Industrial Crops & Products 132 (2019) 12–20

Contents lists available at ScienceDirect

Industrial Crops & Products

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

Mathematical modelling of the drying kinetics of *Jatropha curcas* L. seeds

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ARTICLE INFO

Keywords:
Avhad and Marchetti model
Drying kinetics
Jatropha (*Jatropha curcas* L.)
Mathematical modelling
Moisture ratio

ABSTRACT

Jatropha (*Jatropha curcas* L) seed contains non-edible oil, which is suitable for biodiesel production. The present research focused on the mathematical modelling of the drying kinetics of Jatropha seeds at a storage moisture content. The non-pretreated seeds (whole seeds) and pretreated seeds (crushed seeds) were dried at five different air temperatures (313, 323, 333, 343 and 353 K) in a standard heating furnace. The moisture loss from the seeds was systematically recorded, converted to moisture ratio, and fitted to four semi-theoretical drying mathematical models: Lewis, Henderson and Pabis, Page and Avhad and Marchetti models. The fitness of the models were compared using the coefficient of determination (R^2), chi-square test (X^2), root mean square error (R_{MSE}), mean bias error (MBE), and mean absolute error (MAE). It was found that an increase in the air temperature caused a reduction in the drying time of both the whole and crushed seeds. From the tested models, the Avhad and Marchetti model showed the best fitting to the experimental data with R^2 varied from 0.9914 to 0.9969 and 0.9908 to 0.9917 for all tested temperatures for the whole seeds and crushed sees of Jatropha, respectively. The Avhad and Marchetti mode showed superior fit to the experimental data at the drying temperature of 313 K with R^2 of 0.9969 for the whole seed, and at 333 K in case of crushed seeds for which the R^2 value was 0.9917. The activation energy values of 33.53 and 32.885 KJ mol^{-1}were obtained for the whole and crushed seeds, respectively when the best-fitted model was used.

1. Introduction

Biodiesel has been used as an alternative fuel to fossil engines. This importance of biodiesel has increased as a result of the depletion of world petroleum reserves, increased demand for fuels, and the negative environmental impacts of exhaust gases from fossil fuels (Kamel et al., 2018; Singh and Singh, 2010). Jatropha (*Jatropha curcas* L.) is one of the plants with promising potential for the production of biodiesel (Salehi Jouzani et al., 2018; Siqueira et al., 2012), and the production of biodiesel from the seed of this plant has been promoted due to its social, economic and environmental positive effects compared to the fossil fuels (Eckart and Henshaw, 2012; Pandey et al., 2012; Zahan and Kano, 2018). Moreover, investigations on the selection of the most promising accessions of Jatropha plants to get better oil yield, and oil with higher quality for biodiesel production are continued (Alburquerque et al., 2017; Kumar and Das, 2018).

Sustainable production oil crops and biodiesel without affecting food security is highly desirable towards meeting the increasing global energy demands (Mazumdar et al., 2018). Due to the presence of major toxic compound (the phorbol esters) in Jatropha seed oil (Amkul et al., 2017; Becker and Makkar, 2008; He et al., 2017), production of biodiesel from the seeds does not compete with human consumption (Becker and Makkar, 2008). Using non-edible biodiesels feedstocks such as Jatropha could be a good alternative to overcome the problems that could occur due to continuous conversion of edible oils to biodiesel (Atabani et al., 2012; Sajjadi et al., 2016). Jatropha can well adapt to dry and marginal lands with low soil fertility, and thus, it does not compete for arable lands (Atabani et al., 2013; Basili and Fontini, 2012).

Renewable biodiesels that can be produced within the petroleum importing countries will enable the countries to be less dependent upon the imported fossil oil. Biodiesel production also creates employment opportunity to the rural people through cultivation of oil producing plants, and this could contribute to the improvement of the domestic economy (Datta and Mandal, 2014). The seedcake produced as the by-product of oil extraction can be changed to organic fertilizer through

Abbreviations: a, model coefficient; CS, crushed seeds; FFA, free fatty acids; k, drying constants (min^{-1}); MAE, mean absolute error; MBE, mean bias error; M_e, equilibrium moisture content; M_0, initial moisture content; MR, moisture ratio; M_t, moisture content at time t; N, model coefficient; PSCS, particle sizes of the crushed seeds; R^2, coefficient of determination; R_{MSE}, root mean square error; t, drying time (min); WS, whole seeds; wt. %, weight percentage; X^2, chi-square test

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https://doi.org/10.1016/j.indcrop.2019.02.012
Received 16 July 2018; Received in revised form 4 February 2019; Accepted 6 February 2019
Available online 11 February 2019

composting and serve as an alternative to mineral fertilizer (Olowoake et al., 2018). Composted organic fertilizer is ecofriendly, and very important and useful input for the enhancement soil health, and it could reduce the N_2O emissions caused by nitrogen containing chemical fertilizers (Basili and Fontini, 2012). Moreover, biodiesel could be used as a fuel alternative in diesel engines to improve combustion efficiency and decrease emissions of the major air pollutants (Teixeira et al., 2012).

Generally, the benefits of using biodiesel include its renewability and biodegradability, low sulfur content and having natural lubricity (Sajjadi et al., 2016), non-toxicity, domestic origin and contributions to the reduction of most pollutant emissions (Firoz, 2017; Sajjadi et al., 2016).

Jatropha seeds can be harvested at different fruit maturity stages, and proper postharvest processing is required as the postharvest management of the seeds greatly affects the quality the product. One of the important factors that one has to give due consideration during postharvest processing is the moisture content of the seed, and it should be lowered just after harvest to minimize the loss of quality (Lang and Farouk, 2013). Thus, determination of the moisture content of the oilseed is an unavoidable operation during seed harvesting, transportation, storage, grinding and oil extraction as seed moisture affects the quality of the seed, the oil and biodiesel to be produced (Soltani et al., 2014). According to Brittaine and Lutaladio (Brittaine and Lutaladio, 2010), Jatropha seeds that are harvested from green, yellow and brown fruits could have different moisture contents and should be dried to a moisture content of around 6–10% prior to storage.

Lower Jatropha seed moisture contents compared to the normal seed storage moisture content were recommended by several researchers in order to obtain higher percentage of oil yield, and quality oil and biodiesel. For instance, in the extraction of Jatropha oil from ground kernel with moisture content of 0.912%, using Soxhlet extractor and hexane as solvent, Kadry (Kadry, 2015) obtained a maximum of 45% oil that contained 0.9% free fatty acids. The author reported that the oil obtained did not need pretreatment with acid for basic catalyzed biodiesel productions (Kadry, 2015) as its free fatty acid is less than 1% (Botero et al., 2017; Kombe and Temu, 2017). In biodiesel production from Jatropha seeds through in situ transesterification by alkaline catalyst, seeds with moisture content less than 1% was also used to prevent saponification (Kartika et al., 2013). Moreover, drying seed could reduce the amount of chemical input during in situ biodiesel production process. Haas and Scott (Haas and Scott, 2007) found that a reduction of 60% methanol and 56% sodium hydroxide input when soybean flakes with moisture contents of 7.4% were dried to the lowest moisture content (to around 0%) before in situ transesterification.

