Innovative Food Science and Emerging Technologies 42 (2017) 64–72

Contents lists available at ScienceDirect

Innovative Food Science and Emerging Technologies

journal homepage: www.elsevier.com/locate/ifset

A mathematical model for tailoring antimicrobial packaging material containing encapsulated volatile compounds

Mia Kurek[a], Yannick Laridon[a], Elena Torrieri[b], Valérie Guillard[a,*], Astrid Pant[c], Cornelia Stramm[c], Nathalie Gontard[a], Carole Guillaume[a]

[a] Joint Research Unit Agropolymers Engineering and Emerging Technologies, UMR 1208 INRA/SupAgroM/UM/CIRAD, 2 Place Pierre Viala, 34060 Montpellier, France
[b] Food Science and Agricultural Department, University of Naples Federico II, Via Università 100, 80055 Portici, NA, Italy
[c] Fraunhofer IVV Institute for Process Engineering and Packaging, Giggenhauser Straße 35, Freising, Germany

ARTICLE INFO

Keywords:
Active packaging
Diffusion-reaction
AITC
Experimental validation
PLA

ABSTRACT

A mathematical model describing the water content-dependent release of an antimicrobial agent (allyl isothiocyanate (AITC)) from a bio-based film to the packaging headspace was implemented. The system was characterised experimentally by assessing release kinetics and diffusivities. The model was validated by comparing simulations to experimental data. In spite of the high complexity of the system coupling moisture and antimicrobial diffusion within the packaging material and then release into headspace, the presented model provides a good enough reproduction of experimental conditions. A sensitivity study conducted on the model showed that the release kinetics of the antimicrobial agent were the most influential parameters, and that the diffusivity of moisture and AITC within the film have negligible impact. The model was then used to demonstrate the efficiency of such packaging for shelf-life optimization as it successfully inhibited the growth of bacteria. This work provides a framework that can be used for decision support systems.

Industrial relevance: This work is relevant to industrial considerations as it provides a framework for decision support systems to help manufacturers and researchers to tailor their active packaging. Indeed, the development of anti-microbial applications for food packaging is a time-consuming task, that, if undertaken from a sole experimental point of view, can also be expensive. The use of the simulation framework proposed (that was experimentally validated) helps investigate and compare multiple packaging configurations. Numerical simulation are made by changing the kinetics of release parameters and initial anti-microbial content within the packaging without requiring further experiments, the main issue lying on having plausible values for the parameters.

1. Introduction

Active packaging technologies involve the design and dimensioning of food packaging system for extending food shelf life, while maintaining its quality and safety. Active packaging deliberately incorporates active components intended to release or to absorb substances into, onto or from the packaged food or the environment surrounding the food (Angellier-coussy, Guillard, Guillaume, & Gontard, 2013; Ozdemir & Floros, 2004). They act on different reactions of degradation of food or as a vector of compounds of interest. Antimicrobial packaging acts by direct contact or by emitting some volatile antimicrobial compounds into the headspace to limit microbial growth on the surface of the food. In this last case, the shelf life of the packed food product depends mainly on the composition of volatile compounds in the headspace, which in turn determines the growth rate of microorganisms. This headspace composition is a function of the diffusion of the active agent into the polymer matrix and its release rate from the packaging toward the headspace and of the environmental conditions (e.g. temperature, relative humidity) that could strongly impact the aforementioned transfer rates (Mascheroni, Guillard, Gastaldi, Gontard, & Chalier, 2011). In this context, the dimensioning of the antimicrobial packaging material is meant to adjust the quantity of active agent to add in the material during processing, knowing among others, its diffusivity into the matrix, its release conditions and its expected effect on the targeted degradation reaction (e.g. microbial growth). Mathematical models of mass transfer are very helpful to achieve this task and constitute a real decision support tool for researchers and packaging and food manufacturers.

During the last few years, the research on antimicrobial food packaging material has significantly increased as an alternative method

* Corresponding author.
E-mail address: valerie.guillard@umontpellier.fr (V. Guillard).

http://dx.doi.org/10.1016/j.ifset.2017.05.014
Received 29 March 2016; Received in revised form 19 April 2017; Accepted 31 May 2017
Available online 01 June 2017

Nomenclature

c	concentration ($kg \cdot kg^{-1}$)
j	mass flux ($kg \cdot kg^{-1} \cdot m \cdot s^{-1}$)
h_m	mass transfer coefficient ($m \cdot s^{-1}$)
Bi	Biot number
D	diffusivity ($m^2 \cdot s^{-1}$)
a_w	water activity
k	AITC release rate (s^{-1})
α	pre-exponential release rate term (s^{-1})
β	fitting parameter for release rate
C	Guggenheim constant
K	correction factor
X_m	monolayer moisture content ($kg \cdot kg^{-1}$)
m	mass (kg)
e	thickness (m)
S	surface (m^2)
V	volume (m^3)
IE	inclusion efficiency
MIC	minimum inhibitory concentration ($kg \cdot m^{-3}$)
μ	growth rate (h^{-1})
μ_{max}	maximal growth rate (h^{-1})
N	microbial population (log $CFU \cdot g^{-1}$)
N_{max}	maximal microbial population (log $CFU \cdot g^{-1}$)
I_i	screening index
S_i	first-order Sobol sensitivity index
S_{T_i}	total Sobol sensitivity index
RMSE	root mean squared error

