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Journal of Food Engineeringjournal homepage: www.elsevier.com/locate/jfoodeng**Development of a prototype of a colourimetric temperature indicator for monitoring food quality**Vinicius B.V. Maciel ^a, Cristiana M.P. Yoshida ^b, Telma T. Franco ^{a,*}^a UNICAMP – State University of Campinas, School of Chemical Engineering, Av. Albert Einstein, 500, CP 13083-852 Campinas, SP, Brazil^b UNIFESP – Federal University of São Paulo, Department of Exact and Earth Science, Av. Prof. Artur Riedel, 275 Diadema, SP, Brazil**ARTICLE INFO****Article history:**

Received 18 October 2011

Received in revised form 18 January 2012

Accepted 28 January 2012

Available online 8 February 2012

Keywords:

Chitosan

Anthocyanin

Temperature indicator

Coating

Card paper

ABSTRACT

Temperature greatly affects the food quality and safety. Temperature control is an important condition during distribution and storage. An intelligent and biodegradable temperature indicator packaging material was developed based on a natural and heat-sensitive pigment (anthocyanin – ATH). Different concentrations of ATH were incorporated into chitosan matrix films (2.00%, w/w) that were applied as a surface coating on card paper, forming the chitosan card paper system (CH-Sys). The novelty of this work is an alternative packaging material that is biodegradable, simple manufacturing and indicates temperature variations in a specific range by irreversible visual colour changes. The CH-Sys changed irreversibly the colour from light violet to light yellow in response to different temperature exposition (40 °C until 70 °C), independently of luminosity (0 or 1000 lx). The results indicated that the CH-Sys has a great potential for use as a temperature indicator prototype in the specific temperature range studied in this work.

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The application of natural polymer films in the food packaging sector has been investigated due to their biodegradability and ability to retard the transport of moisture, gas, flavour and lipids. Among the natural polymers, chitosan (CH) stands out for forming resistant films with an efficient oxygen barrier ([Yoshida et al., 2009](#)). CH is a partially *N*-deacetylated derivative of chitin, which is commonly found in shells of insects and crustaceans, as well as cell walls of some fungi, and is known as the second most abundant biopolymer in nature after cellulose ([Singh et al., 2009](#)).

Intelligent packaging can be defined as capable of carrying out additional functions (such as detecting, sensing, recording, tracing and communicating) to facilitate consumer decision making, enhance safety, provide information, and warn about problems of quality of the food packed products ([Yam et al., 2005](#)). Examples include biosensors ([Berti et al., 2010; Sankaran et al., 2011](#)), ripeness indicators, time-temperature indicators ([Wanihsuksombat et al., 2010; Mai et al., 2011](#)) and radio frequency identification ([Wang et al., 2010](#)). Colourimetric indicators can be incorporated into a food packaging materials or attached to the inside or outside of a package to detect and monitor changes in the conditions of packed products by visual colour variations ([Ahvenainen, 2003; Brody et al., 2008](#)).

Temperature greatly affects the quality and safety of food and drug products during distribution and storage ([Tsai et al., 2002](#)). In general, appropriate packaging provides effective barrier to gases, moisture and light, but temperature control depends strongly on the transport and storage conditions. [Karel and Lund \(2003\)](#) reported that the control of temperature during transport and storage is an important condition due its effect on growth of microorganisms, metabolic activities and other chemical, sensorial and nutritional reactions. The authors also affirmed that the temperature control on processed food products could be a method to extend the shelf life. According to recommendations United States Department of Agriculture ([USDA, 2011](#)), several microorganisms grow most rapidly in the range of temperatures between 4 and 60 °C, doubling in number in as little as 20 min. This range of temperatures is often called the “Danger Zone”. If the temperature is above 32 °C more than 1 h in these condition the food products should not be consumption ([USDA, 2011](#)). Besides the microbial growth and possible foodborne illness, the high temperature (above room temperature, 21 °C) accelerates physical changes and chemical reactions promoting deterioration on foods ([Karel, 1984](#)). Some nutritional changes may be produced by the food exposition in different environmental factors as high temperatures, moisture, light, water activity and others, causing loss of vitamins (A, C, E), some essential amino acids, essential fatty acids, mineral salts and potential loss protein digestibility ([Karel, 1984](#)). Also can occur change in sensorial characteristics as discolouration of meats, darkening, rancid off flavours, increase in viscosity of oils and loss of volatiles compounds (flavour and aroma) ([Karel, 1984](#)). Furthermore, variations in temperature cause inactivation of active ingredients

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in pharmaceutical products (Shimonini et al., 2001). Hence, it is important to monitor and record the temperature conditions during distribution and storage (Taoukis and Labuza, 2003).

