for water disinfection rather than 255 nm LEDs.	(Aoyagi, et al., 2011)
	B. subtilis ATCC 6633 spores	269 282	175 – 345 *(J/m^2)*	Spore inactivation caused by the 282 nm LEDs is significantly better than for the 269 nm LEDs during the same time span and input power.	(Würtele, et al., 2011)
	E. coli K12 ATCC W3110 *E. faecalis* ATCC 19433	270 310 365 385 405	-	*E. coli* and *E. faecalis* inactivated by 5 log_{10} CFU/ml after 3 and 18 min respectively at 270 nm.	(Lui, et al., 2016)
	Vibrio parahaemolyticus Enteropathogenic *Escherichia coli* *Staphylococcus aureus* *Escherichia coli* DH5α *Salmonella enteritidis*	365	*315 – 672 (J/cm^2)*	All bacteria tested except for *S. enteritidis* were reduced by greater than 5-log_{10} CFU/ml within 75 min at 315 J cm^{-2} of UVA.	(Hamamoto, et al., 2007)

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MS2 coliphage(ATCC 15597-B1) T7 bacteriophage *E. coli* (11229)	255 275		275 nm LEDs produced more efficient microbial inactivation than 255 nm LEDs for T7 and *E. coli*.	(Bowker, et al., 2011)
E. coli CGMCC 1.3373	254 265 280 265/280 (50%) 265/280 (75%)	10.91 – 15.35	265 nm LEDs were more effective than 280 and LP lamp for inactivating *E. coli*. No synergistic effects observed for combined wavelengths.	(G.-Q. Li, et al., 2017)
E. coli K12 ATCC 29425 MS2 coliphage Human adenovirus type 2 ATCC VR-846 *Bacillus pumilus* ATCC 27142 spores	260 280 260/280	12-122.89	***E. coli***: All sources attained 3-log_{10} CFU/ml reduction **MS2:** 260 nm and 280 nm achieved 2 log_{10} CFU/ml inactivation at doses 30.3 and 38.5 mJ/m^2 **Adenovirus:** 4 log_{10} CFU/ml reduction achieved by 280 nm at lower fluence than 260 nm. ***B. pumilus* spores:** 260 nm and the 260/280 nm treatments more effective than the 280 nm for 2 log_{10} CFU/ml reduction.	(Beck, et al., 2017)
Mesophilic bacteria Faecal enterococci Total coliforms Faecal coliforms.	280 365 405 254/ 365 254/405 280/365 280/405	0.73-25.58	Log reductions of 2.3, 2.3, 3.7 and 3.2 were achieved after 30 min treatment at 280/365 nm in Mesophilic bacteria, Faecal enterococci, Total coliforms and Faecal coliforms, respectively.	(Chevremont, Farnet, Coulomb, et al., 2012)
E. coli O157:H7 (ATCC 35150, ATCC 43889, ATCC 43890) *S. Typhimurium* (ATCC 19585, ATCC 43971, DT 104)	278	0.2 – 3	Log reductions of 2.42-4.85, 0.76-4.24 0.52 – 4.97 were achieved for *E. coli*, *S, typhimurium* and *L. monocytogenes*, respectively.	(Shin, et al., 2016)

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	L. monocytogenes (ATCC 7644, ATCC 19114, ATCC 19115). *Escherichia coli B* MS-2 *B. atrophaeus* spores	260	6.2 – 58	4-log_{10} reductions were achieved in all microbes.	(Sholtes, et al., 2016)
Surface decontamination					
Media	*E. coli* O157:H7 (ATCC 35150, ATCC 43889, ATCC 43890) *S. Typhimurium* (ATCC 19585, ATCC 43971, and DT 104) *L. monocytogenes* (ATCC 7644, ATCC 19114, ATCC 19115).	275	0.17-1.67	At maximum treatment time (5 min) *E. coli*, *S. typhimurium* and *L. monocytogenes* showed log reductions of 6.32, 6.05 and 5.39 log_{10} CFU/ml, respectively.	(Shin, et al., 2016)
Tube lumen	*Pseudomonas aeruginosa* biofilm	265	-	Disinfection (100%) was obtained in 10 cm Teflon tubes exposed for 30 min.	(Bak, et al., 2010)
Stainless steel Polyvinylchloride Cutting boards	*C. jejuni*	395±5	0.90 – 1.20	No *C. jejuni* recovered from stainless steel or cutting board surfaces after initial inoculum of 2-4-log_{10}.	(Haughton, et al., 2012)
Media	*Candida albicans* CAD1 *E. coli* K12 Biofilms	365	-	Continuous and Pulsed modes with reported intensity of 0.28 mW cm^2 and distance of 20mm resulted in survival rates of <10% for both continuous and pulsed modes for both bacteria with C. albicans showing higher sensitivity.	(J. Li, Hirota, Yumoto, Matsuo, Miyake, & Ichikawa, 2010b)
Media	*Pseudomonas aeruginosa* PAO1	266 296	20,000 *(J/m²)*	For 296 nm treatment - total inactivation of 24 hr grown biofilms and 3-log_{10} inactivation of 48 and 72 hr grown biofilms.	(Argyraki, et al., 2017)
Solid food					
Chicken	*C. coli* 1140 DF, 1662 DF, 2124 GF *C. jejuni* 323 BC, 1135 DF, 1136	395±5	0.90 – 1.20		(Haughton, et al., 2012)

