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Enhancing brown seaweed *Sargassum muticum* drying using pulsed electric fields and dehydration pretreatments

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The present study on the brown seaweeds *Sargassum muticum* focused on the application of dehydration by pressing or pulsed electric fields (PEF) pretreatment before three drying processes, namely freeze-drying, air impingement drying and natural convection in an oven. Both pretreatments permitted to reduce significantly the initial moisture of the seaweeds with a reduction up to 50 %. The structural modifications induced by these pretreatments had a direct impact on the mechanisms of moisture retention and distribution within the seaweeds, which consequently exerted an influence on drying kinetics. The apparent moisture diffusivity for freeze-drying was significantly improved by PEF passing from 2.98 10⁻⁹ m² s⁻¹ to 3.62 10⁻⁹ m² s⁻¹ after a total disintegration. Concerning oven drying by natural convection, a dehydration by pressing will be preferred because the drying time to reach 0.45 kg kg⁻¹ in dry basis was reduced by 61 min with this pretreatment.

Keywords: air jets, freeze-drying, moisture diffusion coefficient, pretreatments, *Sargassum muticum*

Introduction

The brown seaweeds *Sargassum muticum* (Yendo) Fensholt (Fucales) are present from Norway to Italy and were probably introduced in Europe in the 1970s with oysters *Crassostrea gigas* imported from Canada and Japan. They are now considered as an invasive specie in Europe because the compete with endemic species, colonized original spaces of several plants and oyster farming areas. Because of their good acclimatization of various environments with specific characteristics like the temperature, the luminosity and the salinity, they were able to proliferate rapidly on the European coasts. Despite several eradication trials, in particular in France and England^[1], they are still progressing along the Atlantic coast. Representing an interesting source of components of interests such as proteins and polysaccharides, this biomass can be valorized in the domains of animal health and feed. Moreover, it represents an important quantity of biomass, which is underexploited. However, because of their high moisture content (about 90 % in wet basis. w.b. (w/w)), a stabilization process is necessary before using *S. muticum* in a biorefinery process. In fact, they degrade rapidly after harvesting.^[2] Moreover, the seasonality of the biomass will be also overcome thanks a drying process, providing a dry and stabilized product all year round.

The main focuses of modern drying processes concern the energy saving and also the quality of the final product. An innovative drying process improves heat and mass transfers thanks to the intensification of the hot air-drying process by impacting air jets on the product's surface. A higher local velocity is thus produced^[3] and temperature levels and drying time are also decreased.^[4,5] This drying is the air impingement drying. This process has been used in previous studies with *S. muticum*^[5] and on holopelagic Sargasso^[6] but no pretreatments were applied before drying.

Pulsed electric field (PEF) treatment is often used as a pretreatment for optimizing processes such as pressing, drying and freezing.^[7] Its main application is the preservation of food products, since PEF are based on eradication of pathogenic microorganisms and control of spoilage of biological products.^[8] This treatment can also be used to insert or extract molecules from cells. For macroalgae, the technology is mostly employed to improve extractions of compounds of interest^[9-13] and the study of this process as a pretreatment for dehydration has also been applied to green algae,^[14] but not on brown seaweeds, as far as we know.

This study outlines pretreatments' parameters, which are dehydration by pressing and pulsed electric fields, on brown seaweeds *S. muticum*. Three drying processes, namely freezedrying, air impingement drying and natural convection in an oven, were also applied to study the impact of the pretreatments on initial moisture content, drying kinetics and effective

moisture diffusivity. The aim of this study was to determine the pretreatment and the associate drying process to stabilize *S. muticum* effectively for use in biorefinery processes.

Material and methods

2.1. Raw material

The seaweed used for the experiments of this study were the brown species *S. muticum*. They were harvested without their holdfast on the Kercambre beach in Saint-Gildas-de-Rhuys (N 47°29'20.81'' O 2°49'14.203'') in February 2022. They were then washed from epiphytes and stored at 4 °C in salt water in the darkness until their use. They were washed with freshwater and cut into samples of 0.1 m long for the experiments.

2.2. Dehydration by pressing

The kinetics for the dehydration by pressing were carried out in triplicate with an initial mass of $31.02 \pm 0.01 \ 10^{-3}$ kg of fresh seaweed, previously rinsed and cut into 0.1 m sections. The algae were placed in a drip tray covered with a rubber membrane progressively filled with water until reaching a maximum pressure of 3.2 bar. The extracted juice was recovered in a crystallizer and its mass was read every 10 s on a balance (CP2202S-OCE, accuracy 10^{-2} , Sartorius, Göttingen, Germany). After 5 min of pressing, the algae were stirred and then repressed under the same conditions for a further 5 min. The final mass of the algae was then recorded. Manipulations to obtain pressing kinetics were carried out in triplicate. The pressed algae could then be used directly to study their stabilization by various drying processes.

