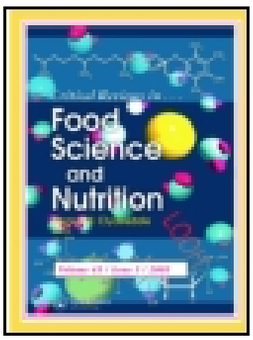

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Critical review of controlled release packaging to improve food safety and quality

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Abstract

Controlled release packaging (CRP) is an innovative technology that uses the package to release active compounds in a controlled manner to improve safety and quality for a wide range of food products during storage. This paper provides a critical review of the uniqueness, design considerations, and research gaps of CRP, with a focus on the kinetics and mechanism of active compounds releasing from the package. Literature data and practical examples are presented to illustrate how CRP controls what active compounds to release, when and how to release, how much and how fast to release, in order to improve food safety and quality.

Keywords

controlled release packaging, antimicrobial packaging, antioxidant packaging, active packaging, target release rate

1 Introduction

Quality loss and microbial safety are major concerns for the food industry and tremendous resources and research efforts have been devoted to address these two issues worldwide. The traditional approach to maintain quality and ensure safety is through adding active compounds such as antioxidants and antimicrobials as ingredients, quickly or instantly, into the food formulation, an approach known as instant addition. Although effective to certain extent, this approach also have limitations due to development of antimicrobial resistance (Zactiti and Kieckbusch, 2009), development of pro-oxidation in lipid foods (Redl et al., 1996), and compliance of regulatory maximum allowable concentration.

Alternatively, when these active compounds are added in a slow release manner, or better still a controlled release manner, superior effect can be achieved. For example, Zhang et al. (2004) found that controlled release of nisin at a constant rate through a syringe pump could be more effective than instant addition to inhibit growth of *Listeria monocytogenes* in growth medium. Zhu et al. (2012b) also showed the same for antioxidant that controlled release of tocopherol at certain constant rates into linoleic acid had better effect than instant addition on inhibitory lipid oxidation in linoleic acid. A question naturally raises: how could controlled release of active compounds be practically achieved in real food systems?

A practical approach to achieve controlled release of active compounds is through controlled release packaging (CRP), a technology to incorporate active compounds such as antimicrobials and antioxidants into the package so that these active compounds can be released in a controlled manner to improve food safety and quality (LaCoste et al., 2005; Yam, 2009). The objective of this paper is to provide a critical review of the uniqueness, design considerations, and knowledge and research gaps of CRP to improve food safety and quality. Knowing the uniqueness of CRP helps identify the appropriate applications, understanding the design considerations helps develop successful CRP systems, and recognizing and filling the knowledge and research gaps are necessary to advance the CRP technology.

2 Uniqueness of CRP

Controlled release packaging (CRP) is a relatively new term first appeared in the literature in 2005 (LaCoste et al., 2005). Why was this new term introduced while the terms antimicrobial packaging and antioxidant packaging already existed? A major reason is to introduce and promote the use of the package as a delivery system to control the release of active compounds to improve food safety and quality, an innovative concept which is substantially different from antimicrobial packaging and antioxidant packaging. CRP may involve antimicrobials and antioxidants as well as other active compounds including flavors, aromas, and plant growth hormones (such as 1-methylcyclopropene to delay fruit ripening), while antimicrobial packaging involves only antimicrobials and antioxidant packaging involves only antioxidants (Figure 1). Research in CRP focuses on releasing systems only, emphasizing on in-depth understanding of the mechanism and kinetics of active compounds releasing from the package. In contrast, research in antimicrobial packaging and antioxidant packaging is more general, involving not only releasing systems, but also non-releasing antimicrobials or antioxidants such as those grafted onto packaging materials, as well as non-releasing systems involving oxygen absorbers and free radical scavengers (Elegir et al., 2008; Gómez-Estaca et al., 2014; Moudache et al., 2016; Moudache et al., 2017; Nerín, 2010; Pezo et al., 2008).

Although the concept of controlled release has been applied to drug delivery and other fields, its application to food packaging is unique due to the requirements of the food and the package. The uniqueness of CRP is that it focuses on the kinetics and mechanism of controlled release—what to release, when and how to trigger the release, how much to release, and how fast to release. What active compounds to release? In addition to single active compounds, combinations of two or more active compounds can also be used to complement one and other; for example, a CRP system may contain two

antimicrobials for different target microorganisms (Lee et al., 2003), or a CRP system may contain both antimicrobial and antioxidant to inhibit microbial growth and lipid oxidation (Lee et al., 2004a). When and how to trigger the release of active compounds? The appropriate time is usually immediately after filling of the food and sealing of the package to avoid premature release occurring before that time. For some CRP systems, moisture from foods such as fresh produce may be used to trigger the release. How much and how fast to release the active compounds? The release rate should closely match the requirement of the kinetics of food deterioration. A rate too slow would result in insufficient amount of active compound to retard food deterioration, and a rate too fast would result in excessive amount of active compounds and their loss due to degradation. Several studies have shown that compared to instant addition, CRP systems may require much less active compounds to achieve the same effects for inhibiting food deterioration (Balasubramanian, 2012; Balasubramanian et al., 2011; Gutiérrez et al., 2010; Manso et al., 2015; Shen, 2012).

Another uniqueness of CRP is that it can have many possible "release rate profiles," defined here as plots of rate of release of active compound versus time. The word "profile" denotes the rate of release of active compound from the package is not constant, but changes with time. In Figure 2, the constant release rate and the diffusion-controlled release rate profile are shown by the dashed line and the dashed curve, respectively, where the area under the line or curve is the total amount of active compound that has been released at a given time. For most CRP systems, the release rates are not constant but change with time. For example, when an active compound is incorporated into a film, its release is often governed by diffusion of the active compound in the film, following the diffusion-controlled release rate profile, characterized by fast release initially with progressively slower release as time passes.

