THIN LAYER CONVECTIVE DRYING OF *CYSTOSEIRA BARBATA* BROWN MACROALGA

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Drying kinetics of Cystoseira barbata brown macroalga were measured and predicted. Thin layer convective drying experiments were conducted in a thermobalance at 50-80 °C and a dehydrator at 55 °C. Drying kinetics were predicted using semi-theoretical kinetic models based on Fick's second law.

Values of effective diffusivity (0.761-1.876×10^{-9} m^2/s), rate constant (0.961-3.040 h^{-n} for Page model, 0.997-2.581 h^{-1} for Newton and Handerson and Pabis (HP) models), and activation energy (26.58-37.39 kJ/mol) were determined for drying in the thermobalance. Page, Newton, and HP models based on kinetic parameters obtained for drying in the thermobalance predicted accurately experimental drying kinetics in the dehydrator. Page model was used to simulate the operation of a conveyor belt dryer with infrared radiation heating.

Keywords: air drying, diffusion coefficient, kinetic model, macroalga, *Cystoseira barbata*

1. Introduction

Macroalgae are valuable sources of biochemicals and biomaterials for the food industry, cosmetics, pharmaceuticals, and nutraceuticals as well as of biofuels [1-10]. Solvent extraction is the traditional technique used to recover different algal compounds, including polysaccarides (alginates, agar, carrageenans, ulvans, fucoidans, laminarans), pigments (β-carotene, lutein, zeaxanthin, fucoxanthin, tocopherol, chlorophyll *a*), sterols (β-sitosterol, fucosterols), polyunsaturated fatty acids (PUFAs), polyphenols, glycoproteins [2,4,6,11]. Biochemical (fermentation, anaerobic digestion) and thermochemical (gaseification, combustion, pyrolysis, hydrothermal liquefaction) routes are usually applied to produce biofuels, *e.g.*, bioethanol, biobutanol, biogas, bio-oil,

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bio-char, from macroalgae [1,3-15]. Extraction of compounds or energy from macroalgae can be performed using either dried or wet biomass.

Air convective drying at temperatures of 25-70 °C is widely used to dried macroalgae prior their processing [16-19]. Drying process is applied to stabilize macroalgal biomass and its compounds by inhibiting the growth of micro organisms and deterioration reactions as well as to reduce the storage volume and transport costs [16-18,20]. Drying kinetics mainly depend on biomass characteristics (structure, shape, thickness) and air stream properties (temperature, pressure, humidity, velocity).

The modelling is a powerful tool in designing, controlling, and optimizing the drying process, predicting its performances, and understanding characteristic phenomena [20-22]. Mathematical models describing the drying dynamics in a continuum porous structure are divided into theoretical, semi-theoretical, and empirical. Theoretical models, based on the differential equations of mass, heat, and momentum balance, are more realistic but contain various process parameters which are often determined by expensive and time-consuming experiments. On the other hand, semi-theoretical models based on Fick’s second law of diffusion and empirical models are widely used to predict thin layer drying kinetics [16-19,20,22-26].

This paper aims at studying the thin layer convective drying of *Cystoseira barbata* brown macroalga in two equipments. Drying experiments were conducted in a thermobalance at 50-80 °C and in a food dehydrator at 55 °C. Characteristic dynamics of convective drying were predicted using semi-theoretical models based on Fick’s second law of diffusion. Kinetic model parameters obtained for drying in the thermobalance were used to predict drying dynamics in the dehydrator as well as in a conveyor belt dryer with infrared radiation heating.

2. Materials and methods

Seaweed material

Cystoseira barbata species was harvested from the southern part of Romanian Black Sea shore in Mangalia (GPS coordinates: latitude N 43° 49.2', longitude E 28° 35.4'), washed with tap water and then with distilled water in order to remove the sand and epiphytes from the surface, chopped to uniform size of about 2 cm length, and stored at 4 °C prior to drying experiments.

Equipments and procedures

Thin layer drying experiments of seaweed were performed using a MB23-Ohaus thermobalance (OHAUS, Parsippany, New Jersey, USA) and a Tribest Sedona Express SDE-P6280 food dehydrator (TRIBEST, Anaheim, California, USA). Air drying tests were conducted at different levels of process temperature, *i.e.*, 50 °C, 60 °C, 70 °C, and 80 °C for thermobalance and 55 °C for dehydrator, until the equilibrium was attained. Macroalga layer thickness was of about 4 mm in all experiments.