As the production and demand of Jatropha seeds increases, the existing and new technologies should be adjusted for the proper functioning of the machineries used for cultivation of Jatropha and postharvest processing of the seeds (Siqueira et al., 2012). Determination of the physical properties of seeds and their relation to the seed moisture content enables the improvement of the design of the equipment used for seed harvesting and postharvest processing (Kumar and Sharma, 2008).

Drying is one of the most important postharvest steps as it directly affects the quality of the oil, which is the main product of the Jatropha (Siqueira et al., 2013). Drying could be defined as the process of moisture removal due to heat and mass transfer between the biological product and the drying air through evaporation, and generally caused by temperature and air convection forces (Perea-Flores et al., 2012). Mathematical modeling of the drying process of the seeds helps to predict the behavior of moisture removal from the seeds, reduce the time and costs of seed drying, and helps in the invention of appropriate drying equipment (Siqueira et al., 2012, 2013).

A few available reports on the mathematical modeling of the drying process of Jatropha seeds and/or fruit focused on the drying kinetics of freshly collected seed or fruits, which contained relatively larger moisture compared to the seed at storage conditions. For instance, the mathematical modeling of drying kinetics of Jatropha seeds (Siqueira et al., 2012; Subroto, 2015) and fruits (Siqueira et al., 2013; Uthman and Onifade, 2016) were carried out on the freshly collected seeds and/or fruits. To the knowledge of the authors, drying kinetic studies on Jatropha seeds at a storage moisture content has not been reported. Thus, the primary objective of this research was to adjusted different drying mathematical models to experimental data from the drying of Jatropha seeds at a storage moisture content under different air temperatures for oil extraction and /or in situ biodiesel production, and select the model that best represents the drying process.

The current paper presents two new issues that have not been done in other works of mathematical modeling of the drying kinetics of Jatropha seeds. Firstly, the current paper deals with the mathematical modeling of the drying kinetics of *Jatropha curcas* L. seeds at a storage condition, and the seeds with moisture content of 6.81% (wt. %) was used for the drying experiments. The drying of seeds at storage might be done when one needs to reduce the moisture contents of the seeds to its minimum for oil extraction and /or for in situ biodiesel production. As aforementioned, in the previous studies of the mathematic modelling of the drying kinetics of Jatropha seeds or fruits, freshly collected seeds/fruits were used for the drying experiments. Secondly, Avhad and Marchetti drying mathematical model (Avhad and Marchetti, 2016), the model recently developed by the combination of the Page model and the Henderson and Pabis model, was used. This drying mathematical model has not been used in the previous studies for Jatropha seeds.

As this study deals with the mathematical modelling of the drying kinetics of Jatropha seeds, its focus is only on the process of moisture removal from the seeds at storage moisture content. Thus, the experiments on the effects of the pretreatment on the oil content and oil composition have not been done for this paper.

2. Methodology

2.1. Materials

Jatropha seed at a storage moisture content was used for the drying experiment. The detailed materials used during the drying experiments was described in the study of the effects of drying temperatures and pretreatment of the seeds on the drying process and physical appearance of different collections of Jatropha seeds and has been presented elsewhere (Keneni and Marchetti, 2018). A bowl-shaped mortar and pestle were used for crushing the pretreated Jatropha seeds. The particle sizes of the crushed seeds was estimated by three different stainless steel sieves with opening sizes of 500 μm, 1 mm and 2 mm woven cloth (Control Group, 15-D2245/J, 15-D2215/J and 15-D2185/J). Digital balance (Mettler-Toledo, PG 5002 Delta Range, Switzerland) with 0.01 mg accuracy was used to weigh the seeds samples. The samples were placed on the pyrex glass petri plates during drying. The drying experiments and determination of the initial and residual moisture contents of the seed samples were performed using a standard heating furnace (Narbetherm P300, Germany).

2.2. Seed drying experiments

The seed drying experiments were performed in the Laboratory of Norwegian University of Life Sciences (NMBU), Faculty of Science and Technology. The initial moisture contents of the Jatropha seed samples used for this experiment has been determined prior to this experiment (Keneni and Marchetti, 2018) by drying 15 g seeds at 105 °C for 24 h (Bamgboye and Adebayo, 2012; Garnayak et al., 2008; Siqueira et al., 2012; Subroto, 2015). Accordingly, the moisture content of the seeds was found to be 6.81% (wt. %), and thus, the moisture content of the seed samples used for the experiment was in the range of the recommended storage moisture content (6–10%) for Jatropha seeds (Brittaine and Lutaladio, 2010).

The pretreated (crushed seeds, CS) and non-pretreated (the whole seeds, WS) were used for the drying experiments. After crushing the seeds using mortar and pestle, the particle sizes of the pulverized seeds were estimated by three different sieves with openings of 500 μm, 1 mm and 2 mm woven cloth. For every drying experiment, ca. 15 g of Jatropha seeds were used and both treatments were duplicated. Thus, the crushed seeds used in the drying experiment was a mixture of four particles sizes. The average proportions (%) of the particle size of crushed seeds (PSCS): PSCS > 2 mm, 2 mm > PSCS > 1 mm, 1 mm > PSCS > 500 μm and PSCS < 500 μm were 14.95 ± 8.2, 29.96 ± 4.38, 35.32 ± 10.5 and 19.77 ± 4.74, respectively.

The pretreated (crushed seeds) and non-pretreated (whole seeds) seeds were placed on separate petri plates, and dried at five different drying temperatures (313, 323, 333, 343, and 353 K) in the standard heating furnace. The seed samples were weighted before inserting into the heating furnace, during the progress of the drying experiments (at the predetermined time intervals) and at the end of the drying experiments. The temperature of the heating furnace was set at the required drying temperature and maintained the set temperature for an hour before placing the samples in the furnace. This was to minimize the fluctuation of the surrounding drying air temperature during drying (Avhad and Marchetti, 2016). During the experiments, the seed samples were removed from the heating furnace at a predetermined time interval, weighted and put back into the furnace by taking less than 10 s to weigh the samples. The experiments were performed until no change in weight had been recorded for three successive weight measurements for the respective drying temperatures, which was assumed as the stage of equilibrium. Accordingly, the drying experiment at a particular temperature was carried out for four days (5760 min) in order to ensure the achievement of the critical moisture level at which no more moisture loss occurred. Every experiments were done twice and the average values were used in data analysis and reporting.

Drying data obtained from the weight measurements of the seed samples at different temperatures and drying times were changed to moisture content data in order to use it in the drying kinetics.