3 Design considerations of CRP

The fundamental concept of CRP is to use the package to modify the concentration of an active compound surrounding the food product inside the package, in order to retard the deterioration of the food and extend its shelf life. It is noteworthy that this concentration may change with time, and consistent to the denotation of the word profile defined above, the term "concentration profile" is defined here as concentration of active compound surrounding the food as a function of time. From a commercial point of view, changing concentration with time has practical value in some situations. For example, it is desirable to have a higher concentration of antimicrobials or antioxidants in the beginning to retard microbial and oxidative deterioration at the beginning to extend shelf life of the product during distribution and storage, but to have a lower concentration when the product reaches the consumer. An important design criteria of CRP is to achieve the optimum concentration profile. How to determine it and how to achieve it? Determining the optimum concentration profile for a specific food item requires the food scientist to conduct a shelf life study using a range of concentration profiles to determine the profile that best fits the distribution and storage environment. Unfortunately, data for determining optimum concentration profiles are scarcely available in the literature. Achieving the optimum concentration profile requires the packaging engineer to develop a package with the desired release rate

profile of active compound. Once the package volume is specified, a mathematical relationship between concentration profile and release rate profile can be obtained through unsteady mass balance analysis. The subsections below describe the design factors that the packaging engineer can select and control to produce desired release rate profiles. Active compound is obviously the first design factor, and its selection depends on efficacy, regulatory compliance, and cost. The second design factor is package composition and structure that form a matrix or carrier, such as a packaging film or a coating, to encapsulate the active compound. The third design factor is the processing methods, such as cast film extrusion and blown film extrusion, which serve the purpose of incorporating the active compound into the packaging polymer and creating an appropriate morphology or structure to allow the release of the active compound. In addition to these design factors, this section also reviews the mathematical models commonly used to summarize the release data, to provide insights of the mechanism (for example, whether the movement of active compound within the polymer matrix follows Fickian diffusion), and to estimate parameter values of diffusivity and partition coefficient. In addition, systems that require trigger to activate release active compounds are reviewed. Target release rate, a concept that connects release kinetics and deterioration kinetics is also discussed.

3.1 Active compounds

Common active compounds used in CRP include mainly antimicrobials for food safety and antioxidants for food quality. There are also attempts to incorporate plant growth hormone regulators, such as 1-methylcyclopropene (1-MCP) and ethylene, to manipulate the ripening process in fresh produce (Mir, 2016; Wood et al., 2017; Wood and Keute, 2016). Certain active compounds can perform multiple functions; for example, some essential oils can be used as both antioxidant and antimicrobial. In recent years, there has been a growing interest to use natural compounds such as essential oil extracts (Albertos et al., 2017; Choulitoudi et al., 2017; Manso et al., 2015) and green tea extract (López de Dicastillo et al., 2011) to replace synthetic compounds.

The molecular shape, size, polarity, and weight of an active compound are important properties that determine its release from the package (Nerin et al., 2016). For example, the release rate profiles of PG, BHT, TBHQ, and BHA from extruded PLA film into ethanol/water mixtures are substantially different due to these properties, with PG having the highest polarity and the fastest release rate among these antioxidants (Jamshidian et al., 2012). This study also reported that although BHT and BHA had about the same polarity, the release of BHA was faster due to its smaller molecular weight.

When two active compounds are incorporated into a packaging polymer, the presence of one compound may influence the release rate of the other compound. Chen el al. (2012b) reported that adding tocopherol to quercetin accelerated the release of quercetin from several packaging polymers due to the plasticizing effect of tocopherol. Graciano-Verdugo et al. (2010) reported that by increasing the concentration of active compounds in packaging film, the release rate was increased due to the plasticizing effect.

The choice of using volatile compounds or non-volatile compounds depends largely on whether there is direct contact between the package and the food. In general, both volatile and non-volatile antimicrobials may be used where there is direct food/package contact (e.g., in a vacuum packed fresh

meat pouch), but only volatile antimicrobials may be used where there is no direct food/package contact (e.g., in a fresh produce bag where there is air space between the produce and the bag). Since microbial inhibition occurs only when the microbes on the food and the antimicrobial from the package come together, it requires that either the microbes to reach the antimicrobial, or the antimicrobial to reach the microbes, or both of them to reach each other.

Where there is direct food/package contact, both volatile and non-volatile antimicrobials can travel toward the microbes, and the microbes can travel toward the antimicrobials, through the liquid/solid or solid/solid interface between the food and the package. Many researchers have studied the use of nonvolatile and volatile antimicrobial for CRP applications where there are direct food/package contact: for example, Dawson et al. (2002) reported that CRP containing non-volatile nisin reduced the microbial count of *Listeria Monocytogenes* on low fat turkey bologna, and Yang et al. (2016) reported that CRP containing volatile clove essential oil reduced the total microbial count on grass carp slice. Where there is no or little direct food/package contact, there is space between the food and the package preventing the microbes and the antimicrobial to reach each other. Since only volatile antimicrobials can travel through space, they must be used where there is no direct food/package contact. Many researchers have studied the use of volatile antimicrobials for CRP systems: for example, Ray et al. (2013) studied the release of chlorine dioxide from PLA film to reduce *Salmonella* spp. and *E. coli* O157:H7 counts on tomatoes, and Pola et al. (2016) studied the release of oregano essential oil released from cellulose acetate film to inhibit fungi growth.

On the other hand, the situation for antioxidants is somewhat different from antimicrobials—non-volatile antioxidant encapsulated in the package can be effective for oxidation inhibition where there is no direct food/package contact. This is because free radicals generated from oxidation and oxygen in the package can travel through space to reach the non-volatile antioxidants in the package. López-de-Dicastillo et al. (2012) showed that EVOH film containing catechin and quercetin was effective to inhibit lipid oxidation in packaged fried peanuts, and Moudache et al. (2017) showed that PE films containing olive leaves extract was effective to inhibit lipid oxidation in fresh pork meat, both by quenching the radical oxidative species in the package headspace. However, such non-volatile and non-releasing antioxidants do not belong to the CRP system. Of course, CRP containing volatile antioxidants is also effective; for example, Zhu et al. (2013) showed that sesamol released from polymeric films into the package headspace was effective to inhibit lipid oxidation of oat cereal.

Due to the mobility of volatile compounds, they are usually more effective than non-volatile compounds for retarding microbial growth and oxidation. However, this mobility can also cause undesirable loss of volatile compounds during processing (such as in film extrusion process where high temperature and shear are involved) and during storage due to premature release of the volatile compound. Lee et al. (2004b) reported that volatile compound BHT had more loss than non-volatile tocopherol during film production and storage. The loss can be reduced by protecting the volatile compounds by encapsulation and barrier layers.

Another important consideration of selecting an active compound is its compatibility (or molecular affinity) with the packaging material, which greatly affects the loading of the active compound and its

release from the packaging material. The compatibility cannot be too high resulting in too slow release, or too low leading to immiscibility of the active compound in the packaging material. For example, Chen et al. (2012b) showed that the release rate of quercetin was much faster from hydrophilic polymers than hydrophobic polymers into 95% ethanol. In hydrophilic materials, the hydrophilic OH groups of quercetin make it miscible in hydrophilic materials, and the hydrophobic ring structure of quercetin prevents it from binding too tightly to the hydrophilic materials, both of which allow quercetin move freely; while in hydrophobic materials, OH groups tend to cluster together resulting in low mobility. Koontz et al. (2010b) also reported similar observation of low release rate of quercetin from hydrophobic material LLDPE film.