Gregory (1996) reported that vitamins and aroma are affected under temperature above 50 °C and luminosity conditions during storage, transport and commercialization.

The major factors that affect lipid oxidation in food products are the presence of catalysts and antioxidants, light wavelength and intensity, oxygen, pressure, water activity and temperature. This reaction produces aldehydes, ketones, hydroxyacids, and hydrocarbons, which are often volatile, and may have strong odor and off flavor. Increasing temperature promotes higher rates of free radical reactions (Karel, 1984).

Time-temperature indicators or integrators (TTIs) are defined as simple, cost-effective and user-friendly devices to monitor, record, and cumulatively indicate the overall influence of temperature history on the food product quality from the point of manufacture up to the consumer (Giannakourou et al., 2005).

New types of temperature indicator devices have recently been described in the literature. A system based on the growth and metabolic activity of a strain of *Lactobacillus sakei* was developed to monitor quality throughout the chilled-food chain (Vaikousi et al., 2008). Yan et al. (2008) developed a new amylase-type temperature indicator based on the reaction between amylase and starch solution. The changes from blue to yellow indicated that temperature of exposure varied from 4 to 37 °C. Nopwinyuwong et al. (2010) obtained an indicator that employed chemical barcode that responds to carbon dioxide, as a spoilage metabolite, through visible colour change when exposed to temperature change from 10 and 25 °C. Ellouze and Augustin (2010) evaluated a biological temperature indicator as a quality and safety indicator for minced beef and spiced cooked chicken slices packed in modified atmosphere that changes colour with the development of microorganisms.

Anthocyanin (ATH) can be used as natural indicator of temperature variations in intelligent packaging systems due to its characteristics of colour change when submitted at different temperatures. The ATH discolouration was observed in blue flowers (Shaked-Sacharay et al., 2002), vegetables extracts (Bolivar and Cisveros-Zevallos, 2004), pomegranate juice (Alighourchi and Barzegar, 2009) and fruits purees (Patras et al., 2009) after exposure to temperatures higher than 30 °C. The discolouration process is usually accelerated with luminosity (Markakis et al., 1957). Shaked-Sacharay et al. (2002) studied the combined effect of elevated temperatures and metal concentrations on the accumulation of ATH in aster flowers and observed that the anthocyanin degraded and became discoloured at higher temperatures (higher than 30 °C).

The biodegradability characteristic is an advantage of natural polymers in comparison of synthetic polymers. The biodegradability of chitosan films with or without palmitic acid applied as a coating on Kraft paper surface was studied. It was found that the systems were degraded in soil in less than 60 days (Reis et al., 2011).

In this present study, a biodegradable packaging material was obtained to be applied as a temperature indicator prototype based on the ATH incorporation into chitosan matrix. This system was formulated as a coating on a card paper surface, and the temperature indicator response was verified by colour parameters changes and mechanical properties.

2. Material and methods

2.1. Materials

Chitosan (Primex, Iceland; degree of acetylation (DA) of 18% and molecular weight (M_w) of 238 000 g mol⁻¹), acetic acid (Synth,

Brazil), anthocyanin (Christian Hansen, Brazil) and card paper triplex TP 250 g m⁻² (Suzano Papel e Celulose Ltd., Brazil).

2.2. Chitosan card paper system (CH-Sys) preparation

Film suspensions were prepared by dispersing chitosan (CH) in aqueous acetic acid according to Yoshida et al. (2009). The stoichiometric amount of acetic acid was calculated from sample weight, taking into account the value of DA to achieve the protonation of all the NH₂ sites (Notin et al., 2006). Anthocyanin (ATH) was added to the filmogenic suspensions and homogenized by magnetic stirring at room temperature (25 ± 1 °C) for 45 min. Different concentrations of CH and ATH were prepared to apply as coating on card paper (Table 1). The suspensions were spread on the card paper surface (0.045 m²) using an 80 µm wire bar coater (TKB Erichsen, Brazil) forming the CH-Sys. The coated paper sheets were dried in an oven at T = 150 °C for 90 s. In this drying process the purple colour characteristics of ATH was maintained, which at higher ATH concentration was not possible note the colour variation.