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	DF, 1146 DF, 1147 DF, 1354 DF, NCTC 11168				
Sliced cheese	*E. coli* O157:H7 (ATCC 35150, 43889, 43890) *S*. Typhimurium (ATCC 19585, 43971, and DT104), and *L. monocytogenes* (ATCC 19111, 19115, 15313)	266 270 275 279	1 – 3	By irradiating sliced cheese only for approximately 10 min - 99.99% of the pathogens were inactivated.	(Kim, et al., 2015)
Lettuce Cabbage	*E. coli* DH5α	365	*6.75 x 10^2 J/m^2*	3-$\log_{10}$ CFU/ml reductions were observed after 90 min.	(Aihara, et al., 2014)
Liquid food					
Apple juice	*E. coli* K12 (ATCC 25253)	254 280 254/365 254/405 280/365 280/405 254/280/365/405		Cloudy apple juice treated with 280 and 280/365 nm for 40 min achieved a 2-$\log_{10}$ reduction Clear apple juice achieved a 4.4-$\log_{10}$ when exposed to 4 LEDs emitting at 280 nm for 40 min.	(Akgün, et al., 2017)
Coloured beverages and Orange juice	*E. coli* DH5α	365	*126 J/cm^2*	Reductions of 2-$\log_{10}$ CFU/ml were achieved.	(Lian, et al., 2010)

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Table 3. Overview of chemical changes reported for UV treated foods.

Sample	UV source	Treatment parameters	Significant findings	Reference
Lipid oxidation				
Sliced Ham	SteriPulse-XL®3000 Pulsed UV-light System, XENON Corporation, Worbun, MA.	Variation of pulsed ultraviolet durations (60. 90 and 120 s) and distances of from light source (4.5, 8.3 and 14.6 cm) were applied.	Oxidative stability of all samples decreased over time and decreased with increased distance from lamp.	(Wambura, et al., 2011)
Chicken breasts	Unfiltered germicidal emitting lamps, Sylvania, G15T8, Phillips, Netherlands.	Reported fluences ranged from 0 – 5 kJ/m^2 at distance of 18 cm.	The TBARS values of irradiated chicken breasts decreased slowly regardless of UVC treatment.	(H. Chun, et al., 2010)
Chicken breast tenderloins	Twelve UVC lamps, 6 of 30 W and 6 of 55 W; OSRAM HNS, OFR, Munich, Germany.	Reported intensity range of 0.62 – 1.95 mW/cm^2 at a distance of 14 cm.	Results demonstrated that UV-C radiation did not significantly affect TBARs values.	(Lazaro, et al., 2014)
Protein oxidation				
Whole milk and Skim milk	Mercruy UVC TUV Philips tube 15 W/G15T8; Philips, Eindhoven, Holland.	Reported intensity of 2.34 x 10^{19} quanta/s for 0-24 hr.	Protein carbonyls increased as a function of irradiation time for both Whole and Skimmed milk also carbonyl moieties were detected after 1 hr of UV treatment.	(Scheidegger, et al., 2010)
Micronutrient degradation				
Ananas comosus (Pineapple)	Unfiltered germicidal emitting lamps, FG15T8-15 W T8 120 V germicidal lamp GRM-0152,	Reported fluence of 4.5 kJ/m^2 applied on each side of produce with treatment durations of 0, 60, 90 s.	Treatment decreased vitamin C content significantly.	(Pan, et al., 2012)

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	Atlanta Light Bulbs Inc., Tucker, Georgia, USA.			
Fragaria x ananassa (Strawberries) Preharvest	160 W; Clean Light Inc., Vineland Station, ON, Canada	Reported fluence of 0.6 kJ/m^2), 40 cm distance from UV-C lamps (254 nm) for 15 s (twice weekly, over 3 weeks)	No statistically significant differences in simple sugars (fructose, glucose, sucrose) and organic acids (citric. Malic and ascorbic acids) observed after treatment.	(Xie, et al., 2016)
Apple juice	Four UV-LEDs (254, 280, 365 405 nm) manufactured by SETI Sensor Electronic Technology Inc., Columbia, SC, USA.	Reported intensities (mW/cm^2) of 0.3± 0.0, 0.3 ± 0.0, 0.8 ± 0.1 and 0.4 ± 0.1 for 254, 280, 365 and 405 nm LEDs, respectively.	Ascorbic acid content completely diminished after UV-LED treatment	

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Research highlights

1.UV light has a limited penetration to achieve microbial inactivation.

2.UV LED light-based technologies can achieve food safety.

3.UV LED light technologies can be used for a range of food.

4.Microbial inactivation occurs mainly due to oxidations.

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