Active compounds can also be modified to speed up or slow down their release. Aytac et al. (2017b) conducted a study to encapsulate tocopherol into γ-cyclodextrin and then use electrospinning to produce nanofibers from the encapsulation complex and PLA. They reported that the release rate of tocopherol was faster for the system with γ-cyclodextrin encapsulation than the system without encapsulation because γ-cyclodextrin improved the tocopherol solubility in the aqueous phase of the food simulant. Siró et al. (2006) incorporated tocopherol/β-cyclodextrin encapsulation complex into LDPE film and showed that the encapsulation complex slowed down the release of tocopherol due to the low solubility of β-cyclodextrin in the fatty food simulant. Other examples of incorporating encapsulated active compounds in packaging include HPMC film incorporating lecithin nanoencapsulated nisin (Imran et al., 2012), HPMC film incorporating PLA/green tea extract nanoparticles (Wrona et al., 2017a), and PE film incorporating encapsulated green tea extract (Wrona et al., 2017b).

It should be mentioned that the use of some active compounds is controversial. For example, BHT and BHA had long been added into plastic films as antioxidants for the film, and later their release from wax paper liner was utilized as a means to inhibit lipid oxidation in cereal product in the 1980s (Labuza and Breene, 1989) until the concern of their negative health effect was raised. Triclosan, an antimicrobial widely used in personal care products since 1960s, have been studied for food packaging applications, but this compound has been banned in Europe since 2010 (Espitia et al., 2016).

3.2 Package composition and structure

In CRP systems, the package serves as a reservoir to incorporate the active compound into its structure, to retain the active compound inside its structure, and to release the active compound from its structure at an appropriate time. Incorporating the active compound into the package structure is accomplished by a process, typically involved the driving force of concentration gradient, to force the active compound into spaces inside the package structure larger than the active compound. Retaining the active compound inside the package structure requires that the spaces where the active compound resides are not too large, as well as tortuous paths exist in the package structure, so that the active compound cannot escape easily and is trapped inside the package structure. Alternatively, a barrier layer or coating can be used to retain the active compound. Releasing the active compound from the package structure can be accompanied by an external trigger such as moisture, heat, or removal of the barrier layer. In

addition, the intermolecular forces between the active compound and the package also plays an important role in the incorporation, retention, and release steps above.

3.2.1 Choices of packaging materials

Both synthetic polymers and biobased polymers have been studied for CRP application. Synthetic polymers are usually studied in the form of packaging film produced using the cast film or the blown film extrusion process. Numerous studies have been conducted to establish the release kinetics of active compounds from food grade synthetic packaging films. In recent years, most papers published in the literature involve the use of biobased polymers because they are perceived to be more environmentally friendly than synthetic polymers.

The structure and morphology of biobased polymers can be modified by cross-linking, processing, and other methods to alter the release rates of active compounds. Gemili et al. (2009, 2010) reported that by manipulating the cellulose film morphology from dense to porous, the release rates of L-ascorbic acid, tyrosine, and lysozyme from the films, as well as the partition coefficients, can be altered greatly. Arabi (2012) reported that by manipulating the composition of stereochemical isomers of PLA, as well as processing methods including drying, annealing, solution casting and extrusion, the crystallinity of the resulted film can be controlled and a wide range of tocopherol release rate profiles can be obtained. Rohini (2008) reported that by controlling the degree of crosslinking in low methyl pectin film using calcium ion as a crosslinker, both release rate and the released amount of nisin can be altered.

Chitosan, a biopolymer derived from shells of shrimp and other sea crustaceans, has inherent antimicrobial activity, and incorporating antimicrobial in chitosan can provide additional antimicrobial efficacy. It has been used alone for CRP such as chitosan incorporating acetic and propionic acids (Ouattara et al., 2000) and in combination with other polymers such as blend of chitosan and cellulose incorporating sodium benzoate or potassium sorbate (Chen et al., 1996), and blend of chitosan and konjac glucomannan incorporating nisin (Li et al., 2006). Lee et al. (2003) used short chain chitosan as active compounds and developed an antimicrobial coating containing both chitosan and nisin, which showed antimicrobial efficacy on inhibiting *L. monocytogenes* and *E. coli* O157:H7.

A wide range of combination of biopolymer and active compounds has been studied for CRP application, and selected examples are listed as follows: PLA incorporating thymol (Wu et al., 2014), PLA incorporating antioxidants including BHA, BHT, PG and TBHQ (Jamshidian et al., 2012), methylcellulose edible film incorporating Ferulago angulate essential oil nanocapsules (Esmaeili and Ebrahimzadeh Fazel, 2016), zein film incorporating lauryl arginate (LAE) (Kashiri et al., 2016), cassava starch film incorporating bixin nanocapsules (Pagno et al., 2016), and pectin-carboxymethyl cellulose films incorporating potassium sorbate (Yu et al., 2017), whey protein film incorporating essential oil (Ribeiro-Santos et al., 2017), and starch film with BHT and starch film incorporating green tea extract (u Nisa et al., 2015). Effectiveness of biopolymer based CRP on various food products were also tested such as soy protein film incorporating nisin for turkey bologna (Dawson et al., 2002), pectin film incorporating nisin for ready-to-eat deli meat (Jin et al., 2009a), starch film incorporating potassium sorbate for cheese (López et al., 2013), alginate based film incorporating nisin for beef (Millette et al., 2007), PLA-cellulose film incorporating nisin for ham (Salmieri et al., 2014), starch film incorporating anthocyanins for olive oil

(Stoll et al., 2017), fish myofibrillar protein film incorporating catechin-kradon extract (Kaewprachu et al., 2017), zein film incorporating lysozyme or zein film incorporating mixture of lysozyme, catechin and gallic acid for fresh Kashar cheese (Ünalan et al., 2013).

3.2.2 Polymer blends

Blending two or more different polymers, usually via the extrusion compounding process, is an approach to produce various polymer blend structures and properties in order to achieve desirable release behavior of active compounds. Chen et al. (2012b) reported that quercetin's release rate from hydrophobic polymer LDPE is slower than its release rate from hydrophilic polymer EVA, and its release rates from LDPE/EVA polymer blends were in between the above two release rates. Arcan and Yemenicioğlu (2013) reported that blending carnauba wax into zein based film effectively slowed down the initial release of lysozyme. Arrieta et al. (2014) reported that the release of catechin, an antioxidant, from PLA-PHB film was accelerated by the addition of ATBC as plasticizer.