2.2.1. Preconditioning

Uncoated and coated card paper sheets were preconditioned at 25 ± 1 °C and 50 ± 2% relative humidity before analysis, in accordance with standard method ASTM D3285 (ASTM, 1999).

2.2.2. Water absorption – Cobb Test

Water absorption capacity was determined in accordance with standard method ASTM D3285 (ASTM, 1999). The weight gain was measured using an analytical balance (AND HR-200 model, Japan). The results are expressed in g m⁻². There were at least 15 replicates per experiment.

2.2.3. Taber stiffness

Taber stiffness was determined using standard method ASTM D5342 (ASTM, 2002). Uncoated and coated card paper sheets were cut into samples of 38 × 70 mm² in machine direction (MD) and the cross-machine direction (CD) using a guillotine (Regmed, Brazil). Taber stiffness was measured at an angle of 15° using Taber stiffness equipment (model RI 5000, Regmed, Brazil). Results were expressed in mN. There were at least 15 replicates per experiment.

2.2.4. Scanning electron microscopy (SEM)

SEM analysis was performed on fractured cross-sections and the surface of gold-sputtered CH-Sys using a LEO 440i scanning electron microscope (LEO Electron Microscopy Ltd., England) under the following conditions: accelerating voltage = 15 kV, distance = 25 mm, current = 200 pA, vacuum = 10⁻⁵ torr (1.3 × 10⁻³ Pa) (Reis et al., 2011). Card paper without coating was studied as reference.

2.2.5. Colourimetric characterization of CH-Sys performance

The colour parameters of CH-Sys were measured during 72 h after different temperature and luminosity conditions exposition. The time was established at 72 h based on preliminary tests. It was observed that the CH-Sys showed efficient colour change during 72 h of exposition and after this the colour parameters maintained similar. A Chroma Meter CR 400 colourimeter (Konica Minolta, Japan) was used. The calibration uses a white calibration

Table 1
CH and ATH concentrations applied as a coating on card paper surface.

Tests	CH (% w/w)	ATH (% w/w)	Total solids (g m ⁻²)
1	2.00	0.25	0.08
2	2.00	0.50	0.09
3	4.00	0.50	0.16

plate. Measurements were performed using the CIE L^* a^* b^* system. The parameter L^* represents the lightness of colours from 0 (dark) to 100 (light), a^* the greenness/redness (negative a^* is green and positive a^* is red) and b^* , the grade of blueness/yellowness (negative b^* is blue and positive b^* is yellow); both move in the two axes that form a plane orthogonal to L^* , and do not have specific numerical limits. Furthermore, the colour parameters most closely related to the psychophysical characteristics of colour, i.e., more related to colour perception. In this case, was calculated the angular coordinates of hue angle [$h_{ab} = \arctan(b^*/a^*)$] were. The parameter h_{ab} refers to the dominant wavelength, starts with 0° and increases counterclockwise (Wyszecki and Stiles, 1982; Sanmartín et al., 2011). The CH-Sys was exposed to a range of temperatures (10–70 °C) and luminosity of 0 and 1000 lx in adapted incubator (Tecnal, model TE-391, Brazil). There were three replicates per experiments.

2.3. Statistical analysis

Statistical analysis was carried out with the Statistic version 7.0 program (Statistic Inc., USA) and differences between the means were detected by the Tukey multiple comparison test.

3. Results and discussion

CH-Sys was characterized by a homogeneous and violet colour system, forming a coated card paper without bubbles or defects. The chitosan-ATH suspension was deposited on the surface, filling the interfibrillar spaces of the cellulose (Fig. 1). The chitosan coated card paper did not present delamination after rigorous handling that could indicate a good compatibility between chitosan film and cellulose fibers. Higher total solid applied as a coating card paper formed a more homogeneous surface. Kato Junior et al. (2011) studying an colourimetric indicator of hydrogen sulfide based on chitosan matrix obtained similar results.