2.3. Mathematical modeling for the drying kinetics

Mathematical modeling of the seed drying is used to determine the optimum drying parameters and the performance of the process. It is essential to select the drying mathematical model that fits best to the drying curves under different conditions (Fudholi et al., 2012). To predict the drying kinetics of Jatropha seeds, mathematical modeling of the process of moisture evaporation from the seed is needed.

In the current study, the drying data from five different drying temperatures were fitted to three selected mathematical models: Lewis, Henderson and Pabis, and Page models. These were the most commonly used mathematical models to predict the drying process of different biological materials (Ghodake et al., 2006). The experimental data were also fitted to the Avhad and Marchetti model, which is a combination of Page model and Henderson and Pabis model and found to be best fitted to the drying kinetics of Hass avocado seeds (Avhad and Marchetti, 2016). The models mentioned above are all semi-theoretical, as in agreement with the references Chukwunonye et al. (Chukwunonye et al., 2016) and McMinn et al. (McMinn et al., 2005). These semi-theoretical models were the most commonly used and discussed models in literature for similar products, and they are used for the current study based on the information obtained from the literature.

In the present experiment, the recorded weight loss data of the seed samples at different time intervals were converted to the moisture loss data. From the initial moisture content of the seed, the moisture content data at different time intervals and the residual moisture contents for different temperatures, the dimensionless moisture ratio (MR) was calculated. Then, the MR as a function of time was used for fitting the mathematical models. The expression used to calculate the MR of Jatropha seed samples was written in Eq. (1).

$$MR = \frac{M_t - M_e}{M_0 - M_e} \tag{1}$$

where, M_0, M_t and M_e refer to the initial moisture content, moisture content at time t, and the equilibrium moisture content for the seed samples, respectively. The equilibrium moisture content (M_e) of Jatropha seeds at each temperature was obtained experimentally by drying the seed samples in the oven until no change in weight occurred for three successive weight measurements (Siqueira et al., 2012).

The activation energy that is required to start the drying process, namely water activation of the seed (Voća et al., 2007), could be found using the Arrhenius equation. It is the energy barrier that should be overcome in order to trigger moisture diffusion during drying (Perea-Flores et al., 2012). In the present study, the drying rate constant "k" and the drying activation energies (Ea) were used to analyze the change of moisture content at different temperatures. Activation energies (Ea) for the crushed and whole seeds of Jatropha were obtained from the slopes of the plots of ln(k) versus T^{-1} predicted using the experimental data for the tested models using Arrhenius equation. Then, the k values calculated from the activation energy for the respective drying temperatures were inserted into the expression of all drying mathematical models. Similarly, the rate constant k was used in the studies of drying kinetics of *Cuminum cyminum* grains (Zomorodian and Moradi, 2010), Jatropha seeds (Siqueira et al., 2012), plantain sample (Oforkansi and Oduola, 2016), and pumpkin fruit slices (Onwude et al., 2016a). Based on the information obtained from the aforementioned publications, the drying rate constant "k" and the drying activation energies (Ea) were used in this paper. Arrhenius equation is presented in Eq. (2).

$$k = A\exp\left(-\frac{Ea}{RT}\right) \tag{2}$$

where, k refers to rate constant, A is the pre-exponential factor, Ea is the activation energy (KJ $mol-^{1}$), R is the universal gas constant (8.314 J mol^{-1}K), and T is the absolute air temperature (K).

2.3.1. Lewis model

Lewis model is the simplest model as it contains only one model constant. The model has been widely applied in describing the drying behavior of different agricultural products (Onwude et al., 2016b). According to Lewis (Lewis, 1921), the change in moisture content in the falling rate period of drying is proportional to the instantaneous difference between the moisture content and the expected moisture content when it comes into equilibrium with the humidity of the surrounding drying air. Lewis model neglects the resistance to the moisture movement from the inner to the surface of the material during drying. Although the model is simple to use, it underestimates the initial parts of drying curve and overestimates later phases (Chukwunonye et al., 2016). Lewis model was used to fit the dryings of black tea (Panchariya et al., 2002), grape seeds (Roberts et al., 2008) and strawberries (Changrue et al., 2008). The expression for the model is presented in Eq. (3).

$$MR = \exp(-kt) \tag{3}$$

where, k is the model constant that follows an Arrhenius equation, and t is the time.

2.3.2. Henderson and Pabis model

The Henderson and Pabis model is related to Fick's second law, and sometimes it is named as bi-parametric exponential model (Zhang et al., 2016). The Henderson and Pabis model has produced good fit in predicting the drying of orange seed (Rosa et al., 2015), African breadfruit (Shittu and Raji, 2011) and dill leaves (Dikmen et al., 2018). The model is presented in Eq. (4).

$$MR = a\exp(-kt) \tag{4}$$

where, a and k are the constants of the model that follow Arrhenius equation.

2.3.3. Page model

The Page model is a two constant empirical modification of the Lewis model that corrects some shortcomings (Zhang et al., 2016). Page model has been used to describe the drying process of bay leaves (Gunhan et al., 2005), cashew kernels (Shittu and Raji, 2011), mango slices (Akoy, 2014), and moringa seeds (Aremu and Akintola, 2016). The model expression can be seen in Eq. (5).

$$MR = \exp(-kt^N) \tag{5}$$

where, k and N are the constants of the model.

2.3.4. Avhad and Marchetti model

The Avhad and Marchetti model is a mathematical model that was developed by the combination of the Page model and the Henderson and Pabis model, and successfully fitted to the drying kinetics of Hass avocado seeds (Avhad and Marchetti, 2016). Avhad and Marchetti model takes into account the benefits of the three discussed models together, and that its fitness could be equally good or better. However, since the model has some parameters to be determinate due to the experimental data, it is not 100% certain that the model will always give equally good or better results. The Avhad and Marchetti model is presented in Eq. (6).

$$MR = a\exp(-kt^N) \tag{6}$$

where, a, k and N are the constants of model.

The coefficients of the drying mathematical models and the regression/statistical parameters were obtained using the optimization mechanism of Microsoft excel solver (Microsoft Excel, 2013) (Oforkansi and Oduola, 2016).

2.4. Comparison of the fitness of the models

The values of five statistical parameters were used to compare the fitness of the data predicted by the drying mathematical models to the drying curves of the experimental data. The parameters utilized include coefficient of determination (R^2), chi-square test (X^2), root mean square error (R_{MSE}), mean bias error (MBE), and mean absolute error (MAE). The statistical parameters for comparison of the models were selected based on other similar publications where these parameters were used for the selection of the most fitted model. This standard procedure has been accepted in the literature for such comparisons. For instance, in the papers of Gunhan et al. (Gunhan et al., 2005), Zomorodian and Moradi (Zomorodian and Moradi, 2010), Sridhar and Madhu (Sridhar and Madhu, 2015), Naderinezhad et al. (Naderinezhad et al., 2016), Oforkansi and Oduola (Oforkansi and Oduola, 2016), and Mazandarani et al. (Mazandarani et al., 2017) similar parameters were used to compare the fitness of different models.