Dimensionless moisture ratio was determined by Eq. (1), where X is the moisture content expressed as mass ratio, X_0 and X_e are initial and equilibrium moisture content, respectively. Values of X_0, which were measured at 120 °C using MB23-Ohaus thermobalance, were in the range of 4.9-6.7 kg_w/kg_{dm}, corresponding to moisture mass fractions of 0.83-0.87 kg_w/kg_{wm}, where the subscripts w, dm, and wm represent water, dry and wet matter, respectively.

$$MR = \frac{X - X_e}{X_0 - X_e} \tag{1}$$

3. Modelling

Thin layer drying dynamics were predicted using four semi-theoretical model based on Fick's second law of diffusion, including a model obtained by simplifying the general solution of Fick's second law (a model called Fick II and given by Eq. (2)) and three models derived by modifying this simplified model, *i.e.*, Page (Eq. (3)), Newton (Eq. (4)), and Henderson and Pabis (HP) (Eq. (5)) [16-19,20,22-26]. Model parameters in Eqs. (2)-(5) are as follows: L (0.004 m) is the thickness of seaweed layer, τ (s) the time, D_{eff} (m^2/s) the effective diffusivity, a, k, and n are kinetic parameters in Eqs. (3)-(5).

$$MR = \frac{8}{\pi^2} \exp\left(-\frac{\pi^2}{L^2} D_{eff} \tau\right) \tag{2}$$

$$MR = \exp\left(-k\tau^n\right) \tag{3}$$

$$MR = \exp\left(-k\tau\right) \tag{4}$$

$$MR = a\exp\left(-k\tau\right) \tag{5}$$

4. Results and discussions

Experimental drying curves

Experimental macroalga drying curves, *i.e.*, MR *vs* τ, r *vs* τ, and r *vs* MR, where r (kg_w/kg_{dm}/h) represents the drying rate defined by Eq. (6), are shown in Figs. 1-3. Dynamics of macroalga MR (Fig. 1) reveal a decrease in MR and final drying time, τ_f (70-140 min), with an increase in the process temperature, t (50-80 °C), and similar time variations of MR for macroalga drying at 70 °C and 80 °C. Experimental data represented in Figs. 2 and 3 highlight larger values of r for low levels of τ and high levels of MR and t as well as similar r values for drying at 70 °C and 80 °C. Moreover, results shown in Figs. 2 and 3 indicate a dominant falling rate period, suggesting that the process is controlled by the internal mass transfer resistance [25].

$$r = \frac{X_{\tau} - X_{\tau+\Delta\tau}}{\Delta\tau} \tag{6}$$

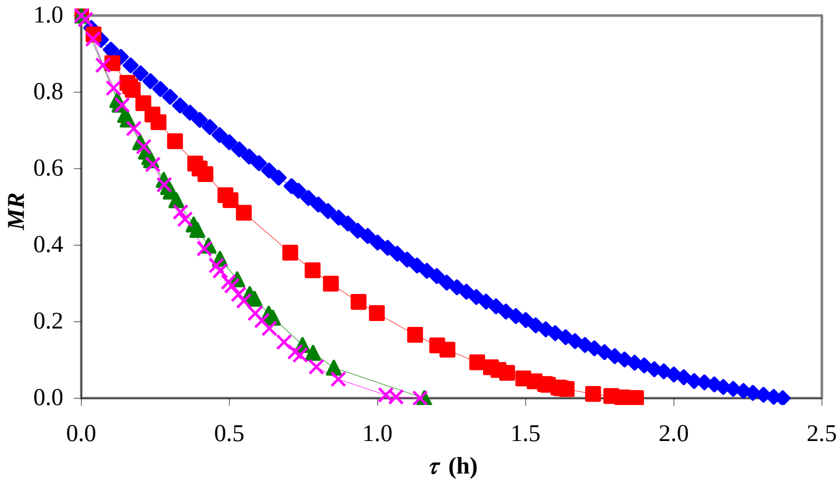

Fig. 1. Macroalga moisture ratio *vs* time for drying in the thermobalance at different values of drying temperature: ♦ 50 °C, ■ 60 °C, ▲ 70 °C, × 80 °C

