

1 **Water adsorption and desorption isotherms of chestnut and wheat**
2 **flours**

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6 **Abstract**

7 Adsorption and desorption isotherms of chestnut and wheat flour at different
8 temperatures (20, 35, 50 and 65°C) were determined over a range of water activity
9 of 0.09-0.91 using a static gravimetric method. According to BET classification
10 the obtained isotherms for both the flours were of Type II. Taking into account
11 IUPAC classification the hysteresis loops were of Type H3. GAB and Chung-
12 Pfoest models were used in order to fit the experimental data with satisfactory
13 results. The goodness of fittings were evaluated by statistical parameters which
14 showed values of $E < 0.046$, $E_{RMS} < 0.026$ and $R^2 > 0.980$. The GAB model gave the
15 best fit to the experimental data for both flours. The net isosteric heats of sorption,
16 estimated by the Clausius–Clapeyron equation, decreased in both flours when
17 moisture content of samples increased. Heats of desorption were higher than the
18 heats corresponding to adsorption at each moisture content and temperature in
19 both flours, but in the case of wheat flour strong differences at low moisture
20 contents were found.

21

22 **Keywords:** chestnut flour, wheat flour, equilibrium moisture content, water
23 activity, sorption heat.

24 Nomenclature

A	Chung-Pfost (Eq. 6) parameter (kg (kg d.b.) ⁻¹)
a _w	Water activity (-)
B	Chung-Pfost (Eq. 6) parameter (kg (kg d.b.) ⁻¹)
C	GAB (Eq. 1) parameter (-)
E	Mean relative percentage deviation modulus (%)
E _{RMS}	Root mean square error (kg (kg d.b.) ⁻¹)
H	Heat of GAB model (kJ mol ⁻¹)
h	Sorption heat (kJ mol ⁻¹)
h _L	Heat of water condensation (kJ mol ⁻¹)
K	GAB (Eq. 1) parameter (-)
N	Number of samples (-)
R	Ideal gas constant (8.314 J (mol K) ⁻¹)
R ²	Coefficient of determination (-)
T	Absolute temperature (K)
X	Equilibrium moisture content (kg (kg d.b.) ⁻¹)

Subscript

cal	Calculated
e	Net isosteric
exp	Experimental
M	Monolayer
N	Multilayer
o	Pre-exponential

26 **Introduction**

27 The chestnut is a seasonal nut (autumn and winter) that belongs to *Fagaceae*
28 family. The European chestnut (*Castanea sativa* Mill) is the most appreciated by
29 size, flavour and industrial processing (Moreira et al., 2005). The value of total
30 European production of chestnuts was approximately about 129,000 t in 2007
31 (FAO, 2009). In order to extend its useful life and consume it is necessary to
32 obtain derived products such as flour and starch, which present two specific
33 advantages: low cholesterol and free gluten contents (products for celiac people).

34 The chestnut flour was marginally obtained by milling of chestnuts with smaller
35 size. Actually it can be considered an interesting product by its characteristics and
36 higher commercial value for products based on chestnut flour has been observed
37 (Chenlo et al., 2007). Its use is more frequent on the manufacturing of a sort of
38 products like snacks, biscuits, cakes, flakes, pasta, purees and creams among
39 others (Harold, 1984; Sacchetti et al., 2004). The chestnut flour has an average
40 content of $6.2 \pm 0.7\%$ of protein, $63.5 \pm 13.8\%$ of starch, $22.9 \pm 9.2\%$ of sugars,
41 $3.6 \pm 1.7\%$ of fat and $3.8 \pm 1.6\%$ of fibre (Demiate et al., 2001; Sacchetti et al.,
42 2004). The wheat flour (*Triticum aestivum* L.) has a content of $11.4 \pm 1.1\%$ of
43 proteins, $82.1 \pm 3.1\%$ of starch, $0.6 \pm 0.0\%$ of sugars, $1.9 \pm 0.3\%$ of fat and $4.1 \pm$
44 1.5% fiber (Danish food composition databank, 2009).

45 One of the most important insects that it is bred in the European chestnut is the
46 chestnut weevil *Curculio elephas* (Debouzie et al., 2002). The *Heteropterous*
47 insects responsible for damage of wheat in Europe are species belonging to
48 *Eurygaster* and *Aelia* (Aja et al., 2004). Delobel and Grenier (1993) studied the
49 development and growth of the cereal weevils of *Sitophilus (zeamais, oryzae,*
50 *granarius* and *linearis)* on non-cereal foods as chestnut, showing the high

51 adaptability of cereal weevils on chestnuts. In order to avoid the loss of quality of
52 chestnut and wheat flours due to insects attack the control of moisture content
53 during the processing has been an ancient method of preservation widely used.
54 This is achieved by either removing water, or binding it such that the food
55 becomes stable to both microbial and chemical deterioration. For this reason much
56 attention has been given to the sorption properties of foods (Al-Muthaseb et al.,
57 2002). Knowledge of the sorption characteristics is essential in regard to stability
58 and acceptability of food products, drying process modelling, design and
59 optimization of drying equipment, calculation of moisture changes which may
60 occur during storage, and for the selection appropriate of packaging materials
61 (Oyelade et al., 2008). The sorption equilibrium is influenced by temperature and
62 water activity (a_w) (Iglesias et al., 1986). The quality of the product on storage is
63 largely depended on the water activity of the product (McMinn and Magee, 1999;
64 Singh and Singh, 1996; Wang and Brennan, 1991) which in turn depends on the
65 moisture content and temperature of storage. Equilibrium moisture content is the
66 minimum moisture content to which foodstuff (i.e. flours) can be dried under a
67 given set of drying conditions. It can be defined as the limiting moisture content
68 approached by a material after it has been exposed to a particular environment for
69 an infinitely long time (Kolor et al., 2006).