Fernandes et al. (2010) observed that the penetration of chitosan on paper sheets occurred gradually showing saturation after the third layer of polymer suspensions. Rhim et al. (2007) applied poli-lactic acid on cardboard surface and observed higher smooth and homogeneous surface, which was associated with greater filling of the pores fibrous structures of cardboard. Han and Krochta (2001) demonstrated using SEM that the coating with whey protein promoted a more smooth and homogenous paper surface.

Reis et al. (2011) applied chitosan coatings with or without palmitic acid on Kraft paper sheets and found that chitosan filling the cellulose fibers network.

3.1. Characterization of CH-Sys performance

The CH-Sys were exposed to different temperatures (20, 40, 60 °C) without (0 lx) or with luminosity (1000 lx) during 72 h. An irreversible and gradual change in colour of the CH-Sys from light purple to slightly yellow was observed (Fig. 2). Luminosity produced a more pronounced colour change. Rein (2005) found that higher luminosities accelerated ATH discolouration present in berries.

The colour parameters (L^* , a^* , b^*) of the CH-Sys were measured along 72 h (Table 2). Parameter a^* did not change significantly ($p \leq 0.05$) after exposition at different temperature and luminosities conditions.

At 20 °C, the L^* parameter of the CH-Sys remained similar without or with luminosity (Table 2), indicating that the luminosities values applied did not promote change in anthocyanin structure in this temperature condition. At 40 °C and 0 lx, no significantly L^* variation of CH-Sys was observed ranging from 76.35 ± 0.04

(0 h) to 76.95 ± 0.08 (72 h) CIELAB units. However, at 1000 lx independently of temperature studied, the L^* values increased significantly during 72 h of exposition, promoting the discolouration of the CH-Sys. At 60 °C, in the absence or presence of luminosity, CH-Sys presented higher L^* values in function of time exposition.

The parameter b^* , associated from blueness (−) to yellowness (+) alterations, changed significantly ($p \leq 0.05$) in all studied temperatures and luminosities conditions (Table 2). The higher variation in b^* values was observed at 60 °C and 1000 lx, changing from -3.95 ± 0.09 to 1.67 ± 0.21 , indicating the tendency of colour alteration CH-Sys for slightly yellow. The luminosity application accelerated the discolouration process of the proposed system.

Hue angle (h_{ab}) of CH-Sys increased significantly with the exposure time at different temperatures and luminosities. At 0 lx, the temperature of 60 °C resulted in higher h_{ab} values in comparison with other temperatures (Table 2), changing a very slightly bluish reddish hue (301.25°) and a red hue (347.37°). At 1000 lx, it was observed the higher variation on h_{ab} values above 40 °C, with results between a slightly bluish red hue (302.69°) and a tendency to yellow hue (37.00°). The h_{ab} values higher than 360.00° could be explained by the h_{ab} parameter that represents a clear and comprehensive visual colour changes occurred in CH-Sys in both luminosities and temperatures studied. It described the tendency to colour change of natural pigment (ATH) present in CH-Sys.

Δb^* was calculated as absolute values from the difference between the initial and final conditions, for CH-Sys exposed in a range of 10–70 °C and different luminosities. The aim of this analysis was to evaluate different formulations (CH and ATH concentrations) applied as coating card paper. The b^* parameter, showed higher change compared to other colour parameters (L^* and a^*).

The difference Δb^* increased with higher temperatures and luminosities exposition and independently of the concentrations of CH and ATH, the yellowness appearance was observed (Fig. 3).

At low temperatures (10 °C), Δb^* absolute values of the CH-Sys remained similar in presence and absence of luminosity. Anthocyanin structure is stable at refrigerator temperatures (Markakis, 1982). At 0 lx, in the range of 20–50 °C, Δb^* values were practically similar, independently of CH and ATH concentration. The colourimetric transition induced by the ATH degradation was observed in temperatures above 50 °C. There was an exponential increase in Δb^* , with pronounced visual colour change. Increasing CH and ATH concentrations from 2.00% CH/0.25% ATH to 4.00% CH/0.50% ATH, a greatest Δb^* change was observed (Fig. 3a). Increasing the ATH concentration from 0.25% to 0.50% and maintaining 2.00% of CH concentration in the range of 20–50 °C, promoted Δb^* values changed from 0.87 to 1.34 and from 1.49 to 1.82, respectively.