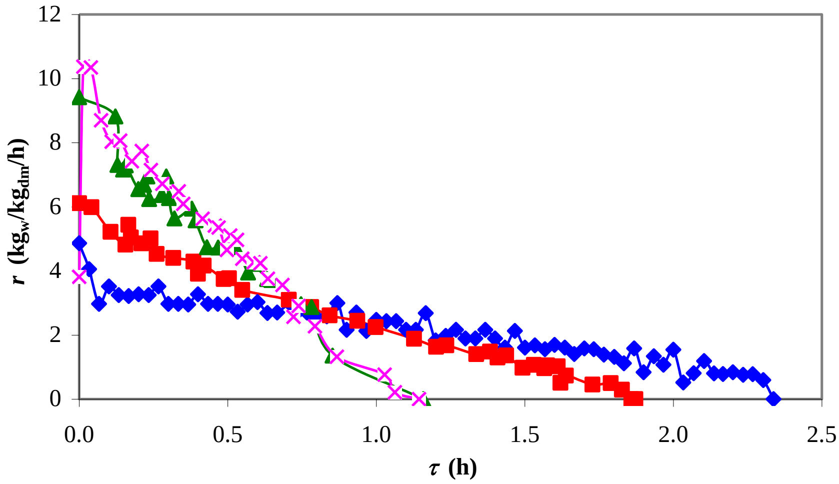

Fig. 2. Macroalga drying rate *vs* time for drying in the thermobalance at different values of drying temperature: ♦ 50 °C, ■ 60 °C, ▲ 70 °C, × 80 °C

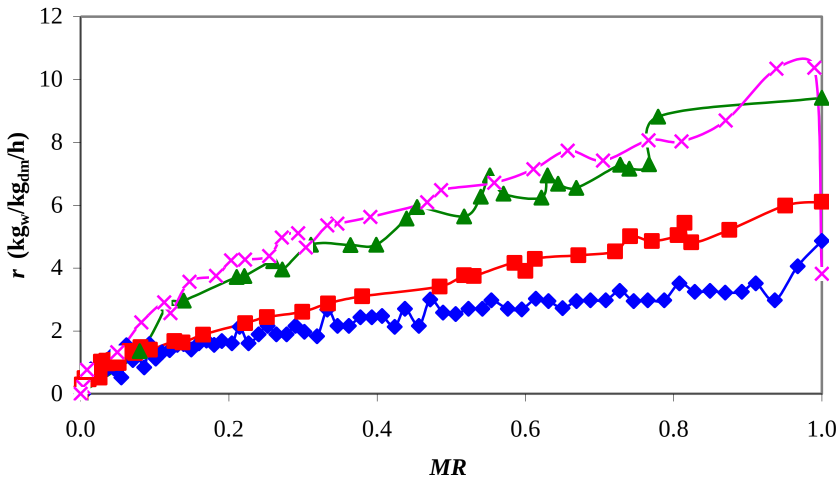

Fig. 3. Macroalga drying rate *vs* moisture ratio for drying in the thermobalance at different values of drying temperature: ♦ 50 °C, ■ 60 °C, ▲ 70 °C, × 80 °C

Predicted drying curves

The values of kinetic parameters (Table 1) obtained by fitting the experimental data, which are summarized in Table 1, reveal the following issues: (i) values of D_{eff} ((0.761-1.876)×10^{-9} m^2/s) and k (0.961-3.040 h^{-n} in Page model, 0.997-2.429 h^{-1} in Newton model, and 1.073-2.581 h^{-1} in HP model) increase with t (50-80 °C); (ii) values of k in Newton and HP models are almost similar at each temperature level; (iii) values of exponent (n=1.176-1.323) in Page model and pre-exponential factor (a=1.052-1.075) in HP model are almost identical for the temperature range considered in the experimental study, their mean values being of 1.257 and 1.064, respectively. Moreover, the values of kinetic parameters are within the ranges which are usually reported for air drying of thin layer biomass [20,22-26].