70 The relationship between moisture content and water activity of food at constant
71 temperature, it is known as moisture sorption isotherm. Water activity can be
72 defined as the ratio between vapour pressure of water in the food and vapour
73 pressure of pure water at the same temperature. The water activity is one of the
74 main control variables in food preservation technology. Moisture sorption
75 isotherms can be constructed either by an adsorption process (i.e. starting from a_w

76 $\rightarrow 0$ of the solid material) or by a desorption process (i.e., starting from $a_w \rightarrow 1$ of
77 the solid material). In the first case, weight gain (water uptake) is measured in the
78 sample by the water adsorption from atmosphere with determined relative
79 humidity and in the second case is measured a weight loss of the material by the
80 water desorption to the atmosphere.

81 The phenomenon of hysteresis is produced when equilibrium moisture content at
82 a given water activity don't present the same value in both processes (Rouquerol
83 et al., 1999). The typical hysteresis curve shows that water is more held at the
84 same a_w (or lower a_w at a given moisture content) for the desorption curve than for
85 the adsorption curve between closure points at each end of the cycle (Bell and
86 Labuza, 2000).

87 Hysteresis loops can be classified into four types, H1–H4, taking into account the
88 IUPAC classification (Sing et al., 1985). Type H1 is a fairly narrow loop with
89 very steep and nearly parallel adsorption and desorption branches. In contrast, the
90 Type H2 loop is broad with a long and almost flat plateau and a steep desorption
91 branch. Types H3 and H4 do not terminate in a plateau at high water activity and
92 the limiting desorption boundary curve is therefore more difficult to establish. The
93 characteristic features of some types of loop are associated with certain well
94 defined pore structures.

95 Analyses of the sorption phenomena can be undertaken in terms of
96 thermodynamic functions, which for food provide an understanding of water
97 properties and energy requirements associated with the sorption behaviour. The
98 total heat of sorption is the total energy required to transfer water molecules from
99 vapour state to a solid surface or vice-versa. It is useful, for example, in predictive
100 drying models and in the design of drying equipment (Fasina, 2006).

101 Two methods are available for the measurement of the total heat of sorption. The
102 first is the direct calorimetric measurement of the heat evolved, and the second is
103 the application of the Clausius-Clayperon equation on the isosteric equilibrium
104 partial pressures of vapour at different temperatures (the 'isosteric' heat of
105 sorption). Calorimetrical measured heats of sorption are much less common than
106 those calculated from the sorption isotherm (Al-Muhtaseb et al., 2002).

107 Moisture sorption behaviour of chestnut and wheat flour could be valuable
108 information on its drying behaviour and storage quality. However, there is very
109 little information available in the literature on sorption behaviour for chestnut
110 flour. This study was motivated by the interest on the knowledge of the chestnut
111 flour, which has a sugars and lipids composition quite different from wheat flour
112 (usually used).

113 The main aim of this work is to determine the sorption isotherms for chestnut and
114 wheat flours over a range of temperatures and water activities commonly
115 experienced for flours. The specific objectives include the presentation of the
116 influence of temperature on sorption isotherms, the hysteresis loop, the fitting of
117 the adsorption and desorption moisture sorption isotherms using two widely
118 recommended isotherm models and the evaluation the net isosteric heats of
119 sorption at different moisture levels for chestnut and wheat flour.

120

121 **Materials and Methods**

122 Raw Materials

123 Commercial chestnut flour was acquired in a local market (Galicia, Spain). Flour
124 particle size was evaluated by previous sieving of the material (Sacchetti et al.,
125 2004). Size distribution of chestnut flour particles was >250 μm (36.3%), 250-200

126 μm (10.6%), 200-125 μm (21.9%), 125-80 μm (6.0%) and $<80 \mu\text{m}$ (25.2%).
127 Wheat flour was also acquired in a local market. Distribution of particle size for
128 this type of flour was 200-80 μm (30.0%) and $< 80 \mu\text{m}$ (70%).
129 Moisture content (AOAC, 1995) of fresh samples in dry basis was 9.1% for
130 chestnut flour and 12.5% for wheat flour. It was necessary to prepare previously
131 the flours for each sorption experiment. For desorption isotherms, samples were
132 wetted by means of the contact with air saturated of water at 50°C during 2 weeks
133 up to achieve 0.5 kg (kg.d.b.)⁻¹. In the case of samples employed to obtain
134 adsorption isotherms, they were introduced in a vacuum oven at 50°C and
135 absolute pressure of 15 kPa during one week to decrease the moisture content up
136 to 0.03 kg (kg.d.b.)⁻¹.

137

138 Sorption isotherms

139 The equilibrium moisture contents for adsorption and desorption of the chestnut
140 and wheat flours were determined at 20, 35, 50 and 65°C by static gravimetric
141 technique. Samples of 0.5g were kept in desiccators with saturated salt solutions
142 (KOH, LiCl, MgCl₂, K₂CO₃, Mg(NO₃)₂, NH₄NO₃, NaCl, KCl and BaCl₂) stored
143 in the controlled temperature chamber (Greenspan, 1977) in order to generate
144 controlled humidity environments in a range from 0.09 to 0.91. The changes of
145 water activity of the salt solutions due to change in temperature were estimated
146 using the relations reported by Labuza et al. (1985). The chestnut and wheat flours
147 samples were weighed and then placed into desiccators with a little quantity of
148 thymol to avoid the microbial degradation. Samples were periodically weighed
149 until reach constant weight ($\pm 0.0004 \text{ g}$) in an analytical balance (Denver SI-234)
150 during approximately two months. After reaching equilibrium, the moisture

151 content of each sample was determined by drying in a vacuum oven (Heraeus
152 Vacutherm VT 6025) at 70°C and 15 kPa (AOAC, 1995). The final equilibrium
153 moisture content was determined gravimetrically in triplicate.