2.3. Pulsed electric fields

Electroporation monitoring studies using pulsed electric fields were carried out on *S. muticum* at field strengths between 200 V cm⁻¹ and 3 000 V cm⁻¹ (voltage from 1 200 V to 18 000 V)

with an interval of 400 V cm⁻¹ produced with two high-voltage generators (MU-17-807-A-FR 22.5 kV, ITHPP, Thégra, France and AHTPM2.5, Effitech, Pau, France). Pulse duration was 2 μ s, with a train duration of 3 s. The rest time between each pulse train was set at 2 min, and the frequency was 1 Hz. The treatment chamber was cylindrical, measuring 6 cm in height and 3.3 cm in diameter.

To ensure a proper treatment, the electrodes were checked and rubbed with abrasive paper to obtain a smooth and clean surface. The initial mass of algae placed in the PEF chamber is $31.03 \pm 0.03 \ 10^{-3}$ kg. The initial temperature of the sample and its initial conductivity (through its impedance) were recorded with an impedance meter (U1733C, Keysight Technology, Les Ulis, France). When all treatments have been applied and the final intensity measured, the sample temperature is recorded. The algae can then be directly stabilized by a drying process.

The cellular matrix decay index Z (-) was calculated as follows:^[15]

$$Z = \frac{\sigma - \sigma_i}{\sigma_d - \sigma_i} \tag{1}$$

with σ (μ S cm⁻¹) corresponding to the conductivity at time t, σ_i (μ S cm⁻¹) is the initial conductivity of the algae and σ_d (μ S cm⁻¹) corresponds to the conductivity of a sample with all cells damaged. Here, this value is obtained by carrying out three freeze/thaw cycles (9 397 ± 101 μ S cm⁻¹).

The received mass energy was calculated with the following equation:

$$E_{PEF} = \sum_{0}^{n} \frac{U \times I \times t_{PEF}}{m}$$
(2)

with E_{PEF} (kW kg⁻¹) corresponds to the energy injected, U (V) is the applied voltage, m (kg) is the mass of the product treated, I (A) corresponds to the current intensity and t_{PEF} (s) represents the treatment time PEF applied. The curves of received mass energy and decay index were modeled according to the following equation:

$$y = a \times \exp(-b \times x) + c \tag{3}$$

with y corresponds to either the received mass energy $(J \text{ kg}^{-1})$ or the decay index (-), x (s) cumulative PEF time and a, b and c are the coefficients of the curve.

2.4. Drying processes

Drying processes were applied after pretreatments in the same drying conditions as untreated control samples. For each drying experiment, either PEF, or dehydration by pressing was used as pretreatment. Seaweeds were dried directly after pretreatments.

Air impingement drying process

The prototype of air impingement dryer used in this study was designed by CIMS company (Sablé-sur-Sarthe. France) and is visible on Figure 1.^[4-6] The impingement jets are shaped by two plenum chambers positioned inside the drying chamber, on which a rig was placed. The air jets configuration consisted of triangular nozzles spaced at 5 10⁻² m with rectangular slots (average width of 3 10⁻³ m).

The experiments were performed at 40 °C at two air velocities based on a previous study^[5] (7 m s⁻¹ and 21 m s⁻¹). Four samples were loaded simultaneously in the drying chamber 30 min after the start of the dryer and with one minute delay. They were weighed at specific time using a precision balance (Acculab ALC-110.4. accuracy 10^{-7} kg. Sartorius. Göttingen. Germany). First, they were weighted after one minute left inside the dryer and the mass was then measured each ten minutes until the final moisture content of 0.11 kg kg⁻¹ (10 %_{wb}) was reached.



Figure 1. Drawing of the air impingement dryer used for the experiments

Freeze-drying

For each point of the kinetics, three replicates of $11.6 \pm 0.1 \ 10^{-3}$ kg were placed inside the freeze-dryer (Virtis Advantage XL-70, SP Scientific, USA). First, they were frozen at -45 °C during 5 h. Then the next step was the primary drying performed at -30 °C at 13.3 kPa over 13 h. Finally, the secondary drying was carried out at 50 °C for 5 h at 6.7 kPa. This temperature of 50 °C was chosen because it corresponds to a measured temperature in the product around 40 °C. At the allotted time and after atmospheric pressure had been applied, the samples were then weighed. These described parameters corresponded to a complete cycle of freeze-drying.