Superscript

u	upper half of desorption cell
l	lower half of desorption cell

Subscript

$AITC-e$	encapsulated AITC
$AITC-f$	free AITC
w	water
f	film
Γ	boundary

to control unwanted microbial growth in foods. The latest progresses in that field have focused on material development with the design of polymeric matrices with tailored mass transfer properties (diffusivity) to control the release (Joo, Merkel, Auras, & Almenar, 2012; Lagaron, Fernandez-Saiz, & Ocio, 2007; Mascheroni, Chalier, Gontard, & Gastaldi, 2010a; Mascheroni et al., 2011; Raouche, Mauricio-Iglesias, Peyron, Guillard, & Gontard, 2011) and on the development of tools based on complex diffusion-reaction systems to predict the antimicrobial release. These approaches aimed at a better design of the antimicrobial packaging by choosing optimal transport parameters for a given situation (Guillard, Issoupov, Redl, & Gontard, 2009; Mascheroni, Guillard, Nalin, Mora, & Piergiovanni, 2010b). Among volatile antimicrobial compounds, allyl isothiocyanate (AITC, a major flavour component of mustard essential oil) has been shown to have a strong antimicrobial activity in its vapour form for very weak added concentrations, compared to others antimicrobial volatile agents (Delaquis & Sholberg, 1997; Raouche et al., 2011). In order to reduce its volatility and its thermal degradation when incorporating it into polymer matrices, usually shaped by using thermo-mechanical processes such as cast-extrusion, AITC can be encapsulated in α- or β-cyclodextrins, prior incorporation in the matrix (Ohta, Takatani, & Kawakishi, 2004).

β-Cyclodextrin (β-CD) is a cyclic oligosaccharide consisting of seven glucopyranose units linked by alpha-(1-4) bonds (Del Valle, 2004). β-CD is most used for its ability to form solid inclusion complexes (host-guest complexes) by molecular complexation with a very wide range of compounds: due to its lipophilic nature, the cavity of β-CD constitutes an appropriate host site for apolar molecules to form inclusion complexes (Fang, Bandaru, Ellis, & Voelcker, 2013). If the complexation is made in aqueous solution, water molecules in the cavity of β-CD are replaced by the more hydrophobic molecules (here AITC) in the solution. This new apolar-apolar association is more stable with a lower energy level (Del Valle, 2004). Once the complex is formed, dried and then rehydrated, the addition of water may cause the breakdown of the system and the release of the encapsulated hydrophobic molecules.

Among possible applications, complexes of β-CD with allyl isothiocyanate (AITC) have been evaluated as a slow-release additive in polylactide-*co*-polycaprolactone (PLA-PCL) biopolymer film packaging. Such encapsulation has been shown to be suitable for long shelf life storage packaging of cheeses (Plackett, Ghanbari-Siahkali, & Szente, 2007; Plackett et al., 2006). The release of AITC from β-CD is known to depend on relative humidity: the higher the relative humidity, the faster and the higher the release. Ponce Cevallos, Buera, and Elizalde (2010) showed that cinnamon and thymol β-CD complexes remained stable up to 75% RH during long storage times (60 days at 25 °C). The guests released from the β-CD complexes were detectable in the region of the water adsorption isotherm at which a sharp increase of water content occurred (84% RH). The authors emphasized that the release of guest molecules was thus governed by the shape of the water sorption isotherm. Li, Jin, and Wang (2007) proposed and validated a release model and showed that depending on the relative humidity (RH) level and on limiting diffusive effects, the release could range from 20% (50% RH and diffusion-limited system) to 100% (non-limiting systems or 98% RH for diffusion-limited systems) of encapsulated AITC. In order to improve knowledge of AITC release mechanisms from polymeric films, it appears thus fundamental to deepen the study of water-β-CD-AITC interactions and especially the impact of water transfer in the film on the diffusion and release rate of AITC. Mathematical modelling of the coupled mass transfer and reaction phenomena prevailing in such a system is crucial to fix its complexity. Release of compounds from β-CD was extensively studied in simplified experimental conditions such as aqueous solutions (for instance Ohta, Takatani, & Kawakishi, 2000; Reineccius, Reineccius, & Peppard, 2003), but as far as we know few of these approaches were then integrated in a full packaging concept (including polymer matrix containing encapsulated active compound and headspace).

In the context of active packaging research, models can provide a better understanding of the interplay between the mechanisms involved. However, the predictive ability of such models depends highly on an adequate experimental determination of input parameters. Huang, Ye, and Yam (2013) proposed a general model for active packaging, but it was not designed for encapsulated compounds and lacked experimental validation. Cerisuelo et al. (2012, 2013) have developed a mathematical model to describe the moisture-dependent release of carvacrol in a hydrophilic EVOH coating on PP film for packaging salmon fillets. Their modelling approach helped the optimization of the package design and the identification of the best environmental conditions that would lead to the achievement of maximum packaging efficiency. In this respect, the use of decision support tools based on mathematical models coupling mass transfer occurring in the packaging (such as diffusion and release of additives, and scavenging of certain species) and reaction in the food product (such as microbial growth, oxidation, and respiration) is helpful to simplify the package design steps by allowing predicting in advance the packaging