154

155 Mathematical modelling

156 Many models have been previously proposed to describe the relationship between
157 equilibrium moisture content and water activity (Iglesias and Chirife, 1976).

158 Experimental data of this study was fitted to following models:

159

160 1. Guggenheim, Anderson and de Boer (GAB) (Van den Berg and Bruin, 1981)

161 GAB model is used in this work to describe the experimental sorption isotherms
162 in the full range of water activity (0.09-0.91) for chestnut and wheat flours and it
163 is given by the following equation:

164

$$165 \quad X = \frac{X_M C K a_w}{[(1 - K a_w)(1 - K a_w + C K a_w)]} \quad (1)$$

166

167 The parameters X_M , C and K have physical significance; depend on the
168 characteristics of the product and show Arrheniusian dependence on temperature
169 (Kim and Bhowmik, 1994):

170

$$171 \quad X_M = X_{M0} \exp(H/RT) \quad (2)$$

$$172 \quad C = C_0 \exp[(h_M - h_N)/RT] \quad (3)$$

$$173 \quad K = K_0 \exp[(h_L - h_N)/RT] \quad (4)$$

174

175 where X_M represents moisture content of the monolayer, C is related to sorption
176 heat of the first layer and K is related to the heat of sorption of the multilayer. The
177 average heat of condensation of water vapour is h_L and it is calculated in the range
178 of temperature from 20 to 65°C by the equation (5).

179

$$180 \quad h_L = 45.04 - 0.0438T \quad (5)$$

181 $h_M - h_N$ and $h_L - h_N$ represent strength of the bounding between water molecules in
182 the respective layers. Particularly, $h_M - h_N$ is the difference between monolayer
183 and multilayer sorption enthalpy, and $h_L - h_N$ represents the difference between the
184 water condensation heat and the multilayer sorption heat (Van den Berg et al.,
185 1981).

186

187 2. Chung and Pfof (Chung and Pfof, 1967)

188 This model describes adequately the behaviour of water sorption isotherms of
189 cereals and cereal starches (Ajisegiri, 1990; Boki and Ohno, 1991; Mok and Dick,
190 1991). The Chung-Pfof equation is recommended as a standard to describe the
191 equilibrium moisture content-water activity data of cereals in ASAE Data D245.4
192 (Chen and Morey, 1989).

193 Chung-Pfof model Eq. (6) is employed in this study to express the experimental
194 sorption isotherms for the hysteresis cycle for chestnut flour and for wheat flour in
195 the full range of water activity (0.09-0.91).

196

$$197 \quad X = A + B \ln(-\ln a_w) \quad (6)$$

198

199 The parameters (X_M , C , K , A and B) of the two employed models (GAB and
200 Chung-Pfost) will be estimate by non-linear regression procedure employing
201 Table Curve software (Jandel Scientific).

202

203 Determination of the net isosteric heat of sorption

204 The net isosteric sorption heat (h_e) is defined by the difference between total
205 isosteric sorption heat and condensation heat. Estimation of the net isosteric heat
206 of sorption at constant moisture contents can be made from the slope of the
207 Clausius-Clapeyron equation:

208

$$209 \quad \ln\left(\frac{a_{w2}}{a_{w1}}\right) = \frac{h_e}{R} \left(\frac{1}{T_2} - \frac{1}{T_1}\right) \quad (7)$$

210

211 Statistical analysis

212 The goodness of the employed mathematical models fitting was tested by
213 calculating statistical parameters like coefficient of determination (R^2), mean
214 relative percentage deviation modulus (E) and root mean square error (E_{RMS}):

215

$$216 \quad E = \frac{100}{N} \sum_{i=1}^N \frac{|X_{exp} - X_{cal}|}{X_{exp}} \quad (8)$$

$$217 \quad E_{RMS} = \left[\frac{1}{N} \sum_{i=1}^N (X_{exp} - X_{cal})^2 \right]^{1/2} \quad (9)$$

218

219 **Results and Discussion**

220

221 Figures 1 and 2 show the experimental data of equilibrium moisture content
222 versus water activity (adsorption and desorption isotherms, respectively) for
223 chestnut flour at different temperatures (20, 35, 50 and 65°C). The sorption
224 isotherms of chestnut flour showed a concurrent increase in equilibrium moisture
225 content with increasing water activity at each temperature. This behaviour was
226 manifested in a sigmoidal curve reflecting that the isotherm can be considered like
227 Type II according the BET classification (Brunauer et al., 1940). It was observed
228 that sorption isotherms of chestnut flour were significantly influenced by
229 temperature. This effect must be described in two different ranges in the studied
230 water activity range, below and above 0.65. At water activities values less than
231 0.65, the equilibrium moisture content of all studied systems decreased when
232 temperature increased. The obtained sorption isotherms of chestnut flour crossed
233 over at this water activity value. Vázquez et al. (2001) showed that desorption
234 isotherms for chestnut crossed over at water activity above 0.6 and it was justified
235 by secreted sugar at the surface at high temperatures. Moreira et al. (2009)
236 developed a simplified algorithm based upon chemical composition that it also
237 predicts the aforementioned behaviour for chestnut in function of water activity
238 value for which all sugars (glucose, fructose and sucrose) are solubilised. This
239 simplified algorithm was employed in this work in order to predict the desorption
240 isotherms of chestnut flour at 25°C with satisfactory results ($R^2 > 0.990$ and $E <$
241 0.100) (data not shown).