Oven-drying

The oven drying was performed at 40 °C in an oven (Type B 50, Memmert, Schwabach, Germany) with flap opened (natural convection). Four replicates were placed in a dish $(1 \times 10^{-1} \text{ m diameter})$ The initial mass of a sample was $5.8 \pm 0.1 \ 10^{-3}$ kg. They were weighed each 10 min with a precision balance (Acculab ALC-110.4, accuracy 10^{-7} kg, Sartorius,

Germany) the two first hours of the drying and then each 30 min until the end of the process. The drying was stopped when a final moisture content of 10 $\%_{wb}$ was reached.

2.5. Variables characteristics of the drying process

The moisture content (kg kg⁻¹d.b.) was calculated thanks the measured mass of the samples during the drying. The equation is the following:

$$X_{db} = \frac{m_t - m_{dm}}{m_{dm}}$$
(4)

where m_t (kg) is the mass of the sample recorded at time t and m_{dm} (kg) is the mass of the sample after drying in oven at 105 ± 2 °C until stabilization.

The moisture content (% w.b.) was calculated as follows:

$$X_{wb} = \frac{m_{water}}{m_t} \times 100 = \frac{m_t - m_{dm}}{m_t} \times 100$$
(5)

where m_{water} (kg) is the mass of water contained in the sample at time t.

2.6. Calculation of the effective moisture diffusivity Deff

During the drying of biological products, the moisture diffusion controls the drying time because it is the limiting transport mechanism. The effective moisture diffusivity D_{eff} (m² s⁻¹) of brown algae *S. muticum* was determined based on Fick's second law defined with the following equation:^[16]

$$\frac{dX_{db}}{dt} = D_{eff} \cdot \frac{d^2 X_{db}}{dx^2}$$
(6)

where t (s) is the time and x (m) the characteristic dimension of the product.

The geometry of the product (infinite slab, infinite cylinder and sphere) determines the corresponding analytical solution, which was defined by Crank.^[16] Some assumptions are

made for the calculation, such as the neglect of external resistance and shrinkage, the effective moisture diffusivity is also considered constant during the drying. These assumptions were detailed by Boy et al.^[4]

In this study, the geometry considered for the algae was infinite slab and the corresponding equation was the following:^[16]

$$\omega = \frac{X_{t} - X_{eq}}{X_{0} - X_{eq}} = \frac{8}{\pi^{2}} \sum_{i=0}^{\infty} \frac{1}{(2i+1)^{2}} \exp\left(\frac{-(2i+1)^{2} \cdot \pi^{2} \cdot D_{eff} \cdot t}{4L^{2}}\right)$$
(7)

where X_t (kg kg⁻¹ d.b.) is the moisture content at time t, X_0 (kg kg⁻¹ d.b.) is the initial moisture content, i (-) is the number of iterations and L (m) is the half thickness (2 10⁻³ m).

In this work, the calculated coefficient was depending on the air velocity, that is why the name of apparent moisture diffusivity D_{app} was preferred for the coefficient instead of D_{eff} .^[5] In fact, the diffusivity calculated by the Fick's diffusional model visible on the equation 7 depends only on the internal material properties, while both external drying conditions and internal material properties have an impact on the coefficient of the present study.

2.7. Statistical parameters

The goodness of D_{app} calculation was estimated by four statistical parameters with the following equations:^[5,17]

adjusted R² = 1 -
$$\frac{\sum_{j=1}^{n} (\omega_{\exp,j} - \omega_{mod,j})^{2} \cdot (N-1)}{\sum_{j=1}^{n} (\omega_{\exp,j} - \overline{\omega}_{\exp,j})^{2} \cdot (N-p)}$$
(8)

$$RMSE = \sqrt{\frac{1}{N} \sum_{j=1}^{N} (\omega_{exp,j} - \omega_{mod,j})^2}$$
(9)

$$\chi^2 = \frac{\sum_{j=1}^{N} (\omega_{\exp,j} - \omega_{\text{mod},j})^2}{N - p}$$
(10)

$$RSS = \sum_{j=1}^{N} (\omega_{exp,j} - \omega_{mod,j})^2$$
(11)

where $\omega_{exp,j}$ and $\omega_{mod,j}$ correspond to the experimental and modeled moisture ratios, $\overline{\omega}_{exp,j}$ is the average of the experimental data, N is the number of observations and p is the number of parameters in the equation. High values of adjusted R² combined with low values of RMSE, χ^2 and RSS indicate a satisfying quality of the fitting.

The results were analyzed with ANOVA with a test of Tukey (p-value < 0.05) in RStudio (Integrated Development Environment for R. RStudio, PBC, Boston, USA).