At 1000 lx and 10–20 °C exposition, Δb^* values were similar (Fig. 3b). Increasing the temperature to 20–30 °C, Δb^* increased exponentially in all formulations. The variation of Δb^* can be associated to the colour change of CH-Sys on blue–yellow scale). Increasing CH from 2.00% to 4.00% and keeping constant ATH at 0.50%, did not observed difference between on Δb^* values of formulations during 72 h at different temperatures and luminosities exposition.

Significant changes in b^* values were also found by Alighourchi and Barzegar (2009) for anthocyanins in pasteurised pomegranate juice stored at 4, 20 and 37 °C for 210 days. In their study, L^* , a^* and b^* values decreased and the most significant colour change was observed after storage at 20 and 37 °C. This change was attributed to the degradation of anthocyanin. According to Lauro and Francis (2000), the anthocyanins could change with temperature, light, oxygen, presence of sugars and enzyme, pH and presence of proteins and metal ions (Mazza and Miniati, 1993; Rein, 2005; Wallace and Giusti, 2008; Aramwit et al., 2010), producing polymers with decreased stability.

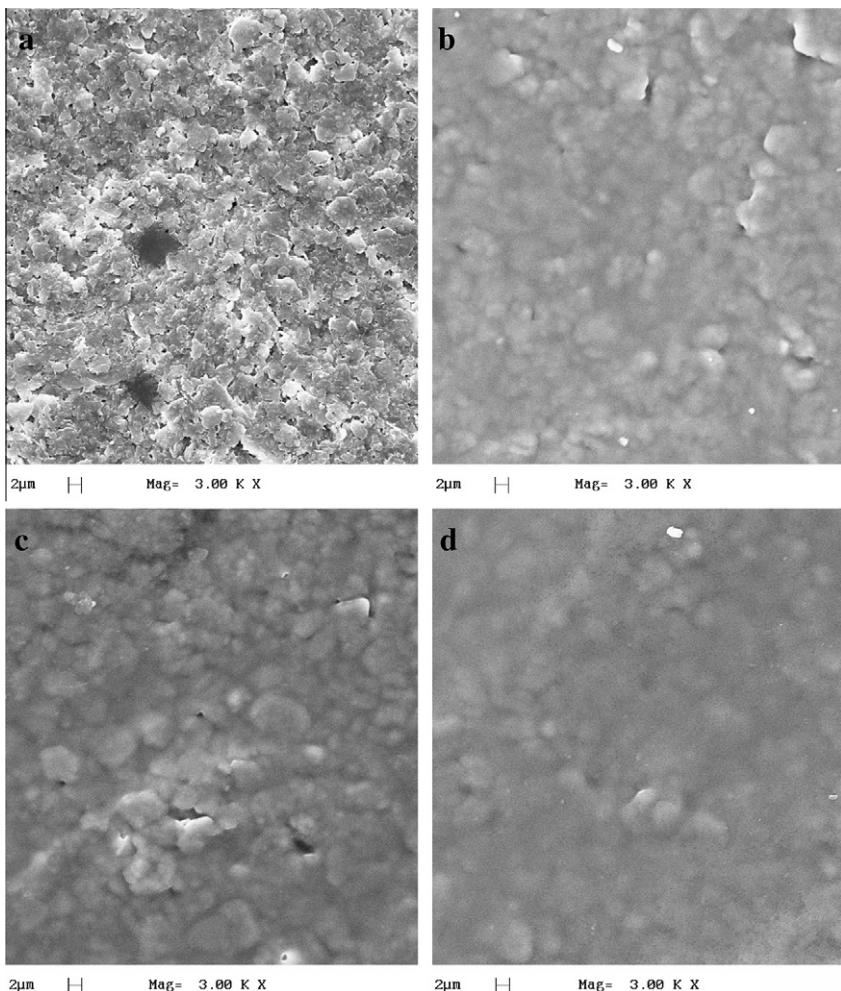


Fig. 1. Micrographs obtained by SEM of the superficial area: (a) card paper without coating, and coated with (b) 2.00% CH/0.25% ATH, (c) 2.00% CH/0.50% ATH; (d) 4.00% CH/0.50% ATH.