242 Experimental adsorption and desorption data of chestnut flour were modelled by
243 means of GAB (Eq. 1) and Chung-Pfost (Eq. 6) equations. Tables 1 and 2 show
244 the values of the parameters of GAB and Chung-Pfost models for adsorption and
245 desorption isotherms applied to chestnut flour and the obtained statistical

246 parameters. Using GAB model were obtained values of E (<0.039 and <0.037)
247 and E_{RMS} (<0.0252 and <0.0247) and R^2 (>0.990 and >0.995) for adsorption and
248 desorption isotherms, respectively. Using Chung-Pfost model were obtained
249 values of E (<0.046 and <0.043) and E_{RMS} (<0.0262 and <0.0259) and
250 determination coefficients (>0.980 and >0.986) for adsorption and desorption
251 isotherms, respectively. Taking into account the recommendations reported by
252 Lomauro et al. (1985), GAB and Chung-Pfost models could be applied
253 satisfactory because E values are below 0.10. Nevertheless, GAB model was
254 applied satisfactorily in whole studied range of water activities and temperatures
255 while that Chung-Pfost model showed high deviations at low (<0.12) and high
256 (>0.80) water activities. Taking into account this result and the lower values of E
257 and E_{RMS} it can be concluded that GAB model gives a more adequate fitting.
258 Figures 1 and 2 also show the fittings of experimental data employing GAB
259 model. The values of GAB model parameters, X_M , K and C , changed with
260 temperature. Particularly, the X_M decreased when temperature increased reflecting
261 less hygroscopicity of the flour at higher temperatures. This may be ascribed to a
262 reduction in a total sorption ability of the foodstuff, which may reflect physical
263 and chemical modifications by temperature (McMinn and Magee, 1999). The
264 values of the parameter C decreased and the values of K increased when
265 temperature increased. These trends have been also found for chestnut (Vázquez
266 et al., 2001). Table 3 shows the values of the GAB parameters (Eqs. 2–4) for
267 adsorption and desorption processes. In view of the results, the sorption
268 characteristics are dependent on the direction to achieve the equilibrium moisture
269 content.

270 In Figs. 1 and 2 it can be observed that, irrespectively of temperature, equilibrium
271 moisture content for desorption, at a specific water activity and temperature, is
272 higher than the corresponding value of adsorption. This fact reflects the presence
273 of hysteresis phenomenon. Fig. 3 shows typical hysteresis effect for chestnut flour
274 at 20 and 65°C (in all the studied temperatures the same behaviour was obtained).
275 The hysteresis cycles for chestnut flour can be classified like Type H3 according
276 to IUPAC classification (Sing et al., 1985). These cycles are presented in the
277 range of water activity from 0.12 up to 0.90. Foodstuffs, like chestnut, with Type
278 II sorption isotherms usually exhibit hysteresis cycles of Type H3 (Rouquerol et
279 al., 1999). Type II isotherms are often obtained with aggregates of plate-like
280 particles, which therefore possess non-rigid slit-shaped pores. Because of delayed
281 capillary condensation, multilayer adsorption is able to proceed on the particle
282 surface until a high water activity is reached. Once the condensation has occurred,
283 the state of the adsorbate is changed and desorption curve follows a different path
284 until the condensate becomes unstable at a critical water activity (Rouquerol et al.,
285 1999).

286 Wheat flour isotherms were also experimentally obtained in order to establish a
287 comparison to chestnuts isotherms. Figures 4 and 5 show adsorption and
288 desorption isotherms, respectively, for wheat flour at different temperatures (20,
289 35, 50 and 65°C). The sorption isotherms of wheat flour showed a similar
290 behaviour respect to the variation of the equilibrium moisture content with water
291 activity. These isotherms are also of Type II. In the literature similar isotherms
292 were reported by Hubbard et al. (1957) for wheat adsorption at 35°C and for
293 desorption at 25 and 35°C and by Ferrer et al. (1966) for wheat adsorption at
294 25.5°C. The main difference with the chestnut flour isotherms is that the

295 equilibrium moisture content of all studied systems decreased when temperature
296 increased. This behaviour can be explained in function of low sugars content of
297 wheat flour. Applying to wheat flour the simplified algorithm reported by Moreira
298 et al. (2009) for desorption at 25°C, it could be checked that in this case sugars are
299 absolutely solubilised in all the range of studied water activity. Satisfactory results
300 have also been obtained with the application of this algorithm for wheat flour
301 ($R^2 > 0.990$ and $E < 0.100$) (data not shown).

302 Modelling of experimental adsorption and desorption data of wheat flour were
303 also evaluated by means of GAB (Eq. 1) and Chung-Pfost (Eq. 6) models. Tables
304 1 and 2 show the values of the parameters of GAB and Chung-Pfost models for
305 water adsorption and desorption isotherms applied to wheat flour and the
306 corresponding statistical parameters. With GAB model were obtained values of E
307 (< 0.030) and E_{RMS} (< 0.0242) and R^2 (> 0.998) for adsorption and desorption
308 isotherms. The fitting using the Chung-Pfost model gave values of E (< 0.040 and
309 < 0.026) and E_{RMS} (< 0.0253 and < 0.0243) and R^2 (> 0.990 and > 0.998) for
310 adsorption and desorption isotherms, respectively. Again, according to Lomauro
311 et al. (1985), $E < 0.1$, and it can be concluded that GAB and Chung-Pfost models
312 were applied satisfactorily in the whole studied range of water activities and
313 temperatures. Figures 4 and 5 also show the fittings of experimental data
314 employing GAB model for wheat flour. In this case, the obtained values of the
315 parameters X_M and C (Table 1) had the same behaviour than for chestnut flour.
316 The parameter K for wheat flour decreased when temperature increased. Such
317 trends of the parameters C and K revealed that the binding energies associated
318 with the mono and multilayer sorption of water to the wheat flour decreased when

319 temperature increased. These behaviours were also found for maize flour by
320 (Oyelade et al., 2008).