R-squared or coefficient of determination (R^2) is the measures of how close the statistical data could fit the regression line (Onwude et al., 2016a). According to Gunhan et al. (Gunhan et al., 2005), the R_{MSE} provides information on the short-term performance and its value is always positive while MBE gives information on the long-term performance of the correlations by comparing the actual deviation between predicted and experimental values term by term. The author also indicated that for both R_{MSE} and MBE, the ideal value is 'zero'.

The value of R^2 is the primary criteria for selecting the best-fit model to the drying kinetics of agricultural products (Doymaz, 2010). During the comparison of the fitness of the models to the experimental drying curves, the most fitted model must have the largest values of R^2, and conversely, it should have the smallest values of X^2, R_{MSE}, MBE and MAE (Younis et al., 2018; Zhang et al., 2016).

The statistical parameter R^2 was calculated according the expressions by Oforkansi and Oduola (Oforkansi and Oduola, 2016). The other statistical parameters were calculated using the equations used by Gunhan et al (Gunhan et al., 2005), Zomorodian and Moradi (Zomorodian and Moradi, 2010), Sridhar and Madhu (Sridhar and Madhu, 2015), Naderinezhad et al. (Naderinezhad et al., 2016), Oforkansi and Oduola (Oforkansi and Oduola, 2016), Avhad and Marchetti (Avhad and Marchetti, 2016), and Mazandarani et al. (Mazandarani et al., 2017). The expression for these statistical parameters were written in Eqs. (7)–(11) as:

$$R^2 = 1 - \left(\frac{\sum_{i=1}^{N}(MR_{pre,i} - MR_{exp,i})^2}{\sum_{i=1}^{N}(MR_{exp,i} - \bar{M}R_{exp})^2}\right) \tag{7}$$

$$X^2 = \frac{\sum_{i,=1}^{N}(MR_{ex,i} - MR_{pr,i})^2}{N - z} \tag{8}$$

$$E_{RMS} = \left[\frac{1}{N}\sum_{i,=1}^{N}(MR_{ex,i} - MR_{pr,i})^2\right]^{\frac{1}{2}} \tag{9}$$

$$MBE = \frac{1}{N}\sum_{i=1}^{N}(MR_{ex,i} - MR_{pr,i}) \tag{10}$$

$$MAE = \frac{1}{N}\sum_{i=1}^{N}|MR_{ex,i} - MR_{pr,i}| \tag{11}$$

where, $MR_{exp,i}$ is the i^{th} experimental moisture ratio; $MR_{pre,i}$ is the i^{th} predicted moisture ratio; $\bar{M}R_{exp}$ is the mean of the experimental moisture ratio; N refers to the number of observations; z represents the number of constants in the models.

3. Results and discussion

3.1. Moisture ratio and seed drying process

The weight loss data collected during Jatropha seed dryings were converted to moisture ratios and their variations as a function of drying time were plotted. Fig. 1a and b show the moisture ratio versus drying time at the drying temperatures of 313, 323, 333, 343 and 353 K. As it can be observed from Fig. 1a and b, rises in the dying temperature resulted in the increasing of the rate of moisture evaporation from the

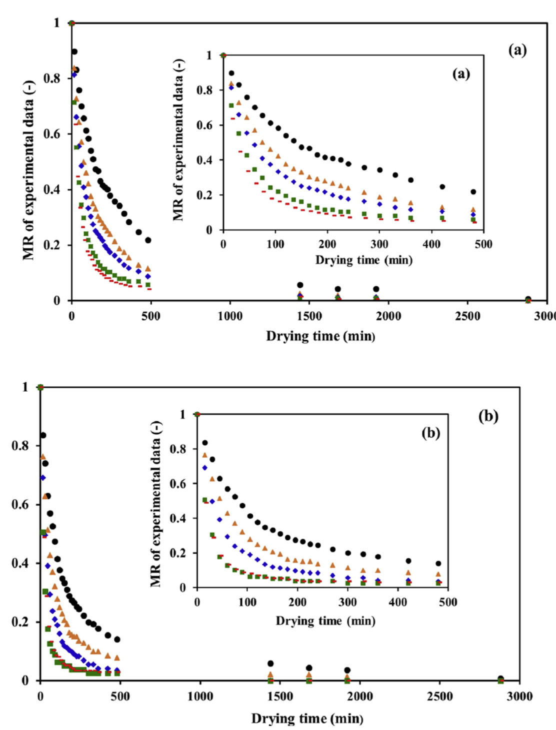

Fig. 1. (a) Moisture ratio vs. drying time at different air temperatures for the whole seeds: (●) 313 K, (▲) 323 K, (♦) 333 K, (■) 343 K and (–) 353 K; (b) Moisture ratio vs. drying time at different air temperatures for the crushed: (●) 313 K, (▲) 323 K, (♦) 333 K, (■) 343 K and (–) 353 K.

seed samples. Similar behavior was observed in a number of agricultural products such as grape seeds (Roberts et al., 2008), pumpkin seeds (Jittanit, 2011), caster bean (Perea-Flores et al., 2012), fresh Jatropha seeds (Siqueira et al., 2012), sorghum grains (Resende et al., 2014), orange seeds (Rosa et al., 2015) and Hass avocado seeds (Avhad and Marchetti, 2016). According to Siqueira et al. (Siqueira et al., 2012) and Rosa et al. (Rosa et al., 2015), rising in temperature increases the difference between the vapor pressure of the drying air and that of the seed samples, thus, higher temperature results in greater and faster water removal.

As it could be seen from Fig. 1a, in the WS of Jatropha, the drying time required to reduce the moisture ratio to below 0.5 when using 313 K was about 5 times more than that required at 353 K drying temperature. The time taken to reduce the moisture ratio of CS to below 0.5 at 313 K was also 6 times longer than when it was dried at 353 K, as it can be seen in Fig. 1 b. Moreover, the reduction of moisture ratios of the WS to below 0.5 when drying at 313 K and 353 K required twice more times compared to that of the CS dried at the respective temperatures. The greater moisture evaporation rate for the CS compared to that of the WS might be due to the greater surface area exposed to the drying temperature in the case of the CS.

In the drying of the whole seeds (Fig. 1a) and the crushed seeds (Fig.1b) of Jatropha, the moisture evaporation rate was faster at the beginning due to high level of water to be removed, and decreasing as the equilibrium moisture content approached. According to Sandeepa et al. (Sandeepa et al., 2013), in the drying experiment, the decreasing of drying rate from the initial to the end of drying shows the non-existence of constant rate period or the existence of constant rate for an insignificant period of time relative to the entire time of drying. During drying, the rate of moisture evaporation will be higher at the early stage of drying due to larger moisture content of the seeds, and reduces as the moisture content decreases.