Tabel 1 contains also the values of root mean square errors (RMSE) determined based on experimental and predicted data. Tabulated results highlight that Page model predicted better the experimental dynamics (RMSE=0.018-0.026) than Fick II (RMSE=0.159-0.213), Newton (RMSE=0.033-0.053), and HP (RMSE=0.028-0.047) models. Experimental drying dynamics, *i.e.*, *MR vs τ*, and those predicted by Page model (Eq. (3)) are shown in Fig. 4.

Arrhenius equation (Eqs. (7) and (8)) was selected to express the variation of D_{eff} and k with drying temperature, where E_a is the activation energy for moisture diffusion process, R (8.314 J/mol/K) the ideal gas constant, T (K) the absolute temperature, $D_{eff,0}$ (m^2/s) and k_0 (h^{-n} for Page model and h^{-1} for Newton and HP models) the effective diffusivity and rate constant at infinite temperature. Values of Arrhenius parameters, which were determined from the slope and intercept of the straight line given by plotting lnD_{eff} *vs* 1/T and ln$k$ *vs* 1/T, are summarized in Table 2.

$$D_{eff} = D_{eff,0} \exp\left(-\frac{E_a}{RT}\right) \tag{7}$$

$$k = k_0 \exp\left(-\frac{E_a}{RT}\right) \tag{8}$$

The data obtained for the drying in the thermobalance, *i.e.*, values of D_{eff} and k specified in Table 2 and mean values of exponent (n_{mean}=1.257) in Page model and pre-exponential factor (a_{mean}=1.064) in HP model, were used to predict drying dynamics in the dehydrator. The results shown in Fig. 5 indicate a good agreement between experimental values of MR for the drying in the dehydrator at 55 °C and those predicted by Page (RMSE=0.025), Newton (RMSE=0.027), and HP (RMSE=0.024) models for the drying in the thermobalance at 50-80 °C.

Table 1

Values of model kinetic parameters and root mean square errors

Temperature (°C)	Model	Parameter symbol (unit)	Parameter value	RMSE
50	Fick II	D_{eff} (m^2/s)	0.761×10^{-9}	0.213
	Page	k (h^{-n})	0.961	0.026
		n	1.323	
	Newton	k (h^{-1})	0.997	0.053
	HP	k (h^{-1})	1.073	0.047
		a	1.075	
60	Fick II	D_{eff} (m^2/s)	1.132×10^{-9}	0.195
	Page	k (h^{-n})	1.621	0.021
		n	1.245	
	Newton	k (h^{-1})	1.508	0.046
	HP	k (h^{-1})	1.625	0.039
		a	1.071	
70	Fick II	D_{eff} (m^2/s)	1.252×10^{-9}	0.159
	Page	k (h^{-n})	2.594	0.018
		n	1.176	
	Newton	k (h^{-1})	2.204	0.033
	HP	k (h^{-1})	2.349	0.028
		a	1.052	
80	Fick II	D_{eff} (m^2/s)	1.876×10^{-9}	0.212
	Page	k (h^{-n})	3.040	0.018
		n	1.284	
	Newton	k (h^{-1})	2.429	0.043
	HP	k (h^{-1})	2.581	0.037
		a	1.060	

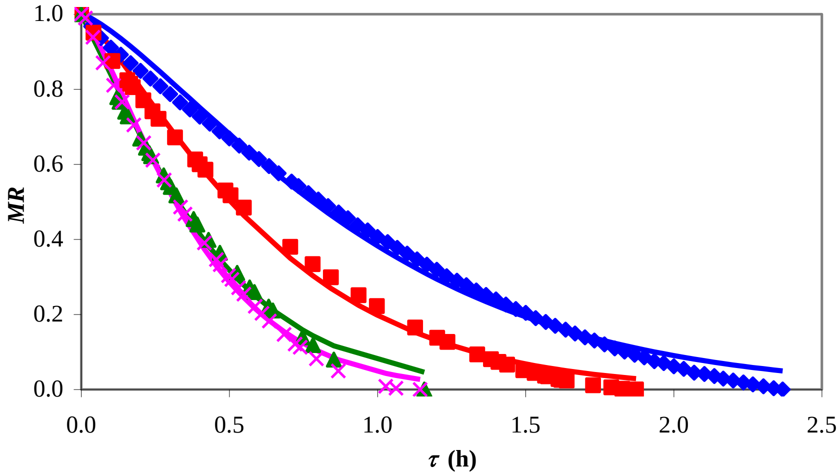

Fig. 4. Macroalga moisture ratio *vs* time for drying in the thermobalance at different values of drying temperature: ♦ 50 °C, ■ 60 °C, ▲ 70 °C, × 80 °C
(bullets: experimental, lines: Eq. (3) (Page model))