321 Fig. 6 shows hysteresis effect for wheat flour at 20 and 65°C (in the rest of the
322 studied temperatures the same behaviour was obtained). The hysteresis cycle for
323 wheat flour can be observed at water activities below 0.70. This is the main
324 difference in relation to the hysteresis cycle for chestnut flour (water activity
325 range from 0.12 to 0.90). Figueirido and Ribeiro (1987) reported that hysteresis
326 can be often observed at very low water activity values in systems with the
327 presence of micropores. This phenomenon can be associated with the expansion
328 of little stiff structures, the non-reversible adsorption of molecules with similar
329 size than the pores and the existence of non-reversible chemical adsorption.

330 Net isosteric heats of sorption for chestnut and wheat flours at the mean
331 temperature of the tested temperature (42.5°C) for both adsorption and desorption
332 data are shown in Fig. 7. Heats of adsorption and desorption decreased
333 exponentially when moisture content increased. The high values of h_e at low
334 moisture contents can be explained because the water molecules are tightly
335 bounded to the food, constitute a monolayer, and therefore the amount of energy
336 required to remove these water molecules is very high. At a fixed moisture
337 content, h_e was higher in desorption than the heats involved in adsorption
338 processes in both flours. The values of h_e were approximated to zero at values
339 above 0.13 kg (kg.d.b.)⁻¹ for chestnut flour and above 0.20 kg (kg.d.b.)⁻¹ for wheat
340 flour. Values of h_e decreased from 48 to 1 kJmol⁻¹ for both adsorption and
341 desorption processes when moisture content of chestnut flour increased from 0.03
342 to 0.08 kg (kg.d.b.)⁻¹. The h_e changed from 23 and 36 to 1.5 kJmol⁻¹ for adsorption
343 and desorption processes, respectively, for wheat flour when moisture content

344 increased from 0 to 0.20 kg (kg.d.b.)⁻¹. These trends for adsorption and desorption
345 heats were similar to many cereal grains (Chung and Pfof, 1967). At moisture
346 contents below 0.1 kg (kg.d.b.)⁻¹ the differences between the values of h_e for
347 processes of desorption and adsorption were higher for wheat flour than chestnut
348 flour due to the aforementioned different chemical compositions and porous
349 structure of both flours. The change in the values of the heat of sorption with
350 moisture content regarding to the latent heat of vaporization of pure water,
351 provides valuable data for energy consumption calculations and subsequent
352 design of drying equipment (McMinn and Magee, 2003).

353

354 **Conclusions**

355 Based on the obtained results, it is concluded that the equilibrium moisture
356 content for adsorption and desorption processes of wheat flour decreases as the
357 temperature increases, at the same water activity, following the trend of the most
358 food materials. Water sorption isotherms chestnut flour follows an opposite
359 behaviour at water activities above 0.65. Based on statistical parameters, GAB
360 model is considered more adequate than Chung-Pfof model to fit the
361 experimental data for adsorption and desorption processes of both flours. At the
362 same water activity value, the equilibrium moisture content is higher for
363 desorption than adsorption isotherms and the found hysteresis loops for chestnut
364 and wheat flours are classified like Type H3.

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368

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493 **Captions for Figures**

494

495 Fig. 1. Influence of temperature on adsorption isotherms of chestnut flour. Lines
496 correspond to the GAB model (Eq. 1).

497

498 Fig. 2. Influence of temperature on desorption isotherms of chestnut flour. Lines
499 correspond to the GAB model (Eq. 1).

500

501 Fig. 3. Hysteresis cycles of chestnut flour isotherms at 20 and 65°C. Lines
502 correspond to the GAB model (Eq. 1).

503

504 Fig. 4. Influence of temperature on adsorption isotherms of wheat flour. Lines
505 correspond to the GAB model (Eq. 1).

506

507 Fig. 5. Influence of temperature on desorption isotherms of wheat flour. Lines
508 correspond to the GAB model (Eq. 1).