Results and discussion

3.1. Dehydration by pressing

The kinetics of dehydration by pressing algae at 3.2 bar is presented on Figure 2. This pretreatment extracted up to $10 \ 10^{-3}$ kg of juice from the seaweed, corresponding to a third of the initial seaweed mass, passing from an initial moisture content of 10.23 kg kg⁻¹ to 5.11 kg kg⁻¹. It can also be seen that half the juice is already extracted when pressing begins. Pressing therefore considerably reduces the moisture content of the fresh seaweeds.



Figure 2. Kinetics of dehydration pretreatment on Sargassum muticum at 3.2 bar

Ten minutes of pressing were chosen for the application of this process as a pretreatment for thermal drying. This pretreatment will also be carried out with a time of 5 min before oven drying and air jets.

3.2. Pulsed electric fields kinetics

Preliminary studies on the application of pulsed electric field technology on seaweeds have demonstrated an improvement of the extraction of phlorotannins and proteins.^[9,18] Electroporation of the algal cell membrane therefore improves the accessibility of compounds present in the cells.

The Figure 3 presents the disintegration index of *S. muticum* calculated during PEF treatments for different electric field intensities. The disintegration of the product increases with high applied field strength. At 200 V cm⁻¹, for example, the final decay index is 0.09 ± 0.02 compared with 1.04 ± 0.06 at $3\ 000$ V cm⁻¹. A critical value is reached for each intensity during the pretreatment.



Figure 3. A) Pulsed electric fields kinetics of *Sargassum muticum*, B) Mass energy received during pulsed electric fields pretreatment

These disintegration curves are used to determine the PEF treatment parameters to be applied, notably the electric field intensity and treatment time, in order to achieve the desired level of disintegration (Z = 0 intact cells; Z = 0.5 when 50 % of cells are damaged; Z = 1 all damaged cells).

During PEF treatment, the product is subjected to electrical pulses, and thus receives energy, this enables electroporation of the cell membranes. The Figure 3 illustrates the amount of mass energy received as a function of the cumulative PEF time to which the product was submitted.

It can be seen that the higher the electric field intensity, the greater the mass energy received. For example, at 200 V cm⁻¹, the mass energy received at the end of treatment is 1.5 ± 0.08 J kg⁻¹, compared with 864.9 ± 35.4 J kg⁻¹at 3 000 V cm⁻¹. A plateau is reached for each intensity, in line with the trays reached during electroporation decay. It can therefore be concluded that the greater the mass energy received by the product, the more likely it is to disintegrate. This result is consistent with recent research on pulsed electric fields disintegration of yellow turnips, for which the disintegration index Z increased with the intensity of PEF treatment.^[19]

The shape of the coefficients obtained by applying equation 3 for mass energy received and disintegration index as a function of cumulative PEF time is presented on Figure 4.



Figure 4. Modeling parameters of A) the mass energy received and B) the disintegration index

The coefficients derived from modelling are consistent with increasing field strength, for example, the coefficient "b" presents a slight decrease at the last electric field strength and is also greater at the first received mass energy strength.

Disintegration also causes the product's temperature to increase as observed by Iranshahi et al.^[20] The temperature of the algae was measured before the first pulse train and at the very end of the treatment. The temperature difference for each applied intensity is visible on Figure 5.



Figure 5. Temperature increases causing by pulsed electric fields treatment

The temperature increases only slightly for intensities up to 1 400 V cm⁻¹. For higher intensities, however, the product heats up by more than two degrees, which is less than the significant twenty degrees observed for the pretreatment at high intensity of apple.^[20] The two-minute pause between each train limits this temperature rise. Two intensities have been chosen for the combination of pulsed electric field pretreatment and drying: 1 800 V cm⁻¹ for moderate algae disintegration (Z = 0.5) and 3 000 V cm⁻¹ for total disintegration (Z = 1). The number of trains will be adapted according to the desired disintegration indices.

3.3 Drying kinetics

Drying processes using freeze-drying, air jets impacting at two air velocities (7 m s⁻¹ and 21 m s⁻¹) and a temperature of 40 °C, and natural convection in an oven at 40 °C, were optimized by the application of pretreatments. For each process, a control sample is presented in addition to the samples pretreated either by pulsed electric fields or by pressing.