Table 2

Values of Arrhenius equation parameters

Model	Activation energy		Pre-exponential factor		R^2
	Symbol (unit)	Value	Symbol (unit)	Value	
Fick II	E_a (kJ/mol)	26.58	$D_{eff,0}$ (m^2/s)	1.55×10^{-5}	0.955
Page		37.39	k_0 (h^{-n})	1.14×10^6	0.965
Newton		29.08	k_0 (h^{-1})	5.32×10^4	0.954
HP		28.62	k_0 (h^{-1})	4.83×10^4	0.952

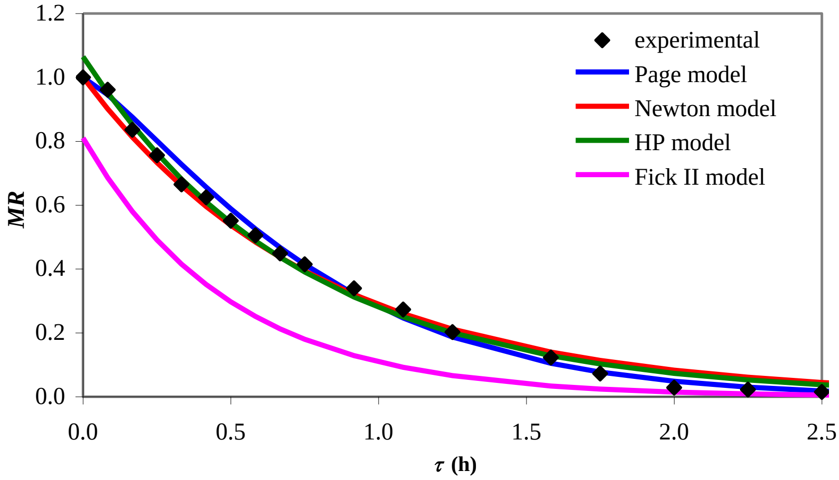

Fig. 5. Macroalga moisture ratio *vs* time for drying in the food dehydrator at 55 °C

These semi-theoretical models can be used to simulate the variation of macroalga moisture content along the length of a conveyor belt dryer with infrared radiation heating (CBDIR) [27]. Algal biomass could be quickly dried in a CBDIR at temperatures of 20-60 °C, which are generally recommended in order to not deteriorate its active compounds [15-19]. For instance, spatial variation of macroalga *MR* can be predicted based on characteristic parameters of Page model (E_a, k_0, and n) according to Eq. (9), where d (m) is the distance measured from the conveyor belt input, w_b (m/h) the linear velocity of conveyor belt, and T (K) the absolute temperature.

$$MR = \exp\left(-k_0 \exp\left(-\frac{E_a}{RT}\right)\left(\frac{d}{w_b}\right)^n\right) \tag{9}$$

Equilibrium water mass fraction of dried *Cystoseira barbata* macroalga, x_e (kg_w/kg_{wm}), was measured at different levels of air temperature, t (°C), and relative humidity, *RH* (%). Experimental equilibrium data, which are shown in Fig. 6, reveal that the values of x_e are in the ranges reported in the related literature [28-30]. Experimental equilibrium results of *Cystoseira barbata* macroalga species were fitted using a modified Oswin model expressed by Eq. (10) [30], where a, b, and c are adjustable parameters. Model parameters, which were determined by minimizing the root mean square error (*RMSE*) between experimental and predicted values of x_e, were as follows: a=10.01, b= -0.050 $(°C)^{-1}$, and c=0.391. Results presented in Fig. 7 highlight a good agreement between experimental and predicted data (*RMSE*=0.002). Values of equilibrium water mass ratio of dried macroalga, X_e (kg_w/kg_{dm}), were determined by Eq. (11) depending on those of equilibrium water mass fraction, x_e (kg_w/kg_{wm}).