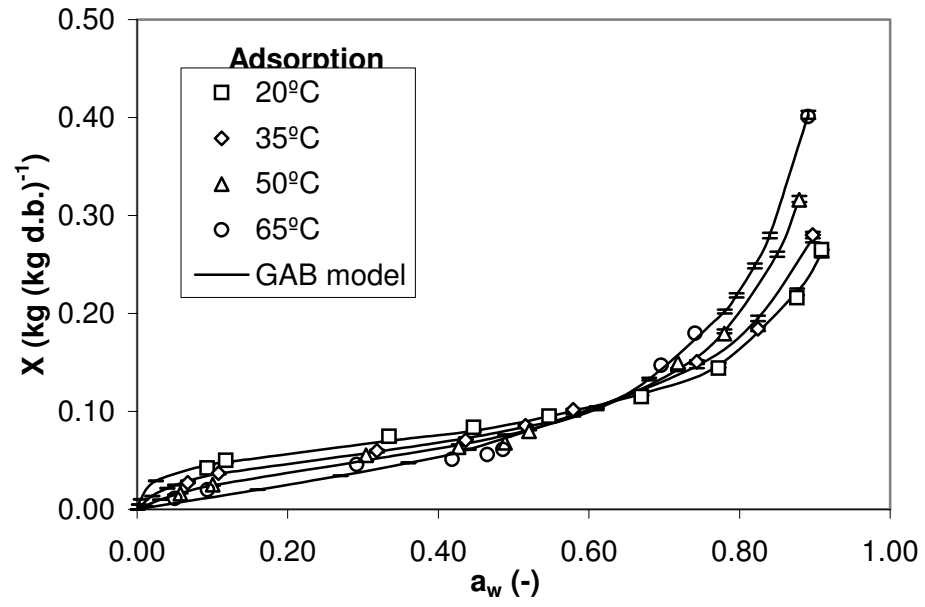
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510 Fig. 6. Hysteresis cycles of wheat flour isotherms at 20 and 65°C. Lines
511 correspond to the GAB model (Eq. 1).

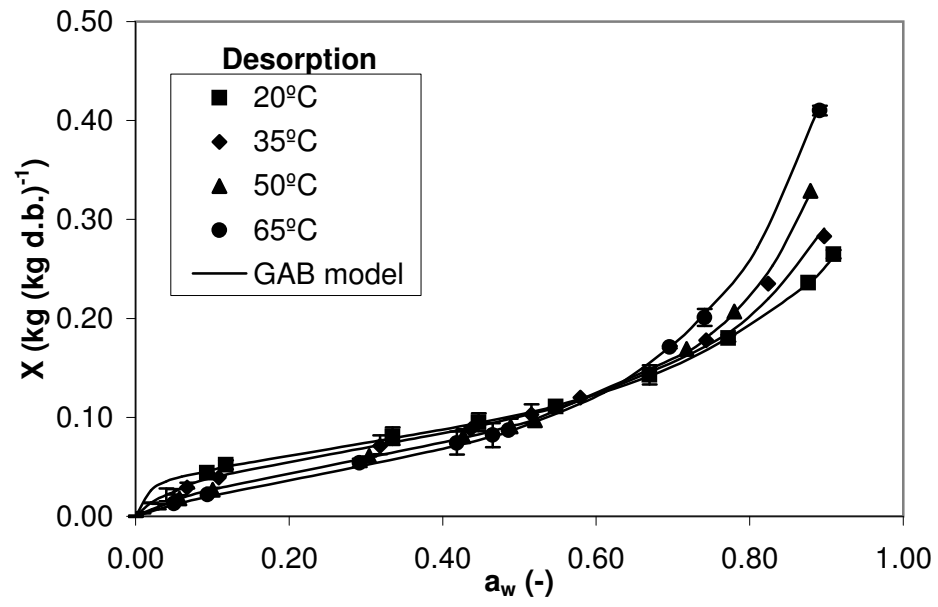
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513 Fig. 7. Effect of moisture content on the net isosteric heat of sorption for
514 adsorption and desorption isotherms of chestnut flour and wheat flour at 42.5°C.