3.2. Determination of activation energy

Fig. 2a and b show the plot of ln(k) versus T^{-1} for WS and CS of Jatropha, respectively at the five drying temperatures when Avhad and Marchetti model (Avhad and Marchetti, 2016) was used. The computed value of the activation energies and pre-exponential factors for the employed models are also presented in Table 1. As it can be seen from Table 1, the activation energy value for the whole seeds and crushed seeds of Jatropha varied from 23.67 to 36.06 and 32.88 to 45.75 KJ mol^{-1}, respectively for all mathematical models used. The activation energies of the WS and CS of Jatropha were in line with those reported for other agricultural products such as sorghum (Resende et al., 2014), grape seeds (Roberts et al., 2008), sliced, and crushed Hass avocado seeds (Avhad and Marchetti, 2016) and castor oil seeds (Perea-Flores et al., 2012).

As it can be observed from Table 1, the activation energy of the CS was greater than that of the WS and this was unexpected as the rate of water evaporation in the crushed seed was faster than that of the WS. In the study of activation energy of water release rate from corn kernel, Voća, et al. (Voća et al., 2007) found that the drying rate constant k significantly increased with the increasing of drying air temperature, and described activation energy as the energy that needs to be supplied to kernels for initiating the moisture release. The authors concluded that if the activation energy is higher, the moisture release from the kernels became slower. Generally, the values of activation energy are related to the nature of a materials to be dried, and thus, if water is more strongly bounded to the structure of the material, it will be more difficult to removed it (Bezerra et al., 2015). The present result was in contrary to the finding of Avhad and Marchetti (Avhad and Marchetti, 2016) in which the activation energy of the crushed Hass avocado seeds (24–32 KJ mol^{-1}) was found to be less than that of the sliced (34–36 KJ mol^{-1}) and non-pretreated (43–129 KJ mol^{-1}) seeds.

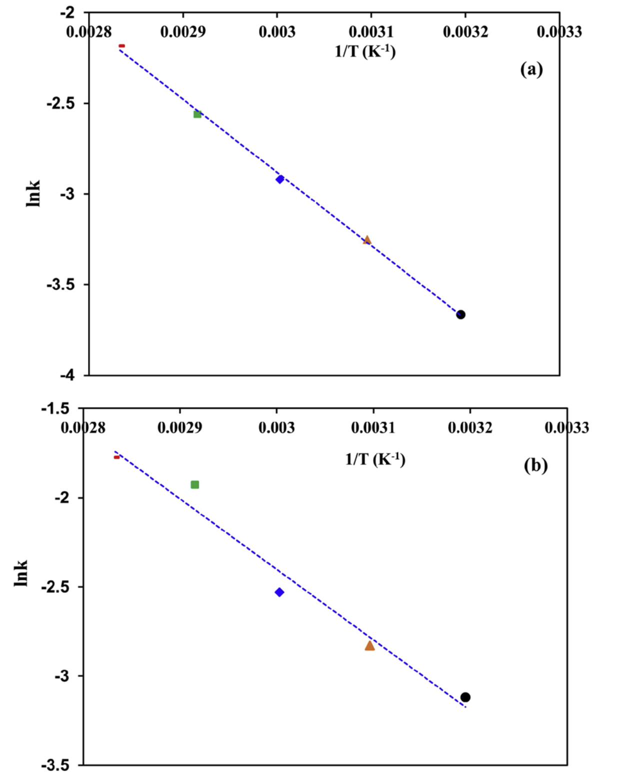

Fig. 2. (a) Arrhenius plot between ln(k) versus 1/T for the whole seeds of Jatropha using Avhad and Marchetti model: (●) 313 K, (▲) 323 K, (♦) 333 K, (■) 343 K and (▬) 353 K; (b) Arrhenius plot between ln(k) versus 1/T for the crushed seeds of Jatropha: (●) 313 K, (▲) 323 K, (♦) 333 K, (■) 343 K and (▬) 353 K.

Table 1
Estimated activation energy and pre-exponential factor for the whole seeds and crushed seeds of Jatropha.

Models	Whole seeds		Crushed seeds	
	Ea (KJ mol^{-1})	A	Ea (KJ mol^{-1})	A
Lewis	35.5956	3.8372×10^3	42.333	8.253×10^4
Henderson and Pabis	36.063	3.891×10^3	45.753	2.563×10^5
Page	23.6706	2.8911×10^4	35.7759	3.468×10^4
Avhad and Marchetti	33.533	1.0064×10^4	32.885	1.288×10^4

3.3. Mathematical modeling of seed drying

The predicted data by the employed mathematical models were fitted to the drying curves of the experimental data of the WS and CS of Jatropha to select a model that best describe the drying process of the seeds. Figs. 3a–c and 4 a–c show the comparison of the four drying mathematical models and the experimental data obtained for the WS and CS of Jatropha, respectively at 313 K (the lowest), 333 K (medium) and 353 K (the highest) air temperatures of the experiments. As it could be seen from the graphical presentations, all the employed models could describe the drying kinetics of the Jatropha seeds. However, the selection of best fit mathematical model was based on the values R^2, X^2, R_{MSE}, MBE and MAE, and as it was aforementioned, the selection of best fit is primarily based on the values of R^2 (Doymaz, 2010). In the mathematical modelling of the drying kinetics castor oil seeds, Perea-Flores et al. (Perea-Flores et al., 2012) accepted the mathematical models with R^2 values greater than 0.97 as fit models to the

