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Performance of a non-invasive methodology for assessing oxygen diffusion in liquid and solid food products

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ABSTRACT

Based on the measurement of local oxygen partial pressure kinetic, a non-invasive methodology was proposed to assess O_2 diffusivity (D_{O_2}) in liquid, viscous and solid matrices. This new method was compared with a previous invasive method, developed by the same group, based on the same principle. The new method has proven to be essential to measure D_{O_2} in solid food matrices where invasive methods usually failed. It was successfully used to obtain D_{O_2} of cooked ham and processed cheese which were found respectively equal to $0.450 \pm 0.004 \times 10^{-9}$ $m^2 \cdot s^{-1}$ and $1.15 \pm 0.11 \times 10^{-9}$ $m^2 \cdot s^{-1}$ at 20 °C. D_{O_2} was also evaluated as a function of temperature (from 5 to 30 °C) and viscosity in lipid-based matrices. These results have permitted to determine activation energy of the diffusion and have revealed that increasing viscosity of the lipid matrices tested did not impact their D_{O_2} values.

1. Introduction

Oxygen, promoting most of the food degradation reactions, is generally considered arprejudicial to the long shelf life of most of foodstuffs. The packaging system tends to remove and/or control the oxygen from the headspace to avoid these reactions, by using, for example, Modified Atmosphere Packaging (MAP). In MAP systems, O_2 permeates through the packaging material from the external atmosphere toward the headspace and then, diffuses from the headspace into the food. The mastering of the O_2 level into MAP relies on the selection of packaging materials with suitable O_2 permeability and by controlling O_2 dissolution/diffusion into the product by adding, for instance, antioxidants. There are mathematical models of mass transfer aiming at predicting the evolution of O_2 content in the food/packaging system, thus permitting to properly dimension and design the system (*e.g.* calculation of the initial quantity of antioxidant to add in the food etc.) (Bacigalupi et al., 2013; Cagnon et al., 2013; Chaix et al., 2015; Pénicaud et al., 2011, 2009). In all cases, the interest of those models is restricted to the available input parameters for mass transfer, such as diffusivities. Recent reviews of the literature have revealed the scarcity of data concerning the diffusivity of O_2 (D_{O_2}) one of the most important parameters required for the modelling of O_2 transfer in food (Chaix et al., 2014; Pénicaud et al., 2012).

Oxygen diffusion in food is usually described by the well-known Fick's second law (Fick, 1855). This equation connects the variation of O_2 concentration in food samples with time and the spatial variation of concentration according to the diffusivity coefficient, D_{O_2}. Obtaining D_{O_2} needs three steps: (a) an experimental approach that enables to obtain O_2 variation according to either time or position in the food, (b) a mathematical solution of Fick's 2nd law, with initial and boundaries conditions which represents well the experimental set-up to model O_2 transfer in the food, and (c) the good fitting of this model to the experimental data by adjusting D_{O_2}. Therefore D_{O_2} is not measured but identified. It involves having a mastered set-up and methodology to obtain O_2 sorption/desorption in food, and an appropriate mathematical model. D_{O_2} is thus not easy to acquire, especially in solid food products, due to the difficulty of treating such a type of material with the existing technologies of O_2 monitoring generally more suitable for liquids (Chaix et al., 2014).

Most of the D_{O_2} data of the literature were obtained in liquids such as water, salted water, synthetic or natural oils, fruit juices, etc.

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Acronyms and abbreviations

μ	dynamic viscosity
B	bias between the two methods
$C_{O_2,F}$	concentration of dissolved O_2 in sample ($mol \cdot kg^{-1}$)
D_0	pre-exponential factor (Arrhenius equation)
D_{O_2}	diffusion coefficient of O_2 ($m^2 \cdot s^{-1}$)
D_{O_2-I}	from invasive device
D_{O_2-NI}	from non-invasive device
e	thickness of sample (m)
Ea	activation energy ($J \cdot mol^{-1}$)
i	point on the grid in discretized expression of 2nd Fick's law
m	number of data in RMSE calculation
MAP	modified atmosphere packaging
n	number of subregions in sample (thickness equal Δx)
N_2	nitrogen
O_2	oxygen
ODE	ordinary differential equations
p	number of matrices to Bias calculation
$P_{O_2,F}$	food partial pressure of O_2 (Pa)
$P_{O_2,F-pred}$	predicted
$P_{O_2,F-exp}$	experimental
$P_{O_2,HS}$	headspace partial pressure of O_2 (Pa)
$P_{O_2,ini}$	initial food partial pressure of O_2 (Pa)
R	gas constant ($8.314\ J \cdot mol^{-1} \cdot K^{-1}$)
RH	Relative Humidity
RMSE	Root Mean Square Error
t	time (s)
T	temperature (K)
x	distance between interface and measurement point (m)

These values were rare in solid products where only a few papers on the topic could be found. For example, some D_{O_2} values could be found for beef muscle (Noriega et al., 2008; Zaritzky and Bevilacqua, 1988) for agar gel (Adlercreutz, 1986; Miller et al., 2003; Pénicaud et al., 2010; Sato and Toda, 1983), copra oil (Pénicaud et al., 2010) and lard (Davidson and Cullen, 1957). But most of these products were models and not real food matrices. Whatever the kind of product (liquid or solid, model or real), D_{O_2} was found to vary a lot for a given matrix from one study to another, leading to difficult comparison between data. For instance, analysis of seven different papers of the literature revealed that, in water, O_2 diffusivity would vary at 20 °C between 1.7 and 2.5×10^{-9} $m^2 \cdot s^{-1}$ (Chaix et al., 2014) (Fig. 1).