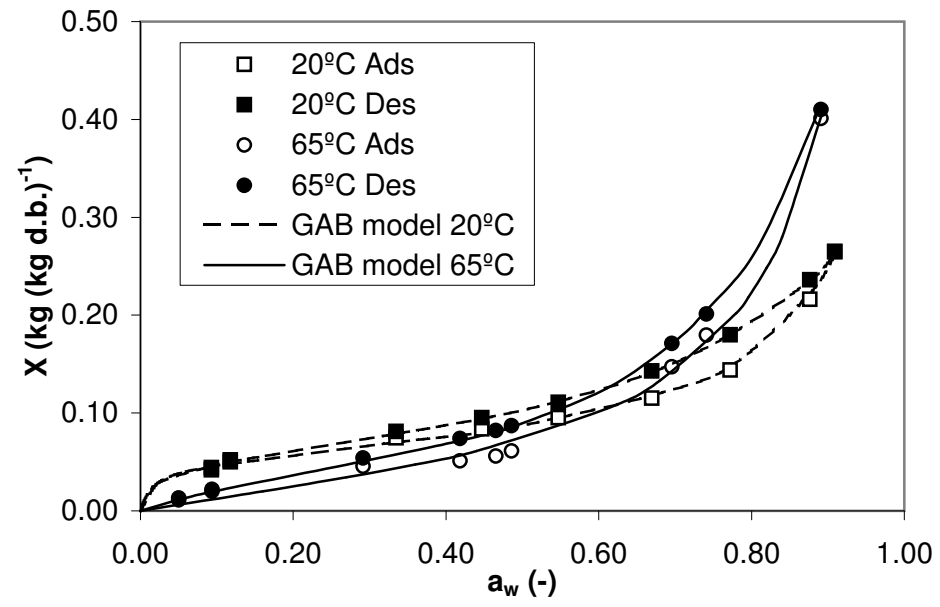
Figure_1



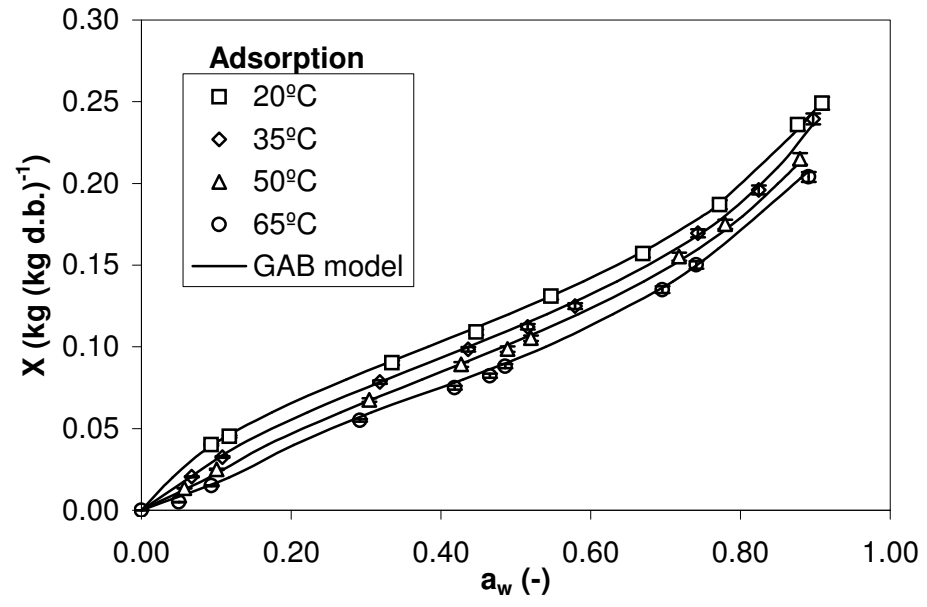
Figure_2



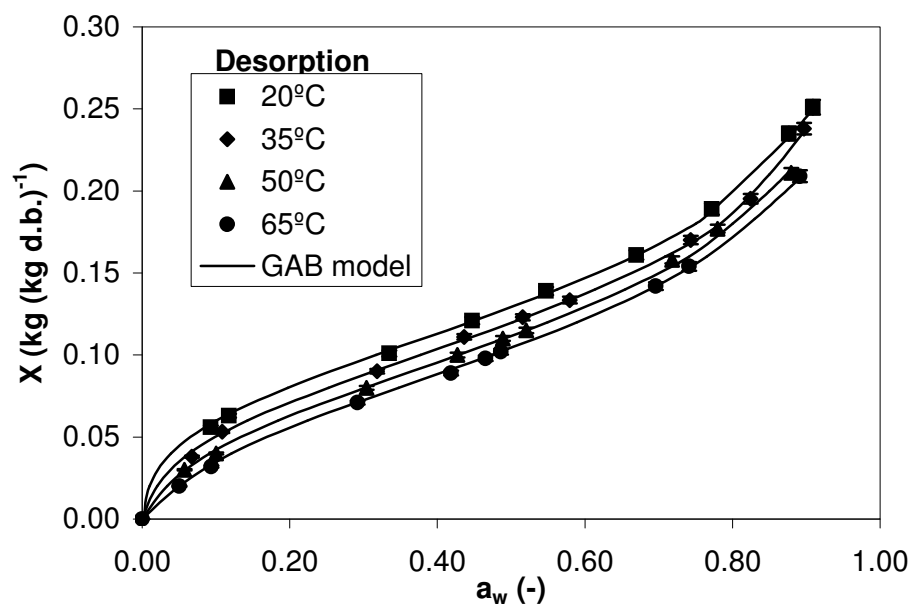
Figure_3



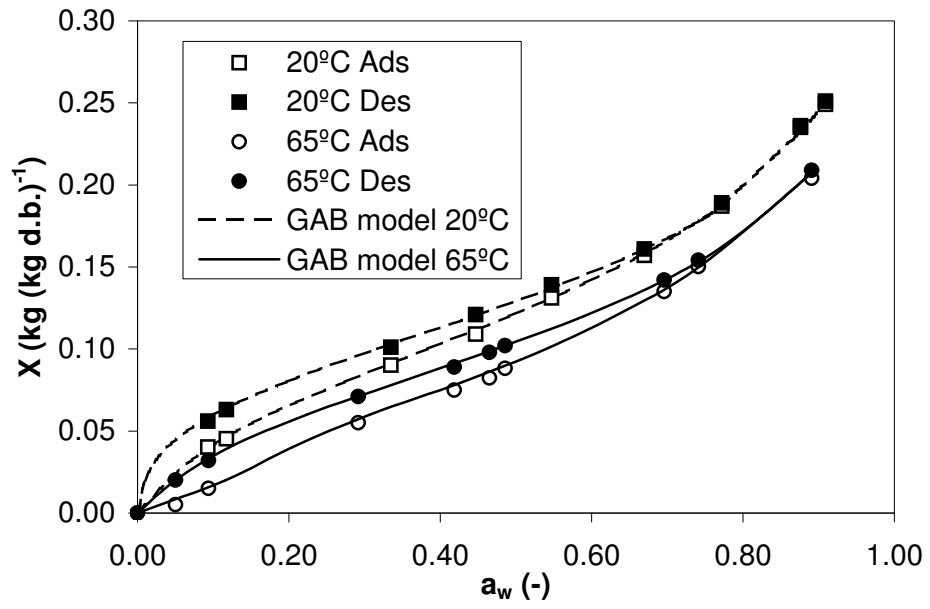
Figure_4



Figure_5



Figure_6



Figure_7

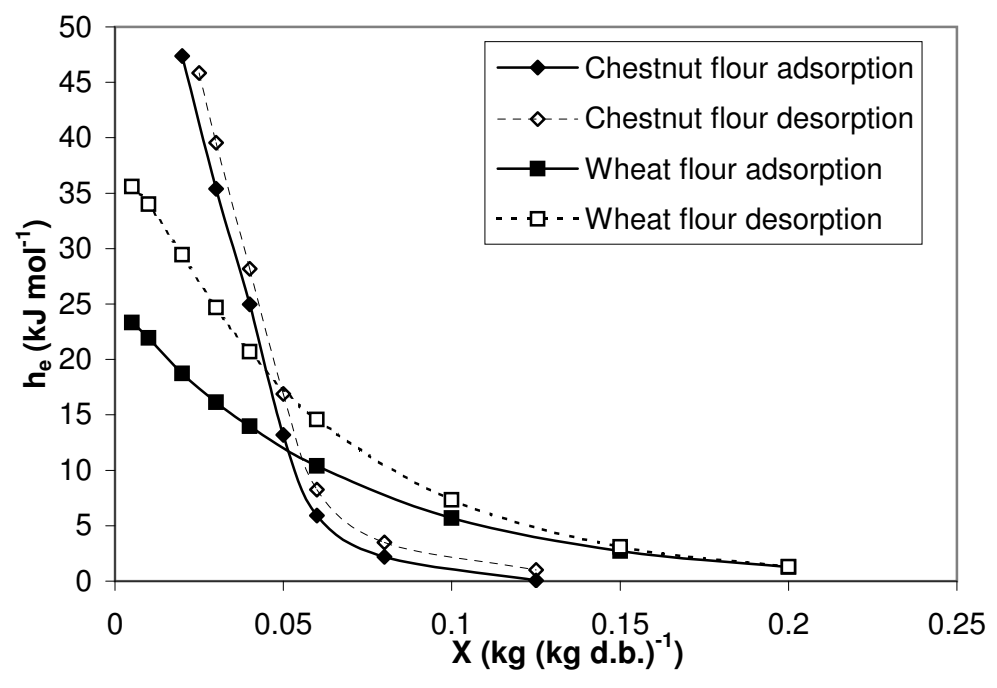


Table 1. Values of GAB model parameters (Eq. 1) and statistical coefficients for adsorption and desorption isotherms of chestnut flour and wheat flour at several temperatures.

Parameters		Chestnut flour				Wheat flour			
Temperature(°C)		20	35	50	65	20	35	50	65
Adsorption	X_M	0.049	0.046	0.044	0.043	0.087	0.086	0.085	0.084
	C	60.5	21.5	9.14	6.01	9.70	6.62	5.27	3.73
	K	0.902	0.931	0.982	0.999	0.751	0.742	0.731	0.720
	R^2	0.990	0.996	0.996	0.995	0.999	0.998	0.999	0.998
	E	0.039	0.035	0.034	0.037	0.024	0.029	0.023	0.030
	E_{RMS}	0.0252	0.0244	0.0243	0.0247	0.0230	0.0241	0.0231	0.0242
Desorption	X_M	0.062	0.061	0.057	0.055	0.089	0.086	0.084	0.080
	C	23.7	14.5	7.27	4.81	20.1	14.6	10.9	9.77
	K	0.859	0.885	0.944	0.978	0.722	0.710	0.709	0.708
	R^2	0.995	0.996	0.999	0.999	0.999	0.998	0.999	0.999
	E	0.037	0.036	0.023	0.024	0.023	0.030	0.023	0.022
	E_{RMS}	0.0247	0.0245	0.0238	0.0239	0.0238	0.0242	0.0237	0.0221