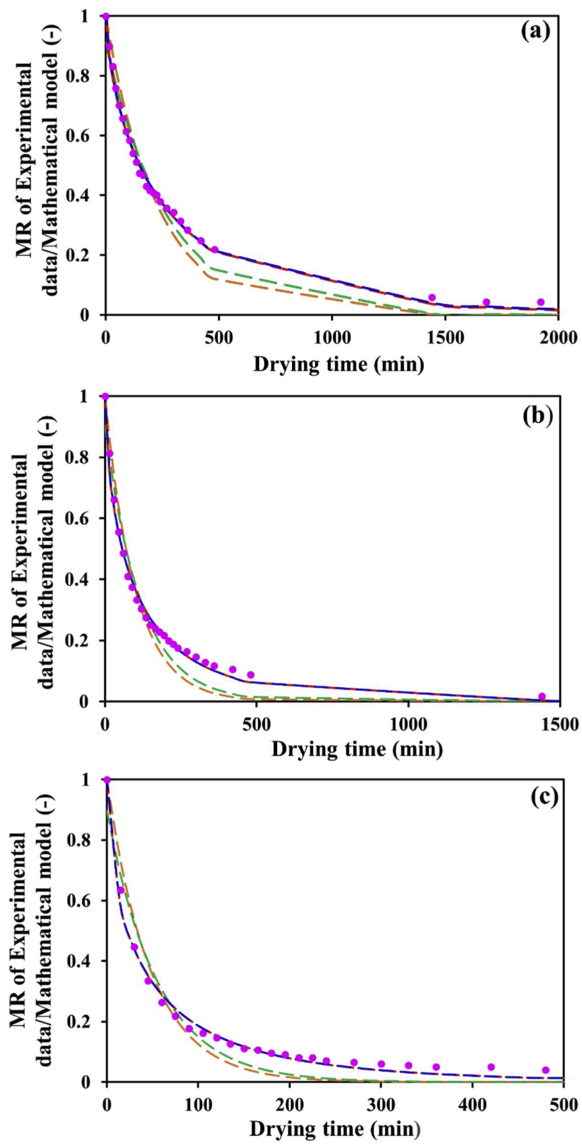

Fig. 3. Comparison of the experimental and predicted moisture ratios using the four different drying mathematical models at (a) 313 K, (b) 333 K and (c) 353 K for the whole seeds of Jatropha: (●) Experimental data, (—) Lewis model, (—) Henderson and Pabis model, (—) Page model and (—) Avhad and Marchetti model.

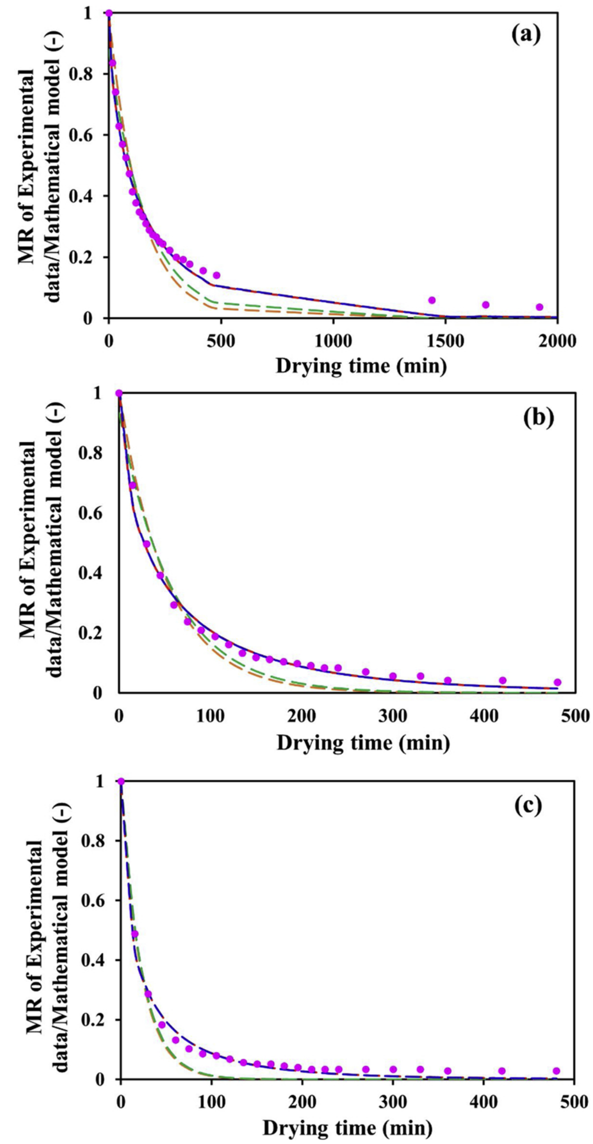

Fig. 4. Comparison of the experimental and predicted moisture ratios using the four different drying mathematical models at (a) 313 K, (b) 333 K and (c) 353 K for the crushed seeds of Jatropha: (●) Experimental data, (—) Lewis model, (—) Henderson and Pabis model, (—) Page model and (—) Avhad and Marchetti model.

experimental data.

The calculated statistical parameters for the WS and CS of Jatropha for all the four models and the drying air temperatures (313–353 K) is presented in Table 2. As it can be seed from Table 2, the values of R^2, X^2, R_{MSE}, MBE and MAE for the WS of Jatropha for all the drying models and drying temperatures ranged from 0.9278 to 0.9969, 2.37×10^{-4} and 4.29×10^{-3}, 0.01454 and 0.06427, $1.95 \times 10^{-3}$and 2.97×10^{-2}, and 0.01171 and 0.05559, respectively. In the CS of Jatropha, the R^2, X^2, R_{MSE}, MBE and MAE values changed between 0.9361 and 0.9917, 4.18×10^{-4} and 4.01×10^{-3}, 0.02039 and 0.06224, 3.64×10^{-3} and 0.04043, and 0.01639 and 0.05318, respectively.

From the four mathematical models, the Avhad and Marchetti model was found to show best fit to experimental data, with the values of coefficient of determination ranging from 0.9914 to 0.9969 and 0.9908 to 0.9917 for the WS and CS of Jatropha, respectively when analyzing all temperature ranges. In the drying of the WS, the values of R^2 for Avhad and Marchetti model were the closest to 1 when compared to that of all other models used. The maximum value of R^2 (0.9969) and the smallest values of X^2 (2.37×10^{-4}), R_{MSE} (0.01454), MBE (1.95×10^{-3}) and MAE (0.01171) were obtained when the whole seeds were dried at 313 K and the Avhad and Marchetti mode (Avhad and Marchetti, 2016) was employed. Moreover, in the drying of crushed seeds of Jatropha, the maximum R^2 value (0.9917) was obtained when the seeds were dried at 333 K and Avhad and Marchetti model was used. The smallest values of E_{RMS} (0.02039) and MBE (3.64×10^{-3}) for crushed seeds were also found when Avhad and Marchetti model was used.

The Page model was found to have a satisfactory fitting with the experimental data as well. Although the fitness of Page model was comparable to that of Avhad and Marchetti, the latter model was found to be superior to fit to the experimental data. As it could be seen from Table 2, the value of R^2 for Avhad and Marchetti model were slightly larger than that of Page model while the X^2 and other statistical parameters for Avhad and Marchetti model were found to be smaller compared to that of the Page model.

Fig. 5a and b show the fitness of Avhad and Marchetti model with

Table 2
Results obtained from the statistical analysis of the four selected drying mathematical models at 313–353 K temperatures for the whole seeds and crushed seeds of Jatropha.