$$x_e = \frac{(a+bt)}{100}\left(-\frac{RH}{100-RH}\right)^c \tag{10}$$

$$X_e = \frac{x_e}{1-x_e} \tag{11}$$

Variations of macroalga water mass ratio (X) along the length of conveyor belt, which were simulated using Eqs. (9)-(12) for different values of air temperature (t) and relative humidity (*RH*), are shown in Fig. 8. Depicted results indicate lower values of X for higher levels of t and lower levels of *RH*. Moreover, the effect of temperature is significant. Similar simulation procedures can be developed for all semi-theoretical models considered in this study. These procedures are important because for a real drying process in a CBDIR, the air temperature and relative humidity are random external process factors and their measurement allows a rapid control of the process (*e.g.*, by changing the belt velocity and IR power) in order to obtain an exit moisture content of algal biomass (X_{exit}) close to that corresponding to the equilibrium state (X_e).

$$X = X_e + MR(X_0 - X_e) \tag{12}$$

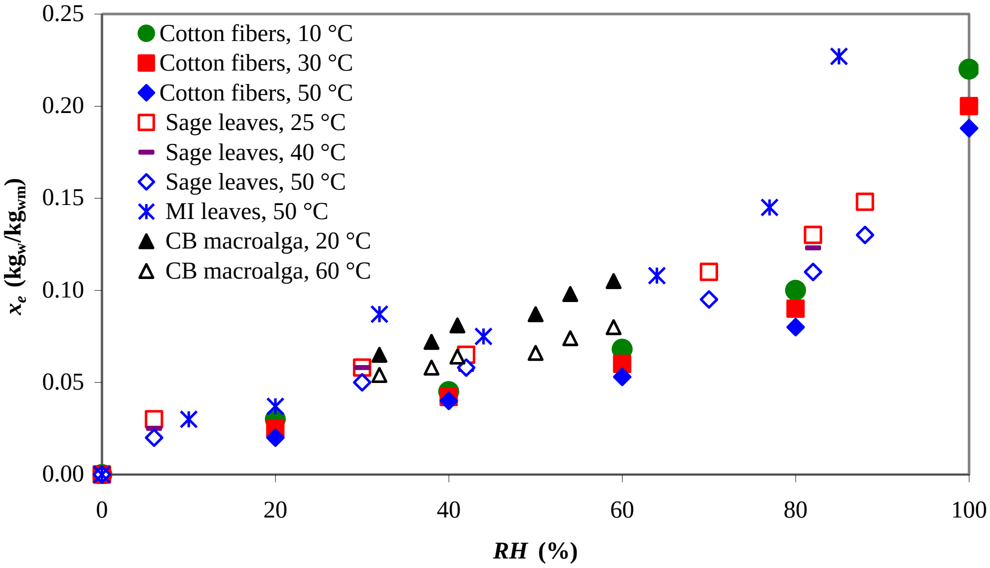

Fig. 6. Equilibrium water mass fraction of dried materials depending on air temperature and relative humidity for different systems: air-cotton fibers [27], air-sage (*Salvia officinalis* L.) leaves [28], air-*Maytenus ilicifolia* (MI) leaves [29], air-*Cystoseira barbata* (CB) macroalga (this study)

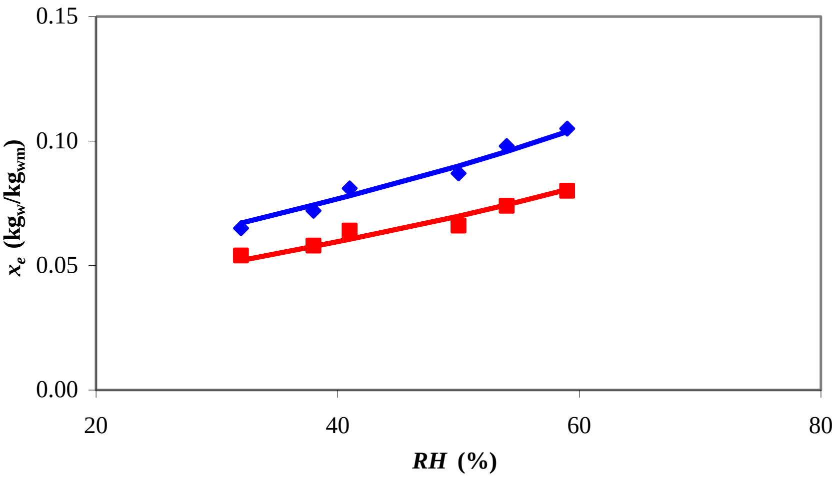

Fig. 7. Equilibrium water mass fraction of dried macroalga *vs* relative humidity at 20 °C (♦) and 60 °C (■) (bullets: experimental, lines: Eq. (10))