Table 2. Values of parameters of Chung-Pfost model (Eq. 6) and statistical coefficients for adsorption and desorption isotherms of chestnut flour and wheat flour at several temperatures.

Parameters		Chestnut flour				Wheat flour			
Temperature(°C)		20	35	50	65	20	35	50	65
Adsorption	A	2.79	2.29	1.51	1.23	4.26	3.57	3.34	3.02
	B	0.065	0.073	0.099	0.123	0.067	0.068	0.066	0.063
	R ²	0.980	0.987	0.993	0.991	0.999	0.990	0.998	0.997
	E	0.046	0.042	0.038	0.039	0.022	0.040	0.026	0.032
	E _{RMS}	0.0262	0.0258	0.0251	0.0252	0.0239	0.0253	0.0242	0.0246
Desorption	A	3.15	2.12	1.74	1.53	5.85	5.11	4.59	4.13
	B	0.071	0.095	0.104	0.121	0.061	0.061	0.060	0.058
	R ²	0.986	0.989	0.992	0.996	0.999	0.998	0.999	0.999
	E	0.043	0.041	0.038	0.035	0.023	0.026	0.024	0.024
	E _{RMS}	0.0259	0.0256	0.0251	0.0244	0.0240	0.0243	0.0239	0.0241

Table 3. Parameters of GAB model of the Eqs. (2), (3), and (4) in terms of temperature.

		C_0	K_0	X_{M0}	H	(h_M-h_N)	(h_L-h_N)
		(-)	(-)	(kg (kg d.b.) ⁻¹)	(kJmol ⁻¹)	(kJmol ⁻¹)	(kJmol ⁻¹)
Adsorption	Chestnut flour	$1.19 \cdot 10^{-6}$	2.03	$1.80 \cdot 10^{-2}$	2.41	42.9	-1.98
	Wheat flour	$9.01 \cdot 10^{-3}$	0.547	$6.69 \cdot 10^{-2}$	0.641	16.9	0.773
Desorption	Chestnut flour	$1.07 \cdot 10^{-4}$	2.36	$2.41 \cdot 10^{-2}$	2.33	30.1	-2.48
	Wheat flour	$7.27 \cdot 10^{-2}$	0.626	$4.14 \cdot 10^{-2}$	1.87	13.6	0.337