Model	Temperature (K)	Whole seeds					Crushed seeds				
		R^2	X^2	E_{RMS}	MBE	MAE	R^2	X^2	E_{RMS}	MBE	MAE
Lewis	313	0.9607	2.82×10^{-3}	0.05217	0.005330	0.045256	0.9361	4.01×10^{-3}	0.06224	0.01201	0.05318
	323	0.9397	3.75×10^{-3}	0.06008	−0.00946	0.05189	0.9372	3.61×10^{-3}	0.05898	0.02363	0.05199
	333	0.9278	4.29×10^{-3}	0.06427	0.02604	0.05559	0.9441	3.12×10^{-3}	0.05467	0.03314	0.05066
	343	0.9356	3.48×10^{-3}	0.05787	0.01858	0.05071	0.9442	2.69×10^{-3}	0.05077	0.00717	0.04071
	353	0.9304	3.40×10^{-3}	0.05724	0.02972	0.05056	0.9536	2.19×10^{-3}	0.04577	0.04043	0.04231
Henderson and Pabis	313	0.9775	1.67×10^{-3}	0.03948	0.00908	0.03426	0.9583	2.72×10^{-3}	0.05026	0.01408	0.04218
	323	0.9646	2.29×10^{-3}	0.04601	0.00555	0.03742	0.9539	2.76×10^{-3}	0.05055	0.02205	0.04356
	333	0.9506	2.82 × 10-3	0.05318	0.02124	0.04627	0.9534	2.72×10^{-3}	0.04990	0.03018	0.047441
	343	0.9495	2.84×10^{-3}	0.05123	0.01991	0.04371	0.9508	2.48×10^{-3}	0.04763	0.01527	0.04081
	353	0.9412	2.99×10^{-3}	0.05262	0.027791	0.04715	0.9563	2.16×10^{-3}	0.04444	0.03820	0.04131
Page	313	0.9964	2.63×10^{-4}	0.01563	3.63×10^{-3}	0.01223	0.9908	5.98×10^{-4}	0.02356	6.61×10^{-3}	0.01881
	323	0.9959	2.65×10^{-4}	0.01565	3.37×10^{-3}	0.01366	0.9913	5.19×10^{-4}	0.02190	6.01×10^{-3}	0.01889
	333	0.9926	4.55×10^{-4}	0.02051	438×10^{-3}	0.01554	0.9915	4.96×10^{-4}	0.02128	4.81×10^{-3}	0.01639
	343	0.9912	4.91×10^{-4}	0.02130	5.89×10^{-3}	0.01760	0.9888	5.66×10^{-4}	0.02274	0.01128	0.02052
	353	0.9918	4.18×10^{-4}	0.01965	5.78×10^{-3}	0.01590	0.9907	4.18×10^{-4}	0.02046	7.58×10^{-3}	0.01676
Avhad and Marchetti	313	0.9969	2.37×10^{-4}	0.01454	1.95×10^{-3}	0.01171	0.9913	5.86×10^{-4}	0.02288	5.51×10^{-3}	0.01877
	323	0.9962	2.53×10^{-4}	0.01496	2.48×10^{-3}	0.01336	0.9916	5.23×10^{-4}	0.02152	4.81×10^{-3}	0.01917
	333	0.9931	4.45×10^{-4}	0.01985	3.03×10^{-3}	0.01589	0.9917	5.08×10^{-4}	0.02101	3.64×10^{-3}	0.01701
	343	0.9914	5.00×10^{-4}	0.02104	5.04×10^{-3}	0.01791	0.9889	5.88×10^{-4}	0.02262	1.09×10^{-2}	0.02039
	353	0.9919	4.31×10^{-4}	0.01952	5.36×10^{-3}	0.01610	0.9908	4.78×10^{-4}	0.02039	7.48×10^{-3}	0.01721

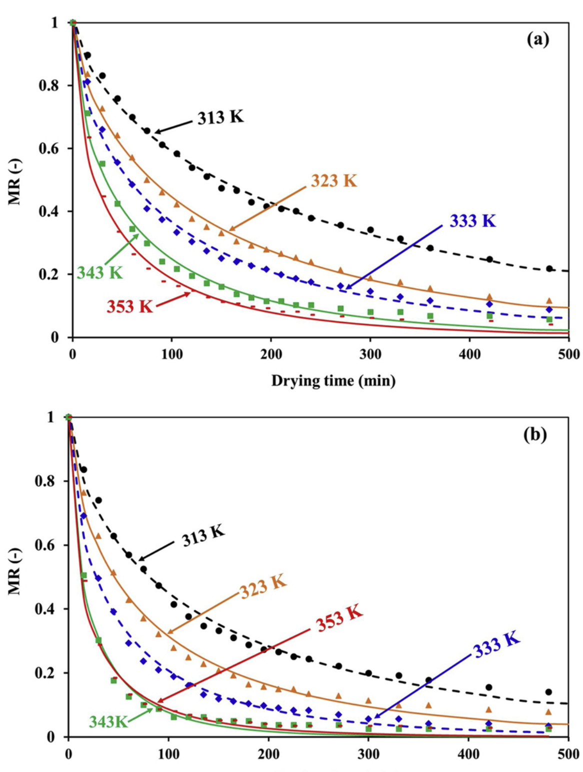

Fig. 5. (a) Comparison of the experimental and predicted moisture ratios using the Avhad and Marchetti drying mathematical model at 313–353 K air temperatures for whole seeds of Jatropha: (●) Experimental data, () model; (▲) Experimental data, (—) model; (♦) Experimental data, (—)model; (■) Experimental data, (—) model; (—) Experimental data, (—) model. (b) Comparison of the experimental and predicted moisture ratios using the Avhad and Marchetti drying mathematical model at 313–353 K air temperatures for crushed seeds of Jatropha: (●) Experimental data, () model; (▲) Experimental data, (—) model; (♦) Experimental data, (—)model; (■) Experimental data, (—) model; (—) Experimental data, (—) model.

Table 3
Constants of the Avhad and Marchetti model for the whole seeds and crushed seeds of Jatropha.

Temperature	Whole seeds			Crushed seeds		
	k (min^{-1})	a	N	k (min^{-1})	a	N
313	0.0255	1.0263	0.6663	0.04186	1.0219	0.6459
323	0.0379	1.0186	0.6684	0.0619	1.0244	0.6399
333	0.0552	1.0267	0.6349	0.0894	1.0241	0.6258
343	0.0786	1.0180	0.6264	0.1264	1.00028	0.6538
353	0.1098	1.0092	0.5936	0.1752	1.0068	0.5725

the experimental data for temperatures varying from 313 K to 353 K for the WS and CS of Jatropha. The values of the constants (k, a and N) obtained for the Avhad and Marchetti model are also presented in Table 3. As it can be seen from Table 3, the values of the drying rate constants k (moisture release rate constant) found to increase with the drying temperature for both the WS and CS as expected. The estimated values for the parameter a and N were not constant and found to vary with the drying air temperature, and both parameters tend to decrease with the rising of the drying temperatures and increasing of drying rate constant k. In contrary to the present finding, Simal et al.(Simal et al., 2005) reported the study in which the calculated value for N parameter for Page model did not exhibited temperature dependence, and considered as a constant parameter (N = 0.796).

Figs. 6a–c and 7 a–c show the predicted moisture ratio by Avhad and Marchetti model versus experimental moisture ratio of the WS and CS of Jatropha, respectively at 313 K (the lowest), 333 K (medium) and 353 K (the highest) air temperatures of the experiments. As it is evident from Figs. 6a–c and 7 a–c, the predicted moisture ratio by Avhad and Marchetti model generally banded around the straight line which showed the suitability of the model in describing the drying behavior of the WS and CS of Jatropha, respectively.