Colour variation on the CH-Sys was associated with changes in the chemical structure of ATH molecules. Thermal degradation can produce alteration in ATH structures that depends on the severity and heating conditions. The mechanisms of ATH degradation are still relatively unknown. Chemical structures and the presence of other organic acids have a strong influence (Patras et al., 2010). Markakis et al. (1957) suggested that the pyrylium ring of ATH opens, forming a chalcone structure as a first degradation step. Chalcone is derived from three acetates and one cinnamic acid. It

has a yellow pigmentation and is a precursor of the biosynthesis of flavonoid (Fig. 4). Adams (1973) reported that ATH could decompose upon heating into a chalcone structure and be further transformed into a coumarin glucoside derivative by the loss of the B-ring.

3.2. Properties of CH-Sys

The grammage, water absorption capacity and Taber stiffness results for uncoated and coated card paper were measured (Table 3). These properties could indicate the application form of CH-Sys.

Water resistance is an important property which can determine the behaviour of card paper in various applications. The water absorption of the CH-Sys decreased significantly when applying 2.00% CH and at 0.25% and 0.50% of ATH. Although at 4.00% of CH, the water absorption capacity increased that it could be associated to higher chitosan concentration, which is a hydrophilic polymer and has an affinity for water molecules (Yoshida et al., 2009). Bordenave et al. (2007) studied the potentiality of bioactive food packaging based on chitosan-coated papers and show that chitosan film decreased the water absorption of paper despite the very low amount of total solids. The incorporation of chitosan as a papermaking additive in paper and paperboard production or as a surface coating, had previously been investigated (Kuusipalo et al., 2005; Kjellgren et al., 2006; Bordenave et al., 2007; Fernandes et al., 2009). Different concentrations of chitosan were applied

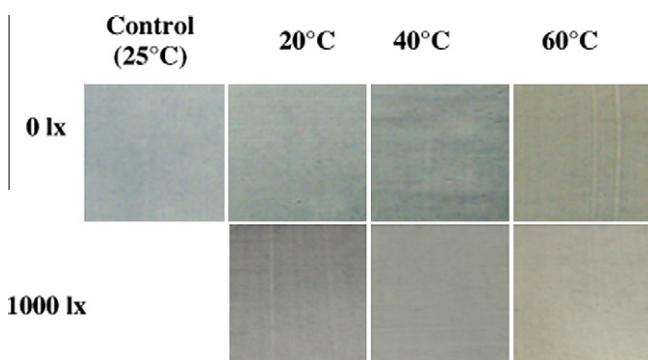


Fig. 2. Visual colour transition of CH-Sys exposed at different temperatures and luminosities after 72 h.

Table 2Values of CIELAB colour parameters (L^* , a^* , b^* , h_{ab}) of CH-Sys exposure at different temperature and luminosities conditions.