Polymer blends is also used to improve physical properties of packaging films. Li et al (2006) study on polymer blends of konjac glucomannan and chitosan showed that chitosan concentration altered physical property of the resulted film. Muppalla et al. (2014) reported that clove oil incorporated in CMC–PVA film was effective to improve microbial safety in ground chicken and blending PVA into CMC can improve the physical property of the resulted film.

3.2.3 Multilayer structure

Multilayer film structure is a common approach to achieve "controlled release", with each layer serves a particular function. In multilayer structure, active compounds can be placed into surface layer which is in direct contact with food; for example, LDPE containing 4% tocopherol as direct contact layer was coextruded with HDPE and EVOH for application of milk powder package, in which HDPE and EVOH were used to improve water vapor and oxygen barrier properties for the package (Granda-Restrepo et al., 2009). A common type of active surface layer is achieved through coating technology, and it is important to note that active coating layer may not be a permanent physical layer and may be disintegrated over time. Active compounds can also be placed into the core layer; for example, skin layers without any active compounds were coextruded with core layer containing tocopherol to prevent direct exposure of tocopherol to the ambient environment and thus protect it from oxidation in extruded films (Chen et al., 2012b). Selected examples of multi-layer structure CRP are listed in Table 1.

3.3 Processing methods

Depending on the processing methods, the release behavior of active compounds can be quite different (Jamshidian et al., 2012, 2013). By utilizing different processing methods, a wider range of release profiles can be obtained for various applications. To produce CRP films, traditional processing methods such as cast film extrusion, blown film extrusion, and solvent casting may be used. In recent years, innovative technologies have also been explored for CRP development.

Smart blending is an innovative technology, based on the principle of chaotic advection (a subfield of fluid mechanics), that uses a smart blender attached to one or more extruders to produce polymer blend films with morphologies not achievable by conventional extrusion methods (Zumbrunnen et al.,

2006). The strength of smart blending is its ability to produce unique polymer blend morphologies that can be used to release active compounds at desirable rates (LaCoste et al., 2005). Obinata (2006) used smart blending to study the release of tocopherol from polymer blend films and suggested that the release of tocopherols could be controlled by manipulating polymer compositions and film morphologies. Jin et al. (2009b) also used smart blending to alter the release rates of tocopherols from LDPE/PP and LDPE/HDPE films.

Another innovative technology is electrospinning to produce polymer fibers containing active compounds. The electrospinning equipment consists of a high voltage supply, a syringe with a small needle, an electronic pump to control the syringe, and a metal collecting screen (Teo and Ramakrishna, 2006). In the electrospinning process, a strong electric field is used to charge a polymer solution inside the syringe and force the polymer solution to discharge from the needle; as the discharged solution jet travels in air, its solvent evaporates leaving behind charged fibers to be collected on the metal screen as a nanofiber mat. (Huang et al., 2003). The fibers are in nanoscale typically ranging from 50 to 500 nm. The nanofiber mat has high surface area and porous morphology (Chronakis, 2005) to enable higher loading of active compound. Bio-based food packaging polymers including cellulose and polysaccharides have been produced using electrospinning technology (Schiffman and Schauer, 2008). A research group at Bilkent University has produced both antioxidant and antimicrobial CRP using electrospinning technology (Aytac et al., 2014; Aytac et al., 2017a; Aytac et al., 2017b). A research group at Spanish National Research Council has also produced antimicrobial CRP using electrospun zein fiber incorporating cinnamaldehyde and thymol (Cerqueira et al., 2016; Torres-Giner et al., 2014).

3.4 Mathematical models to analyze release kinetic data

The release of active compounds from a polymeric packaging film involves three steps: (1) molecular diffusion within the film towards the film/food interface, (2) mass transfer across the interface, and (3) dispersion into food or desorption into package headspace (Lee et al., 2008). In most studies reported in the literature, diffusion in the film is assumed to be the slowest and rate determining step.

Mathematical models are useful to summarize release kinetic data, predict release behavior, and provide mechanistic insights. Diffusivity and partition coefficient are two model parameters commonly used to describe the release behavior of an active compound from a polymer film to a food or food simulant. Diffusivity indicates how fast the active compound moves within the film, and partition coefficient indicates how much the active compound is released from the film to the food at equilibrium.

The most popular mathematical models to describe release behavior are derived from differential equations based on one-dimensional Fickian diffusion with appropriate initial and boundary conditions (Crank, 1975). These models typically require the following assumptions: (1) diffusion of active compound in polymer film is the rate determining step in the release process, (2) no structural change in the polymer film during the release process, and (3) the active compound can be readily desorbed from the film into the food, (4) active compound in film is homogeneously distributed initially, (5) initial concentration of active compounds in food is zero, (6) no concentration gradient of active compound exists in food, (7) partition coefficient and diffusivity are constant at a given temperature, (8)

interactions between food simulant and film are absent or negligible, and (9) no degradation of active compound occurs.

Partition coefficient (K) estimation:

$$K_{F,P} = \frac{C_F}{C_P} \text{ or } K_{P,F} = \frac{C_P}{C_F}$$

where C_F and C_P are concentrations of active compound in food and package at equilibrium, respectively.

Diffusion coefficient (diffusivity) estimation

Crank's diffusion model 1:

$$\frac{M_{f,t}}{M_\infty} = 1 - \sum_{n=1}^{\infty} \frac{2\alpha(1+\alpha)}{1+\alpha+\alpha^2 q_n^{\,2}} \exp\left(-\frac{D q_n^{\,2} t}{L_P^{\,2}}\right) \quad (Model\,1)$$

$$\alpha = K_{FP}\frac{V_F}{V_P}$$

where V_F is volume of food, V_P is volume of the package, K_{FP} is partition coefficient, and $q_n s$ are non-zero positive roots of $\tan q_n = -\alpha q_n$, $M_{f,t}$ is mass of active compound migrated to the food at time t, M_∞ is mass of active compound in the food at equilibrium, L_P is the film thickness, D is diffusivity of active compound in the film, and t is time. The estimation of D can be accomplished using some computer programs such as Microsoft Excel and Matlab

When the food simulant is infinite compared to the film. Model 1 can be simplified to Model 2.

$$\frac{M_{f,t}}{M_\infty} = 1 - \sum_{n=1}^{\infty} \frac{8}{(2n-1)^2 \pi^2} \exp\left(\frac{-D(2n+1)^2 \pi^2 t}{L_P^{\,2}}\right) \quad (Model\,2)$$

When $M_{f,t}/M_{P,0}$ is <0.6, Model 2 can be simplified to Model 3:

$$\frac{M_{f,t}}{M_{P,0}} = \frac{4}{L_P}\left(\frac{Dt}{\pi}\right)^{0.5} \quad (Model\,3)$$

where $M_{P,0}$ is the initial loading of active compounds in the package. D can be estimated from the slope of the plot of $M_{f,t}/M_{P,0}$ versus $t^{0.5}$.