During drying by natural convection in an oven at 40 °C, both pretreatments, i.e., dehydration by pressing and application of two degrees of cell disintegration by PEF, were studied. On Figure 6, the initial moisture content of the seaweeds is higher for non-pretreated algae (10.23 kg kg⁻¹), then for PEF-treated algae (8.44 kg kg⁻¹ for Z = 0.5 and 8.11 kg kg⁻¹ for Z = 1), and finally for algae previously dehydrated by pressing (6.23 kg kg⁻¹ for 5 min pressing and 5.11 kg kg⁻¹ for 10 min pressing with stirring). Dewatering by pressing therefore reduces the initial moisture content of the algae by 39 % to 50 %. For PEF, water loss from fresh algae is in the order of 17 % to 20 %, depending on the level of disintegration. This reduction in seaweed water content means shorter drying times. As a result, to reach 0.45 kg kg⁻¹ moisture in dry base (30 % in wet basis), the shortest time corresponds to seaweed pressed 10 min with a time of 225 min. It then takes 229 min to reach the same moisture content for algae pressed 5 min, then 262 min and 282 min for algae disintegrated by PEF at Z = 0.5 and Z = 1 respectively. This time is close to the 286 min needed to reach this moisture content with untreated algae.



Figure 6. Moisture content in dry basis of *Sargassum muticum* oven dried (triangle) and pretreated by pressing (square) or by pulsed electric fields (round)

It turns out that pretreatments only reduce drying times up to a certain point, since from 330 min onwards all curves are confounded and the effect of pretreatments on moisture is no longer visible. The benefit of pretreatment is therefore useful for reducing algae moisture up to 0.15 kg kg⁻¹. In the literature, the application of electric fields as a pretreatment to drying is widespread, particularly on vegetables and fruit. This pretreatment has been shown to significantly reduce drying times.^[20-24] For example, drying times are reduced by 6.9 % to 8.2 % for 70 °C drying of carrots with PEF treatments of 5.63 kJ kg⁻¹ and 8 kJ kg⁻¹,^[25] and can be reduced by up to 34.7 % under optimal conditions for PEF pretreatment of *Capsicum annuum*.^[26] In general, the PEF conditions with the best results on drying kinetics correspond to total disintegration $(Z = 1)^{[25-27]}$ or at least with high disintegration (Z > 0.8).^[24] However, when drying onions following PEF treatments,^[28] concluded that kinetics was further improved for medium disintegration (Z = 0.53). This observation is in line with our results, since drying times are shorter for Z = 0.5 than for Z = 1. The hypothesis associated with these results is related to the structure of the algae, since the softening of the seaweeds caused by the PEF would result in greater collapse of the intracellular structure, hindering mass transport and thus reducing drying speed. Indeed, the pretreatments of dehydration by pressing and by PEF modified the cellular structure of the algae. Pressing compressed the cells, while PEF created pores in the cell membranes. These structural changes directly affect the way moisture was retained and distributed in the algal tissue, and thus influenced drying kinetics. Drying time was reduced by improving water diffusion during drying, thanks to the disruption and electroporation of cell membranes following pretreatments. Kim et al.^[24] also concluded that PEF pretreatment of carrots reduced drying time through improved moisture diffusivity.

For air jets drying process, dehydration pretreatment by pressing was applied to the fresh *S. muticum* material. The pretreatment reduces the initial moisture content of the algae prior to the thermal drying process (Figure 7). Indeed, the initial moisture content of fresh seaweed is 9.30 ± 0.67 kg kg⁻¹ on average against 6.28 ± 0.27 kg kg⁻¹ after 5 min pressing and 5.45 ± 0.21 kg kg⁻¹ after 10 min pressing.



Figure 7. Moisture content in dry basis of *Sargassum muticum* dried by air jets impacting (triangle) and pretreated by pressing (square)

A reduction in moisture of between 31 % and 34 % is thus observed for seaweed pressed for 5 min, and a loss of moisture in the order of 46 % to 37 % for 10 min pressing with stirring after 5 min pressing. For the two drying velocities studied, pressing reduces drying times: for example, to reach 30 % moisture content in wet basis (0.45 kg kg⁻¹) at 7 m s⁻¹, it takes 20 min, 19 min and 16 min respectively for untreated, 5 min pressed and 10 min pressed seaweeds. Drying times improved by an average of 4 min at 21 m s⁻¹. From 21 min, however, the curves merge for all algae and all velocities combined. This drying time reduction was due to a lower initial moisture content in seaweed and a higher apparent moisture diffusivity.^[29]

As the curves meet after 21 min of drying, an enlargement was carried out in order to observe the differences between the kinetics (Figure 7). It can thus be seen that the dry-base humidities of untreated algae are higher than those of previously dehydrated algae, and that pressing time also influences drying kinetics. Indeed, for a pressing time of 10 min, the dry-base moisture content of the algae is lower than that of algae pressed for 5 min to 21 min, for both drying speeds. In addition, the curve for algae dried at 21 m s⁻¹ is lower than that for

seaweeds dried at 7 m s⁻¹, demonstrating the influence of air velocity on drying kinetics as shown in Le Loeuff et al.^[5] This finding is reflected in the apparent diffusion coefficients (results presented later in this section).