Exposure time (h)	0 lx				1000 lx				
	0	24	48	72	0	24	48	72	
20 °C	L^*	76.37 ± 0.07 ^a	76.53 ± 0.30 ^a	76.63 ± 0.24 ^a	76.73 ± 0.36 ^a	76.29 ± 0.09 ^a	76.52 ± 0.08 ^b	76.61 ± 0.06 ^b	76.93 ± 0.02 ^c
	a^*	2.85 ± 0.07 ^a	2.81 ± 0.10 ^a	2.70 ± 0.07 ^a	2.65 ± 0.08 ^a	2.52 ± 0.12 ^a	2.46 ± 0.09 ^a	2.35 ± 0.10 ^a	2.32 ± 0.09 ^a
	b^*	-4.41 ± 0.20 ^a	-3.56 ± 0.20 ^b	-3.26 ± 0.17 ^{bc}	-2.91 ± 0.03 ^c	-4.14 ± 0.29 ^a	-2.89 ± 0.17 ^b	-2.47 ± 0.14 ^{bc}	-2.10 ± 0.12 ^c
	h_{ab}	302.92 ± 1.74 ^a	308.26 ± 2.39 ^b	309.62 ± 1.68 ^b	312.25 ± 1.14 ^b	301.40 ± 1.51 ^a	310.35 ± 0.96 ^b	313.59 ± 0.46 ^b	317.77 ± 1.79 ^c
40 °C	L^*	76.35 ± 0.04 ^a	76.56 ± 0.03 ^b	76.81 ± 0.07 ^c	76.95 ± 0.08 ^c	76.50 ± 0.03 ^a	76.82 ± 0.19 ^a	77.32 ± 0.13 ^b	77.93 ± 0.18 ^c
	a^*	2.82 ± 0.14 ^a	2.73 ± 0.06 ^a	2.65 ± 0.06 ^a	2.60 ± 0.06 ^a	2.61 ± 0.12 ^a	2.52 ± 0.11 ^a	2.46 ± 0.10 ^a	2.34 ± 0.15 ^a
	b^*	-4.37 ± 0.17 ^a	-3.47 ± 0.08 ^b	-3.01 ± 0.02 ^c	-2.38 ± 0.12 ^d	-4.07 ± 0.09 ^a	-1.92 ± 0.31 ^b	-0.73 ± 0.39 ^c	0.07 ± 0.32 ^d
	h_{ab}	302.83 ± 2.08 ^a	308.19 ± 0.72 ^b	311.35 ± 0.45 ^b	317.58 ± 1.04 ^c	302.69 ± 0.68 ^a	322.89 ± 3.41 ^b	343.77 ± 7.72 ^c	5.88 ± 3.81 ^d
60 °C	L^*	76.25 ± 0.04 ^a	76.57 ± 0.03 ^b	76.96 ± 0.04 ^c	77.14 ± 0.06 ^d	76.69 ± 0.28 ^a	77.39 ± 0.32 ^a	78.36 ± 0.36 ^b	79.22 ± 0.20 ^c
	a^*	2.80 ± 0.08 ^a	2.70 ± 0.07 ^a	2.67 ± 0.07 ^a	2.64 ± 0.06 ^a	2.63 ± 0.20 ^a	2.55 ± 0.17 ^a	2.41 ± 0.23 ^a	2.21 ± 0.20 ^a
	b^*	-4.61 ± 0.25 ^a	-2.99 ± 0.27 ^b	-1.50 ± 0.17 ^c	-0.59 ± 0.08 ^d	-3.95 ± 0.09 ^a	-1.43 ± 0.23 ^b	0.08 ± 0.14 ^c	1.67 ± 0.21 ^d
	h_{ab}	301.25 ± 1.24 ^a	312.23 ± 3.02 ^b	330.71 ± 3.32 ^c	347.37 ± 1.83 ^d	303.60 ± 1.36 ^a	330.87 ± 3.86 ^b	3.02 ± 1.14 ^c	37.00 ± 1.40 ^d