Literature reported values of diffusivity and partition coefficient obtained from these mathematical models are summarized in Tables 2-4. Besides single layer films, mathematical models for multilayer films were also available to evaluate the effect of polymer structure on the diffusion of active compounds (Piringer and Baner, 2007).

It is important to validate a model before accepting it. If the model does not fit the data, this fact suggests that the release kinetics does not follow Fickian diffusion and/or some of the model assumptions are violated. For example, liquid from the food or food simulant may cause swelling of the polymer film especially if the film is made of a biopolymer, and the swelling may in turn cause the active compound to move faster and deviate from Fickian behavior. This is the case in which above mentioned assumption 8 is violated, since there is interaction between the food and the film causing the swelling. If the deviation is small, Fickian models may still be marginally acceptable, as in a study where the release of potassium sorbate from swellable sodium alginate film was studied (Zactiti and Kieckbusch, 2009). If the deviation is large, a non-Fickian model accounting for the swelling effect must be used.

The usefulness of these models is limited by their assumptions. For example, virtually all the release studies reported in the literature were conducted using food simulant such as ethanol in a well-stirred container to satisfy the above mentioned assumption 3 that the active compound can be readily desorbed from the film into the food, and the assumption 6 that no concentration gradient of active compounds exists in the food. Although these models provide useful insights of the release of active compound to food simulants, its usefulness is quite limited for describing and predicting the release to real foods.

3.5 Triggered release CRP systems

Triggered release is a desirable feature of CRP systems containing volatile compounds to prevent premature release of the compounds until the release is triggered by an external stimulus. It is particularly useful for gaseous and volatile active compounds. One method for trigger release involve micro or nano-encapsulation, which stabilize the active compound before incorporation into packaging material. Another method for triggered release is to incorporate precursors of an active compound into a CRP system and at an appropriate time, usually immediately before filling and sealing of the package, to trigger a reaction to generate and release the active compound. Examples include SO_2 and ClO_2 CRP systems incorporating SO_2 or ClO_2 precursors to inhibit microbial growth of fresh produce, in which moisture from transpiration of the fresh produce is used to trigger the reaction and release the antimicrobials (Bai et al., 2016; Decco-Italia-srl; Decco-Italia-srl; Ray et al., 2013; Xu et al., 2011). The reactions involved in the systems are shown below.

$S_2O_5^{2-} + 2H^+ = 2SO_2\uparrow + H_2O$ (Reaction 1)

$5ClO_2^- + 4H^+ = 4ClO_2\uparrow + Cl^- + 2H_2O$ (Reaction 2)

In some flavor/aroma release systems, the packaging structure is disrupted or destroyed to trigger the release (Arabi et al., 2012). Selected examples of triggered control system are listed in Table 5.

3.6 Concept of target release rate

To provide guidance for the packaging engineer to design CRP systems, the concept of target release rate was proposed, which is defined as a release rate or a range of release rates of active compounds to effectively inhibit microbial growth or lipid oxidation (Balasubramanian, 2012; Balasubramanian et al.,

2011; Zhu et al., 2012b). Determination of target release rate involves consideration of the food composition, the package, and the distribution and storage conditions.

This definition was first presented by Zhu et al. (2012b) who proved that in order to achieve the maximum inhibition of lipid oxidation in linoleic acid, the release rates of tocopherol cannot be too fast or too slow. Too fast release caused more tocopherol released than linoleic acid free radical produced, and the excess tocopherol formed dimers or other products, making it not available for later stage of lipid oxidation; while too slow release did not supply sufficient tocopherol for inhibiting lipid oxidation. Later, this concept was further developed by Balasubramanian (2012) who found that initial amount of antimicrobial released in the inherent lag period of the target microbial needs to be greater than minimal inhibition concentration (MIC) to achieve high bio-efficacy, and further inhibition can be achieved using smaller amount of antimicrobial. A mathematical model (Model 4) was also developed which takes antimicrobial efficacy (MIC), microbial growth kinetics (lag period) into consideration and correlates them with the release kinetics of antimicrobial from film (diffusivity). Using this target release rate model can help food engineers identify the optimum release rate profile based on the requirement of the food product.

$$\frac{MIC_{IL} \times V_f}{M_{p,0}} = \frac{2 \times A}{V_p} \sqrt{\frac{D \times t_{lag}}{\pi}} \qquad (Model\ 4)$$

where MIC_{IL} is minimum inhibitory concentration for an initial microbial load, $M_{p,0}$ is initial amount of antimicrobial in the packaging film, t_{lag} is time taken for the organism to increase by 1 log, V_f is volume of food, V_p is volume of the package, A is surface area of the polymer, D is diffusivity.

Lee and Yam (2013) also developed a numerical model to identify the target release kinetics of antioxidants and the optimum loading of antioxidant in packaging film based on the reaction kinetics of lipid oxidation. The model was developed by combining a differential equation of hydroperoxides production in lipid oxidation and Crank's diffusion model, and validated using literature data. This research presented a practical example of how to use literature data to design a CRP system.

4 Knowledge and research gaps of CRP

This section summarizes some of the knowledge and research gaps needed to be filled in order to develop commercially viable packages to improve food safety and quality.

4.1 Lack of knowledge about optimum concentration profiles

An important function of CRP is to create an optimum concentration profile of active compound surrounding the food product during distribution and storage. This profile depends on the food deterioration kinetics and the distribution and storage conditions; however, there is a scarcity of useful data in the literature to determine the optimum concentration profiles. Therefore, there is a need to design and conduct experiments to determine optimum concentration profiles for both food simulants and real foods.

4.2 Need for research and development to achieve controlled release

Efforts are needed to develop CRP systems with a wider range of release profiles for various food products. Since most CRP systems reported in the literature seems to release too fast, there is a need to develop systems that can release active compounds in a slower and more sustained manner. However, it should be mentioned that since most release studies reported in the literature were conducted with liquid food simulants in well stirred containers, not conducted with real food under practical distribution and storage conditions—in general, the release of active compound to real foods is much slower than to liquid food simulants.