Freeze-drying was carried out with primary drying at -30 °C for untreated seaweeds and seaweeds previously disintegrated by pulsed electric fields at two disintegration levels (medium: Z = 0.5; and total: Z = 1). Figure 8 presents the results obtained for dry-base moisture kinetics.



Figure 8. Moisture content in dry basis of *Sargassum muticum* freeze-dried (triangle) and pretreated by pulsed electric fields (round) at different disintegration ratio

At the end of the complete freeze-drying cycle (in total 17 h and 50 min), the average final moisture content of the algae was 0.078 ± 0.016 kg kg⁻¹ (7.19 ± 1.36 %). On the other hand, the effect of pulsed electric fields on moisture content can be observed right from the start of the kinetics. The initial moisture content on a dry basis for untreated algae (Z = 0), moderately disintegrated algae (Z = 0.5) and totally disintegrated algae (Z = 1) were 8.53 ± 0.20 kg kg⁻¹, 5.51 ± 0.5 kg kg⁻¹ and 4.64 ± 0.37 kg kg⁻¹ respectively. Seaweeds therefore lose water with this pretreatment. On the other hand, pretreatment has only an influence on freeze-drying kinetics only to 180 min. However, the final moisture content is

lower for the control sample at Z = 0. PEF pretreatment will therefore only be beneficial, in terms of improving drying times, for times in excess of 180 min and therefore humidities below 47 %_{wb}.

Moreover, the curves for the two PEF treatments merge after 300 min of freezedrying, so total disintegration requiring high-intensity pulses will not be favorable compared with moderate disintegration. Indeed, it does not seem worthwhile to push disintegration to its maximum in view of the kinetic results, which are relatively close for the two pretreatments applied. In contrast, Wu and Zhang^[30] demonstrated that potato freeze-drying was further enhanced with the PEF pretreatment at extreme conditions (1 500 V cm⁻¹, 120 μ s and 45 pulses). These conditions resulted in a 31.47 % reduction in freeze-drying time. Freezedrying kinetics performed at 35 Pa and 75 °C on apples were also improved with pulsed electric fields.^[31]

Air velocity in jet drying has an impact on drying kinetics and apparent diffusion coefficients. D_{app} coefficients were calculated between 0 min and 21 min for impact air jet drying, and between 0 min and 180 min for natural convection oven drying, both with n = 0. It turns out that for the control sample, D_{app} is higher for an air velocity of 21 m s⁻¹, with a value of $6.89 \pm 2.59 \ 10^{-9} \ m^2 \ s^{-1}$, compared with $4.53 \pm 0.23 \ 10^{-9} \ m^2 \ s^{-1}$ for a velocity of 7 m s⁻¹. This observation is also valid for the biomass pressed 5 min and 10 min, since for both pretreatments, the D_{app} values are higher than values for the highest air velocity. Increasing drying air velocity therefore improves water diffusivity through the product and accelerates drying kinetics. Coefficient values are shown in Table 1.

Apparent diffusion coefficients for drying by natural convection in the oven at 40 °C are significantly lower than those calculated for drying with air jets, which is totally consistent with the drying kinetics observed. For example, for algae pressed 10 min, D_{app} is $0.228 \pm 0.020 \ 10^{-9} \ m^2 \ s^{-1}$ in the oven, compared with 4.50 ± 0.11 $10^{-9} \ m^2 \ s^{-1}$ at 7 m s⁻¹ and

 $5.70 \pm 1.32 \ 10^{-9} \ m^2 \ s^{-1}$ at 21 m s⁻¹. Air jet drying is therefore significantly faster than natural convection drying for the same temperature of 40 °C. Moreover, for the 5 min pressing applied as a pretreatment, D_{app} is significantly greater than for the control sample, demonstrating that diffusion is accelerated with this pretreatment. To optimize this drying process, and for the dehydration pretreatment by pressing, only 5 min of pressing will be retained. In addition, this pretreatment optimizes diffusion, unlike the pulsed electric field pretreatment. Indeed, the coefficients are $0.193 \pm 0.010 \ 10^{-9} \ m^2 \ s^{-1}$ for average disintegration and $0.165 \pm 0.011 \ 10^{-9} \ m^2 \ s^{-1}$ for total disintegration, compared with $0.194 \pm 0.016 \ 10^{-9} \ m^2 \ s^{-1}$ for untreated seaweeds.