4. Conclusions

In this study, to describe the drying kinetics of Jatropha seeds at a storage moisture content, non-pretreated (whole seeds) and pretreated (crushed seeds) seeds were dried at temperatures ranged between 313 and 353 K. The fitness of four different semi-theoretical mathematical

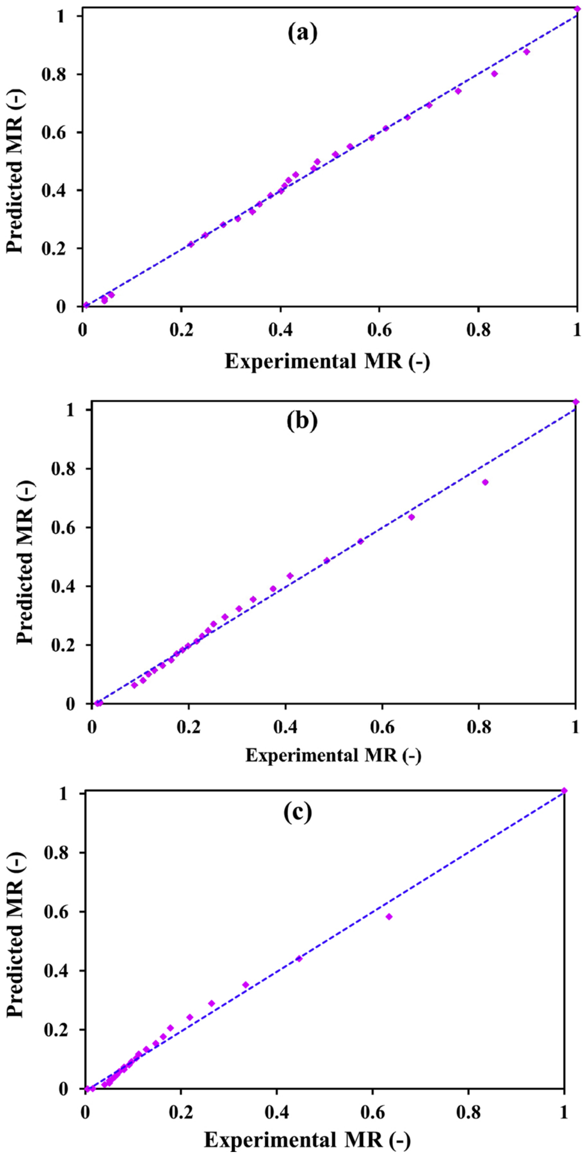

Fig. 6. Predicted moisture ratio by Avhad and Marchetti model versus experimental moisture ratio of the whole seeds of Jatropha at the drying temperatures of (a) 313k, (b)333 K, (c) 353 K: (♦) predicted moisture ratio and (–) regression line.

models (Lewis model, Henderson and Pabis model, Page model and Avhad and Marchetti model) to drying curves were compared by employing coefficient of determination, chi-square test, root mean square error, mean bias error, and mean absolute error. It was found that the moisture removal rate increased with the rising of the oven air temperature and decreased with time due to the reduction of the seed moisture content. Avhad and Marchetti model and the Page model gave better and a more comparable fit to the experimental data than the other models. However, the Avhad and Marchetti model with R^2 ranged from 0.9914 to 0.9969 and 0.9908 to 0.9917 for the whole seeds and crushed seeds, respectively for all the drying temperatures and models was found to show best fit to the drying kinetics of the Jatropha seeds at a storage moisture content. The Avhad and Marchetti model showed superior fit to the experimental data at the drying temperature of 313 K with R^2 of 0.9969 for the whole seed, and at 333 K in case of crushed seeds for which the R^2 value was 0.9917. The activation energies of the whole and crushed seeds of Jatropha when using Avhad and Marchetti model were found to be 33.53 and 32.885 KJ mol^{-1}, respectively.

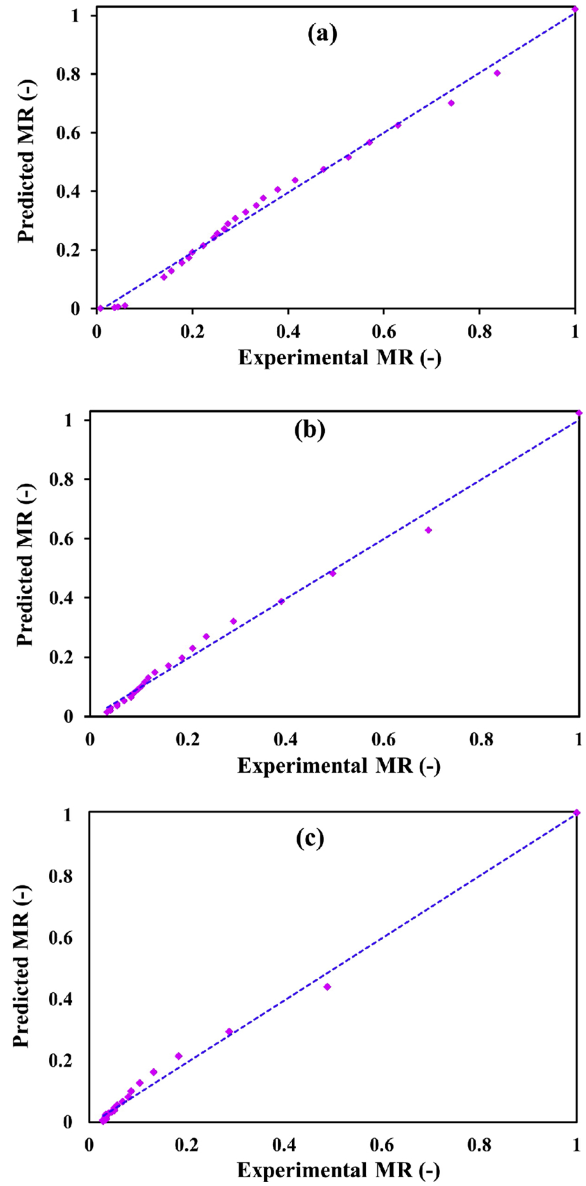

Fig. 7. Predicted moisture ratio by Avhad and Marchetti model versus experimental moisture ratio of the crushed seeds of Jatropha at the drying temperatures of (a) 313k, (b)333 K, (c) 353 K: (♦) predicted moisture ratio and (–) regression line.

Declarations of interest

The authors declare no competing financial interest.

Acknowledgements

The financial support from the Norad/EnPe project entitled "Research and Capacity Building in Clean and Renewable Bioenergy in Ethiopia", Agreement No. 2015/8397 to the Norwegian University of Life Sciences (NMBU), is gratefully acknowledged.

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