More precisely, there is a need to develop CRP systems with optimum release profiles corresponding to the optimum concentration profiles of given food products under given storage and distribution conditions. In other words, the design objective of CRP is to match the release kinetics with the food deterioration kinetics, where matching here means that the release rate should not be too slow leading to ineffective inhibition of food deterioration due to insufficient amount of active compound, nor the release rate should not be too high leading to loss of excessive amount of active due to chemical deterioration.

4.3 Lack of release kinetic data for real food products

Although researchers have studied the release kinetics of active compounds into food simulants, the results from those studies are insufficient to predict the release of active compounds into real food products, because real food products are much more complicated than food simulants. There is a need to design and conduct efficient experiments to obtain release kinetic data for real food products.

4.4 Challenges to develop commercial biobased packaging films

The development of new biobased packaging films faces two major challenges. The first challenge is that new biobased packaging films should demonstrate competitive advantage over cellophane, a biobased polymer commercialized over a century ago, before the commercialization of synthetic polymers. Cellophane has excellent mechanical, barrier, and optical properties, better than most new biobased materials proposed in recent journal articles. However, the market of the cellophane has been declining significantly during recent decades due to its higher cost compared to synthetic polymers and its production is considered to be environmental unfriendly because it involves the use of toxic carbon disulfide and other organic solvents. Most new biobased polymer films also suffer from the problem that their production requires toxic or environmental unfriendly solvents.

The second challenge is that information from the literature is of little value to develop commercial biobased films. It is because most studies used the solution casting method to produce films in petri dishes--this is not a commercial viable process to produce large volume of films, and since the morphology and properties of the film depends on the film production process, the usefulness of results from studies using petri dishes and solution casting method is questionable. In addition, commercial equipment for producing biobased film is lacking and need to be developed.

4.5 Regulatory challenges

Incorporation of active compounds into the package may facilitate unintended migration of other packaging additives into food, especially for the systems involving micro and nano-encapsulation of active compounds, as some of the encapsulants may migrate with active compounds into the food. The possible migration of undesirable compounds may cause product safety and regulatory compliance issues, presenting major hurdles that must be dealt with in order to transfer CRP technology from benchtop to reality.

5 Closing remarks

There has been a growing interest to develop controlled release packaging for improving food safety and quality, as evidenced by the publication of numerous journal articles related to this technology in recent years. The uniqueness of this emerging technology is that it uses the package to delivery active compounds to the food product, in a controlled manner, in order to provide more effective inhibition of food deterioration than using the traditional method of instant addition. Attempts for controlled release of active compounds should involve creativity and careful consideration of the design factors (described in Section 3) to best fit the situation. Although this innovative technology may have passed the early development stage and showed great promises to improve food safety and quality, significant knowledge and research gaps (described in Section 4) still exist, and filling those gaps requires research efforts from multidisciplinary teams with areas of expertise in food science, packaging technology, and material science. The commercial success of this technology also requires industry participation, regulatory approval, and market acceptance.

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Abbreviations

1-MCP	1-methylcyclopropene
AA	acetic acid
AITC	allyl isothiocyanate
ATBC	acetyl tributyl citrate
BHA	butylated hydroxyanisole
BHT	butylated hydroxytoluene
CA	cellulose acetate
CD	cyclodextrin
CMC	Carboxymethyl cellulose
EVA	ethylvinyl acetate
EVOH	ethylvinyl alcohol
LAE	lauryl arginate
LMP	low methoxylpectin
LDPE	low density polyethylene
LLDPE	linear low density polyethylene
MFC	microfibrillated cellulose
NC	nitrocellulose
PE	polyethylene
PEO	poly(ethylene oxide)
PG	gallate
PHB	Polyhydroxybutyrate
PLA	poly lactic acid
PO	paraffin oil
PP	polypropolene

PVA	polyvinyl alcohol
PVDC	polyvinylidene chloride
SPI	Soy protein isolate
TBHQ	tert-butylhydroquinone
toc	tocopherol
w/	with

Reference

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Table 1. Examples of multilayer CRP.

Active compounds	Packaging structure	Effectiveness	Reference
Natamycin and/or nisin	PVDC (active) or NC coated on PE	Inhibition of selected microorganisms in cheese	(Hanušová et al., 2010)
Nisin	HPMC (active) coated on PE		(Chollet et al., 2009)
Nisin	Cellulose coated on PE	Inhibition of *L. monocytogenes* in tofu	(Cha et al., 2003)
Nisin or nisaplin	EC/HPMC (active layer)/EC	Food simulant analysis: EC was effective to delay release of nisin	(Guiga et al., 2010)
Nisin or nisaplin	PVC (active layer) coated on PE	Inhibited microbial growth on f cheese Blaťácké zlato	(Hanušová et al., 2009)
Triclosan	Styrene-acrylate (active) coated on paper	Inhibition of *Enterococcus faecalis* in agar	(Chung et al., 2003)
Lysozyme	Layer by layer assembled chitosan organic rectorite and sodium alginate coated on cellulose acetate	Effective to inhibit growth of *Escherichia coli* and *Staphylococcus aureus*, and extend shelf life of pork for 3 days	(Huang et al., 2012)
Sodium benzoate and potassium sorbate	Pectin, pullulan, and chitosan as edible coating	Effective to reduce weight loss, fruit softening, delayed color degradation and titratable soluble solid in strawberry	(Treviño-Garza et al., 2015)
Lysozyme	PVOH/PVOH (active)/PVOH	Inhibition of *Micrococcus lysodeikticus* in sterilized water	(Buonocore et al., 2005)
Ascorbic acid and tocopherol	Tetraethyl orthosilicate and a mixture of alcoxysilane containing organic moieties (active) coated on PA/PE	Inhibition of oxidation shown by ferric reducing antioxidant power assay in EtOH	(Bossi et al., 2016)
Marigold extract	LDPE (active) coextruded with HDPE		(Colín-Chávez et al., 2013)
Sesamol or BHT	(1) LLDPE/HDPE (active)/HDPE, (2) HDPE /HDPE (active)/EVA, (3) HDPE/HDPE (BHT)/EVA	Effective to inhibit lipid oxidation in linoleic acid and breakfast cereal	(Zhu et al., 2013)
Cinnamon oil	Active with or without encapsulation in PVA coated on PP and laminated with LDPE	Effective to repel instar larvae in a retail box containing flour	(Jo et al., 2015)
Chitosan	Thin layer of chitosan	Effective to inhibit growth of yeast	(Castillo et

oligomer	oligomers sandwiched in thermoplastic corn starch film	and mold in strawberries, ricotta and flavored breads	al., 2017)

Table 2: Kinetics data based on Model 1.