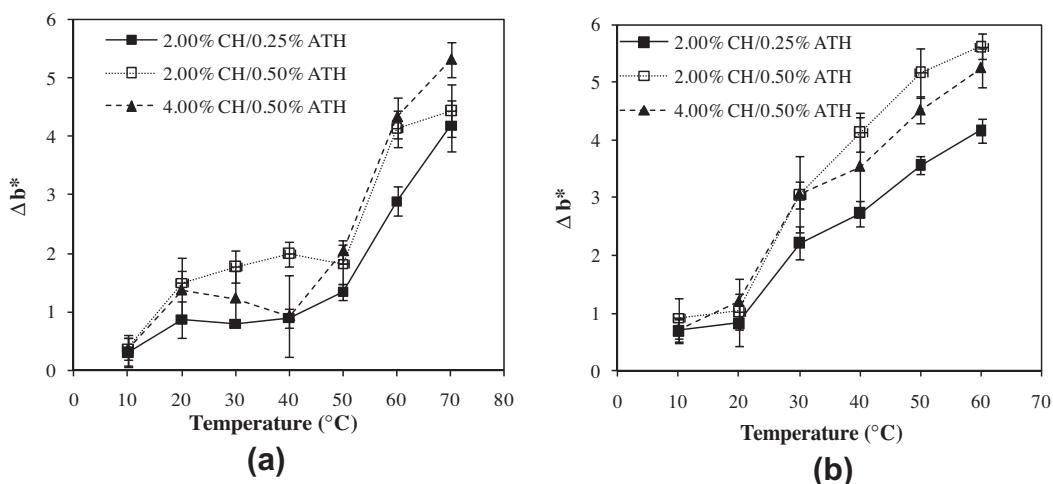
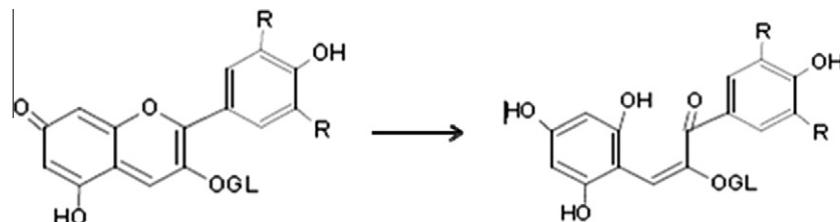
a,b,c,d Means in the same line with different superscripts letters differ significantly ($p \leq 0.05$) in accordance with Tukey's test.Fig. 3. Δb^* of CH-Sys exposed to different temperatures and luminosities: (a) 0 lx and (b) 1000 lx.

Fig. 4. Mechanisms of change in anthocyanin chemical structure after exposure to a high temperature forming chalcones (Markakis, 1982).

Table 3

Grammage, water absorption capacity and Taber stiffness of CH-Sys.

Samples	Grammage (g m^{-2})	Water absorption (g m^{-2})	Taber Stiffness (mN)	
			CD	MD
Uncoated cardpaper	252.71 ± 1.88 ^a	45.48 ± 1.76 ^a	5.60 ± 0.16 ^a	12.97 ± 0.21 ^a
2.00% CH/0.25 % ATH	253.52 ± 1.56 ^a	33.25 ± 1.61 ^b	6.44 ± 0.28 ^b	13.89 ± 0.31 ^b
2.00% CH/0.50 % ATH	253.17 ± 1.71 ^a	35.41 ± 1.66 ^b	6.43 ± 0.17 ^b	14.15 ± 0.09 ^b
4.00% CH/0.50 % ATH	256.39 ± 1.40 ^b	42.14 ± 1.91 ^c	6.29 ± 0.16 ^b	13.98 ± 0.35 ^b

a,b,c Means in the same column with different superscripts differ significantly ($p \leq 0.05$) in accordance with Tukey's test.

as a coating additive in paper and paperboard making, and it was observed that increasing chitosan concentration from 0.10% to 0.75% (w/w), the water absorption decreased significantly

(Kuusipalo et al., 2005). The greaseproof of paper coated with chitosan films did not provide an extra barrier against water absorption (Kjellgren et al., 2006). Reis et al. (2011) applying

chitosan coating with and without palmitic or stearic acid on sheets of *Kraft* paper, observed a reduction in water absorption in order to 35% as compared to uncoated *Kraft* paper.

The CH-Sys did not produce the same reinforcing effect in the MD as in the CD direction. The resistance of the fibres and the rigidity was always higher in the MD. The resistance and flexibility of chitosan films strengthened the cellulose fiber interbonds. Chitosan films had a positive impact on the mechanical properties of coated paper (Fernandes et al., 2010).

4. Conclusions

The visual colour of the CH-Sys irreversible changed from light violet to light yellow after a specific temperature range exposition (40–70 °C). This indicator system has potential applications in different areas that the exposition of temperature in a range of 40–70 °C could promote total or partial degradation, such as food, pharmaceuticals, biological, agricultural, vaccines and other products. The convenience of colourimetric visually detection and simple manufacturing are advantages of this temperature indicator prototype. Also, an efficient and alternative temperature indicator packaging material was obtained based on biodegradable and safe grade materials (chitosan, card paper and ATH). Luminosity accelerated the ATH molecule degradation. Indicators associated with intelligent packaging shows large potential for safe and quality food products.

Acknowledgements

The current study was developed with the support of CNPq, FA-PESP, CAPES and Suzano Papéis e Celulose Ltd.

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