Faced with the experimental difficulty to obtain D_{O_2}, predictive modelling would be interesting to represent D_{O_2} in food as a function of various parameters. But, in spite of the high interest of this approach, very few modelling trials have been attempted. As regards the prediction of the impact of temperature, the well-known Arrhenius equation is generally used (Equation (1)).

$$D_{O_2} = D_0 \exp\left(\frac{-Ea}{RT}\right) \quad (1)$$

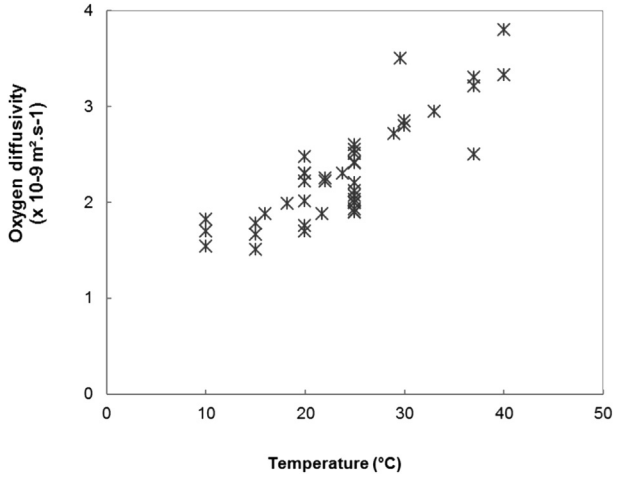

Fig. 1. Oxygen diffusivity of water for temperature ranging from 10 to 40 °C from literature data, as a function of temperature (Data from Langø et al. (1996) and Pénicaud et al. (2010)). Diffusivity data could be uploaded at : http://ceres.agroparistech.fr/atWeb/TableServlet?viewTable=2765&idDoc=1342&id=35116534.

where D_0 is the pre-exponential factor ($m^2 \cdot s^{-1}$), Ea is the activation energy ($J \cdot mol^{-1}$), R is the universal gas constant ($8.314\ J \cdot mol^{-1}\ K^{-1}$), and T is the absolute temperature (K). In theory, the increase of temperature should increase the diffusion of O_2. However, analysis of literature data for D_{O_2} of water has revealed a high variability (Fig. 1) preventing us from calculating any activation energy. Regarding the original values of Ea for D_{O_2} of food products, there were, as far as we knew, no other values than those presented by Simpson et al. (2004). These authors calculated activation energy between 18 and 25 °C (equal to 20.3 $kJ \cdot mol^{-1}$) for D_{O_2} of gelatine from values of D_{O_2} estimated from the fraction of water content in the matrix and D_{O_2} in the water, taken from literature.

Another modelling approach, the Wilke–Chang equation (Wilke and Chang, 1955), related D_{O_2} to the absolute temperature (T) and viscosity (μ) of the medium (Equation (2)).

$$\frac{D_{O_2} \times \mu}{T} = \text{constant} \quad (2)$$

Equation (2) was initially established for predicting D_{O_2} of water, organic liquids, salt and glucose solutions (Jamnongwong et al., 2010; Schumpe and Luehring, 1990) and its extrapolation to solid matrices remains questionable principally faced with the difficulty to express "a viscosity" for solids. It was, nevertheless, used to estimate D_{O_2} of solid foods: for example, Zaritzky and Bevilacqua (1988) estimates D_{O_2} in muscle tissues at 0, 5 and 10 °C respectively from D_{O_2} measured at 37 °C (equal to 1.7×10^{-9} $m^2 \cdot s^{-1}$), by considering the impact of the temperature on the viscosity of the water phase of the product. D_{O_2} in beef was also estimated equal to 0.59, 0.71 and 0.84×10^{-9} $m^2 \cdot s^{-1}$ for 0, 5 and 10 °C. With exactly the same assumptions and same equation, Noriega et al. (2010) estimated D_{O_2} of minced chicken breasts (1.3×10^{-9} $m^2 \cdot s^{-1}$ at 25 °C). In the aforementioned studies, experimental validations of calculations were never performed, making it difficult to conclude about the suitability of the extrapolation of the Wilke–Chang equation to solid food products.

In this work, two methodologies were used to obtain oxygen diffusivity of several model and real food products, liquid or solid ones. To avoid possible bias of invasive methodologies in the measurement of oxygen sorption/desorption in dense product, a non-invasive method was proposed as an alternative to the previous one, which was invasive, described by Pénicaud et al. (2010). This non-invasive method was first validated and then specifically applied to obtain O_2 diffusivity coefficient in solid and dense media.