Active compounds	Packaging	D ($\times 10^{-10}$ cm^2/s)	K*	T (°C)	Release media	Reference
Potassium sorbate	LMP/CMC 8/2	0.0026	*0.00062*	4	95% EtOH	(Yu et al., 2017)
	LMP/CMC 6/4	0.0068	*0.00148*	4		
	LMP/CMC 4/6	0.0093	*0.0029*	4		
	LMP/CMC 0:10	0.0211	*0.00581*	4		
	LMP/CMC 4:6	0.0150	*0.00505*	25		
	LMP/CMC 4:6	0.0205	*0.0083*	40		
	LMP/CMC 4:6	0.0311	*0.01758*	60		
Thymol	SPI	0.0061	452	5	Olive oil	(Hu et al., 2012)
		0.0199	378	20		
		0.0937	233	40		
		0.322	117	60		
Cinnamal-dehyde		0.001	1445	5		
		0.0046	1350	20		
		0.0215	737	40		
		0.0593	290	60		
Vanillin		0.0345	420	5		
		0.0562	392	20		
		0.0904	341	40		
		0.128	286	60		
Eugenol	SPI	0.003	1155	5	Olive oil	(Chen et al., 2012a)
		0.007	1033	20		
		0.032	644	40		
		0.131	323	60		
Isoeugenol		0.01	535	5		
		0.019	382	20		

		0.137	283	40		
		0.373	106	60		
Astaxanth-in	LDPE	0.354	55.54	23	95% EtOH	(Colín-Chávez et al., 2013)
		0.74	11.95	30		
		3.267	9.04	40		
	HDPE/LDPE	0.153	72.78	10		
		0.412	18.12	23		
		0.559	27.32	30		
		NA	27.14	40		
Lysozyme	CA with various CA/H_2O ratios and porosities	1.50–23.3	234 – 826	4	Water	(Gemili et al., 2009)
L-ascorbic acid	CA with various CA/acetone/ H_2O ratios and porosities	1.67–15	169 – 2439	4	Water	(Gemili et al., 2010)
L-tyrosine		0.17–2.5	343 – 39327	4		
Tocopherol	Ziegler-Natta LLDPE	4.2	83%[$]	30	Coconut oil	(Koontz et al., 2010a)
	LLDPE w/ toc β-CD complex	0.047	9%			
	Metallocene LLDPE	2.0	57%			
	Metallocene LLDPE w/ toc β-CD complex	0.018	7%			
BHT	PLA	0.295	61.14	23	95% EtOH	(Ortiz-Vazquez et al., 2011)
		0.895	20.54	31		

[*] K is calculated by $K_{P,F}$, unless data shown in *italic* is calculated by $K_{F,P}$.

[$] Calculated by amount released from film after reaching equilibrium.

Table 3. Kinetics data based on Model 2.

Active compounds	Packaging	D ($\times 10^{-10}$ cm^2/s)	K*	T (°C)	Release media	Reference
Carvacrol	Soy protein coated paper under various RH (60%-80%)	0.0011–0.075	NA	5	H_2O & n-pentane mixture (50/50)	(Chalier et al., 2009)
		0.0085–0.0878		20		
		0.0171–1.38		30		
Quercetin	EVA	133	*0.01*	RT	95% EtOH	(Chen et al., 2012b)
	EVOH	1.77	*0.023*			
	EVOH w/ toc	63.3	*0.024*			
Tocopherol	EVOH w/que	17.5	*0.059*			
	LDPE w/que	33.0	*0.723*			
Tocopherol	HDPE/EVOH/ LDPE (active)	0.234	27.91	20	Whole milk powder	(Granda-Restrepo et al., 2009)
		0.306	12.58	30		
		0.314	5.26	40		
Tocopherol	LDPE w/ various loadings of toc	0.13–0.14	NA	5	Corn oil	(Graciano-Verdugo et al., 2010)
		0.71–0.96		20		
		3.03–5.11		30		
Natamycin	LDPE coated w/ PVDC/NC	0.79	NA	6	Water	(Hanušová et al., 2010)
		1.03		23		
	LDPE coated w/ NC	0.89		23		
BHA	Extruded PLA	59.9	Infinite	40	95% EtOH	(Jamshidian et al., 2012)
		0.36	ND		50% EtOH	
		0.19	1580		10% EtOH	
BHT		2.66	27.3	20	95% EtOH	
		24.1	Infinite	40	95% EtOH	
		0.27	118		50% EtOH	
		2.97	239	20	95% EtOH	
		0.27	26021		50% EtOH	

PG		176.1	Infinite	40	95% EtOH	
		1.02	ND		50% EtOH	
		0.79	1231		10% EtOH	
		30.9	ND	20	95% EtOH	
		1.5	565		50% EtOH	
Tocopherol	LDPE	0.264	NA	7	95% EtOH	(Heirlings et al., 2004)
	LDPE adsorbed on Syloblock	0.165				
	LDPE adsorbed on SBA-15	0.166				
	EVA	0.423				
Nisin	EVA	1130	NA	10	66% H_2O, 32% PO w/ 2% emulsifier	(Lee et al., 2004a)
Tocopherol	EVA	2910				
Nisin and toc	EVA	930 (nisin)				
		2920 (toc)				
Tocopherol	Ecoflex	983	NA	30	95% EtOH	(Marcos et al., 2014)
	Ecoflex/PLA	458				
Olive leaf extract	Ecoflex	196				
	Ecoflex/PLA	219				
BHT	PLA	19.04	12.61	43	95% EtOH	(Ortiz-Vazquez et al., 2011)
Lysozyme	MFC with various additives	0.15–1.0	NA	6	Water	(Cozzolino et al., 2013)
		0.21–1.2		23		
		0.10–1.2		6	10% EtOH	
		0.20–1.3		23		

*K is calculated by $K_{P,F}$, unless data shown in *italic* is calculated by $K_{F,P}$.

Table 4. Kinetics data based on Model 3.