without pretreatn	nents. Dif	fere	nt letters i	n the same	e lin	e indicate s	ignificant sta	tisti	cal differe	ences.						
	Air impacting jets at 7 m s⁻¹															
	Control			Pressing = 5 min			Press	Pressing = 10 min								
$D_{app} \times 10^9 \ [m^2 \ s^{-1}]$	4.53ª	±	0.23	4.41 ^a	±	0.26	4.50ª	±	0.11							
R ²	0.9989	±	0.0007	0.9993	±	0.0006	0.9925	±	0.0020							
RMSE × 10 ²	4.55	±	1.17	3.42	±	1.2	12.2	±	1.53							
$\chi^2 \times 10^2$	6.44	±	1.66	4.84	±	1.76	17.3	±	2.17							
RSS × 10 ²	0.87	±	0.48	0.51	±	0.39	6.03	±	1.49							
	Air impacting jets at 21 m s ⁻¹															
	C	ol	Pressing = 5 min			Press	Pressing = 10 min									
$D_{app} \times 10^9 \ [m^2 \ s^{-1}]$	6.89 ^a	±	2.59	4.55ª	±	0.35	5.70ª	±	1.32							
R ²	0.9948	±	0.0013	0.9881	±	0.0004	0.9810	±	0.0048							
RMSE × 10 ²	12.9	±	1.46	18.2	±	0.31	23.4	±	2.95							
$\chi^2 \times 10^2$	18.2	±	2.06	25.7	±	0.44	33.1	±	4.17							
RSS × 10 ²	6.71	±	1.50	13.2	±	0.45	22.2	±	5.30							
			Nati	ural convect	ion c	drying at 40 °	С									
	C	ol	PE	PEF Z = 0.5			PEF Z = 1			Pressing = 5 min			Pressin	Pressing = 10 min		
$D_{app} \times 10^9 \ [m^2 \ s^{-1}]$	0.194 ^{bc}	±	0.0160	0.193 ^{bc}	±	0.010	0.165 ^c	±	0.011	0.	247ª	±	0.018	0.228 ^{ab}	±	0.020
R ²	0.9929	±	0.0008	0.9942	±	0.0026	0.9928	±	0.0018	0.	9968	±	0.0010	0.9948	±	0.0022
RMSE × 10 ²	3.05	±	0.25	2.31	±	0.63	2.21	±	0.35	2	2.19	±	0.25	2.61	±	0.76
$\chi^2 \times 10^2$	3.27	±	0.27	2.49	±	0.68	2.39	±	0.38	2	2.37	±	0.27	2.82	±	0.86
RSS × 10 ²	1.40	±	0.23	0.78	±	0.38	0.70	±	0.22	C).68	±	0.16	1.01	±	0.54
				Fr	eeze	-drying										
	C	ontro	ol	PEF Z = 0.5			PEF	PEF Z = 1								
$D_{app} \times 10^9 [m^2 s^{-1}]$	2.98ª	±	0.049	3.51 ^b	±	0.17	3.62 ^b	±	0.29							
R ²	0.9892	±	0.0043	0.9760	±	0.0028	0.9732	±	0.0079							
RMSE × 10 ²	18.3	±	3.51	32.7	±	3.08	35.8	±	8.34							
$\chi^2 \times 10^2$	25.9	±	4.96	46.2	±	4.36	50.6	±	11.8							
RSS × 10 ²	17.7	±	5.33	43	±	7.86	53	±	25.4							

Table 1. Coefficient values and statistical parameters for air impacting jets at 7 m s⁻¹ and 21 m s⁻¹. Oven drying and freeze-drying with and without pretreatments. Different letters in the same line indicate significant statistical differences.

PEF pretreatment do not improve diffusion coefficients for oven-drying of *S. muticum* at 40 °C. Better internal diffusion was also observed for medium rather than total decay.^[28] In fact, drying is not necessarily improved by a greater release of water from the product surface caused by PEF, since water evaporation also influences the drying rate. Drying conditions must therefore be consistent with PEF induced tissue disintegration in order to derive benefits for the drying process, including adaptation of the evaporation rate.^[28] However, these results are not consistent with numerous studies that have shown that the more consistent the pretreatment by PEF, the greater the improvement in the effective diffusion coefficient.^[21-23, 25-27] Wiktor and Witrowa-Rajchert^[32] also observed an increase of around 42 % in D_{eff} for microwave assisted convection dried carrots treated with PEF.