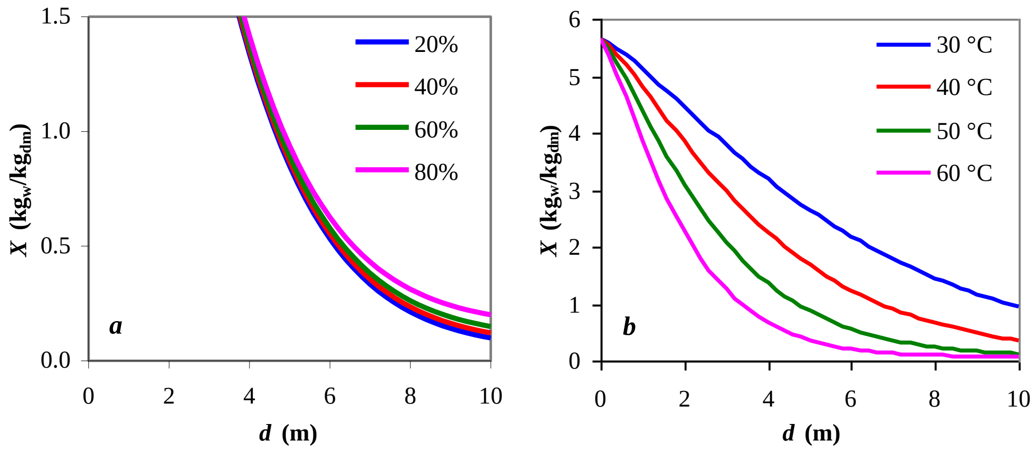

Fig. 8. Predicted variations of water mass ratio of *Cystoseira barbata* macroalga along the length of conveyor belt for different values of (a) relative humidity at 50 °C and (b) air temperature at RH=40% (X_0=5.67 kg$_w$/kg$_{dm}$, w_b=3 m/h, E_a=37393 J/mol, k_0=1.14×10^6 h^{-n}, n=1.257)

5. Conclusions

Thin layer convective drying of *Cystoseira barbata* brown macroalga was performed in a thermobalance and a food dehydrator. Characteristic dynamics of convective drying in the thermobalance at 50-80 °C were predicted using semi-theoretical models based on Fick's second law of diffusion, *i.e.*, Fick II, Page, Newton, and Henderson and Pabis (HP).

Adjustable parameters of kinetic models in terms of effective diffusivity (D_{eff}), rate constant (k), n exponent in Page model, and a pre-exponential factor in HP model were estimated by fitting the experimental data. Values of D_{eff} (0.761-1.876)×10^{-9} m^2/s) and those of k (0.961-3.040 h^{-n} for Page model, 0.997-2.429 h^{-1} for Newton model, and 1.073-2.581 h^{-1} for HP model) were higher at superior levels of drying temperature, whereas n (1.176-1.323) and a (1.052-1.075) were almost invariant. Page model predicted better the experimental dynamics (RMSE=0.018-0.026) than the other models.

Values of activation energy for drying tests performed in the thermobalance, which were determined based on Arrhenius equation, were in the range of 26.58-37.39 kJ/mol. Kinetic model parameters obtained for drying in the thermobalance (at 50-80 °C) were used to predict drying dynamics in the dehydrator (at 55 °C). Page, Newton, and HP models simulated accurately (RMSE=0.024-0.027) experimental drying dynamics in the dehydrator.

A procedure based on Page model was proposed to simulate the operation of a conveyor belt dryer with infrared radiation heating (CBDIR). Moisture content of algal biomass along the length of conveyor belt was predicted for various levels of air temperature and relative humidity. The results obtained in this study could be used to design, optimize, and control the biomass drying in specific equipments.

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