Active compounds	Packaging	D ($\times 10^{-10}$ cm^2/s)	K*	T (°C)	Release media	Reference
Clove essential oil	EVA	0.25	NA	23	H_2O	(Yang et al., 2016)
		0.30			3% AA	
		0.75			10% EtOH	
		3.2			95% EtOH	
Tocopherol	LDPE	4.60	NA	40	95% EtOH	(Zhu et al., 2012a)
	LDPE/PP (75/25)	0.89				
	LDPE/PP (50/50)	0.195				
	LDPE/PP (25/75)	0.107				
	PP	0.0658				
Cinnamald-ehyde	Gliadin w/ various loading of cinnamaldehyde	0.0488–1.31	NA	20	Released into headspace	(Balaguer et al., 2013a)
Quercetin	LDPE	1.15×10^{-5}	*0.001*	RT	95% EtOH	(Chen et al., 2012b)
	EVA/LDPE 50/50	0.00713	*NA*			
	LDPE w/ toc	2.50×10^{-4}	*0.001*			
Potassium sorbate	κ-carrageenan with various pH	35.3–64.2	NA	40	Sodium phosphate buffer	(Choi et al., 2005)
		26.0–29.8		25		
		10.5–12.9		5		
BHT	LDPE	$(6.24–6.26)\times10^{-2}$	NA	5	Cheese	(Soto-Cantú et al., 2008)
Nisin	Acrylic polymer (active) coated paper	420	1.3%$	10	Water	(Kim et al., 2002)
		130	1.6%		2% NaCl	
		110	1.3%		2% citric acid	
		120	1.3%		2% sucrose	
	EVA (active) coated	930	3.2%		Water	

	paper	680	3.6%		2% NaCl	
		1220	4.1%		2% citric acid	
		1130	3.9%		2% sucrose	
Catechin	PLA	0.019	NA	40	50% EtOH	(Arrieta et al., 2014)
	PLA-ATBC	0.83				
	PLA-PHB	0.26				
	PLA-PHB-ATBC	3.5				

*K is calculated by $K_{P,F}$, unless data shown in *italic* is calculated by $K_{F,P}$.

$Calculated by amount released from film after reaching equilibrium.

Table 5. Examples of CRP system requiring external triggers.

Active compounds	Packaging structure	Trigger	Reference
Allyl isothiocyanate	With or without β-cyclodextrin encapsulation in soy protein/PLA film	Moisture	(Vega-Lugo and Lim, 2009)
1-MCP	With α-cyclodextrin encapsulation grafted on polymer substrate	Moisture	(Wood et al., 2017; Wood and Keute, 2016)
Perillaldehyde	β–cyclodextrin/pullulan electrospun membrane	Moisture	(Mascheroni et al., 2013)
Lysozyme	Electrochemically generated alginate matrix cross-linked with Fe^{3+}	Electric signal	(Jin et al., 2012)
Carvacrol	Soy Protein Coated Paper	Moisture and temperature	(Chalier et al., 2009)
Cinnamaldehyde	Gliadin films	Moisture	(Balaguer et al., 2013b)
Carvacrol	Gluten coated paper	Moisture	(Mascheroni et al., 2011)
SO_2 from $Na_2S_2O_5$	Barrier layer/binding layer containing $Na_2S_2O_5$/food contact layer EVA/LDPE	Moisture	(Xu et al., 2011)
SO_2 from $Na_2S_2O_5$	Three layer paper/coated paper	Moisture	(Decco-Italia-srl)
SO_2 from $Na_2S_2O_5$	Multilayer plastic base pad	Moisture	(Decco-Italia-srl)
SO_2 from $Na_2S_2O_5$	Paper label	Moisture	(Wrona et al., 2015)
ClO_2 from $NaClO_2$ and citric acid	PLA	Moisture	(Ray et al., 2013)
ClO_2 from $NaClO_2$ and tartaric acid	(1) $NaClO_2$ incorporated into ROBOND PS-7850 adhesive coated on a polymer substrate, (2) tartaric acid incorporated into PVOH coated on a polymer substrate. ClO_2 releases by adhering the two films together	Moisture	(Bai et al., 2016)
ClO_2 from $NaClO_2$	Tyvek	Moisture and CO_2	(Zhou, 2015)

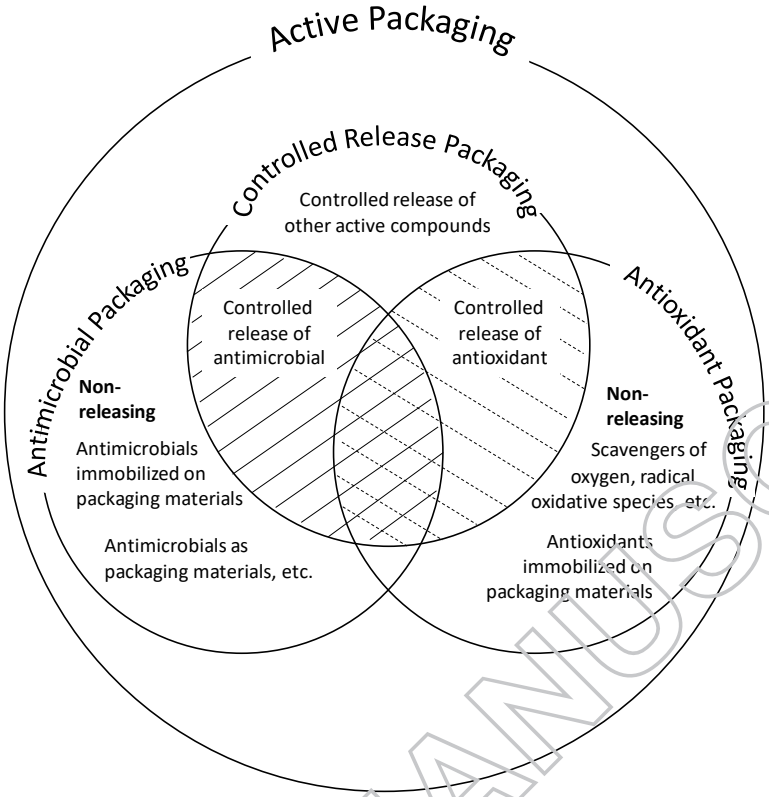

Figure 1: Relationship between different terminologies related to CRP.

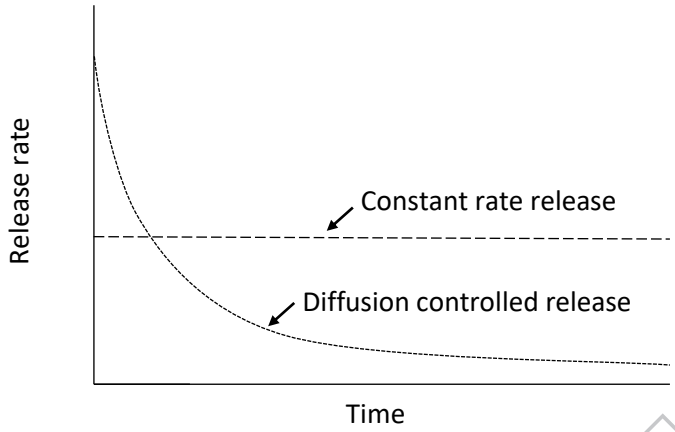

Figure 2. Different release rate profiles of active compounds.