In contrast, PEF improves diffusion during freeze-drying of *S. muticum*, as it does for apples.^[31] Indeed, the significant difference between the D_{app} of untreated and PEF-treated seaweeds suggests that PEF treatment has an impact on diffusion. However, since the difference is not significant between the two treatments applied, then it does not seem relevant to apply total disintegration of algal material. These results also highlight the fact that freeze-drying provides better diffusion $2.98 \pm 0.049 \ 10^{-9} \ m^2 \ s^{-1}$ than oven drying $0.194 \pm 0.016 \ 10^{-9} \ m^2 \ s^{-1}$, but air-jet drying still remains the process with the best diffusion $6.89 \pm 2.59 \ 10^{-9} \ m^2 \ s^{-1}$ at $21 \ m \ s^{-1} \ vs$. $4.53 \pm 0.23 \ 10^{-9} \ m^2 \ s^{-1}$ at $7 \ m \ s^{-1}$. These results are still lower than the diffusion coefficients obtained during apple freeze-drying, since they are $2.4 \ 10^{-8} \ m^2 \ s^{-1}$ for untreated apples and reach $4.20 \ 10^{-8} \ m^2 \ s^{-1}$ for apples pretreated with PEF.^[31]

With regard to drying with air impacting jets, no significant difference between D_{app} was observed for the same air velocity.

Calculation of the diffusion coefficients was used to model the drying curves using Crank's equation. Figure 9 presents the reduced moisture content models obtained for impact air jet drying (A), oven drying (B) and freeze-drying (C). It can be seen that there is a convergence of reduced humidities. The linearization of these curves within the time interval defined for the D_{app} calculation can also be seen on the corresponding figures.



Figure 9. Modelling of the drying curves with Crank's equation and the calculated D_{app} for air jet drying (A), oven drying (B) and freeze-drying (C). Control sample are represented by a triangle, sample pretreated with PEF or dehydration by pressing are represented by a round and the parameters corresponding to the pretreatment applied.

It turns out that for all three drying processes, Crank's^[16] model of the drying curves is satisfactory, but only within the predefined reduced moisture content range. It would therefore

be incorrect to use this model for times in excess of 180 min for convective oven drying, in excess of 21 min for impact air jet drying and in excess of 700 min for freeze-drying.

The apparent diffusion coefficients were calculated over a precise time interval corresponding to the point linearity of the natural logarithm of reduced moisture content. These models, together with the apparent diffusion coefficient, provide a visual description of the phenomena occurring during drying, even if this representation is only a partial view of reality.

Conclusion

Dewatering by pressing and disintegration of the algal material by pulsed electric fields can be applied as pretreatment to drying processes. Pressing kinetics were used to define the times to be applied, a pressing time of 10 min will be preferred because of the reduction up to 50 % of the initial moisture content of the algae. The PEF study quantified the cellular disintegration of seaweeds for different treatment parameters applied and the oven drying kinetics were improved for medium disintegration (Z = 0.5). This pretreatment also significantly improved the apparent moisture diffusivity for freeze-drying.

To conclude, these pretreatments reduced the initial moisture content of the algae and thus shorten drying times to reach 30 % final moisture, and improved apparent diffusion coefficients. The impact of drying, due to exposure of the product to heat, was thus reduced thanks to these pretreatments.

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Declaration of interest statement

The authors declare that they have no conflict of interest.

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Table caption

Table 1. Coefficient values and statistical parameters for air impacting jets at 7 m s⁻¹ and 21 m s⁻¹. Oven drying and freeze-drying with and without pretreatments. Different letters in the same line indicate significant statistical differences.

Figure captions

Figure 1. Drawing of the air impingement dryer used for the experiments

Figure 2. Kinetics of dehydration pretreatment on Sargassum muticum at 3.2 bar

Figure 3. A) Pulsed electric fields kinetics of *Sargassum muticum*, B) Mass energy received during pulsed electric fields pretreatment

Figure 4. Modeling parameters of A) the mass energy received and B) the disintegration index Figure 5. Temperature increases causing by pulsed electric fields treatment Figure 6. Moisture content in dry basis of *Sargassum muticum* oven dried (triangle) and pretreated by pressing (square) or by pulsed electric fields (round)

Figure 7. Moisture content in dry basis of *Sargassum muticum* dried by air jets impacting (triangle) and pretreated by pressing (square)

Figure 8. Moisture content in dry basis of *Sargassum muticum* freeze-dried (triangle) and pretreated by pulsed electric fields (round) at different disintegration ratio

Figure 9. Modelling of the drying curves with Crank's equation and the calculated Dapp for air jet drying (A), oven drying (B) and freeze-drying (C). Control sample are represented by a triangle, sample pretreated with PEF or dehydration by pressing are represented by a round and the parameters corresponding to the pretreatment applied.