FABRICATION OF GRANULAR HUMIDITY INDICATOR

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Abstract: *This work presents the fabrication of the granular humidity indicator for the monitor of the relative moisture in the outdoor environment or in the enclosed areas such as bags, containers, enclosed rooms, boxes which are used for the storage of the weapons and military equipment, food preservation, seed storage in agriculture, controlling fermentation process as well as the plants growing process, etc. Moisture indicator was fabricated from cobalt chloride, desiccant, color-stabilizing agents, and silica gel particles. The obtained indicator can measure the relative humidity in the range of 30 to 70 based on the color changing from the indicator.*

Keywords: Humidity indicator; Cobalt chloride; Silica gel particles.

1. INTRODUCTION

Vietnam is in the typical tropical climate with high humidity, which significantly affects daily livings. The change in the relative humidity (RH) between day and night in seasons is significant. Currently, several moisture devices have been extensively used to measure the RH in many practical applications. However, these devices are expensive and not convenient for operation in small spaces such as bags, boxes, bins. Thus, it is necessary to find new moisture-measure techniques, which are cheap, quickly responsive, convenient, and smoothly operating with high accuracy. One of these techniques is the granular humidity indicator. Many materials have been employed to fabricate granular indicator. Among these, silica gel has become one of the most well-known materials used for humidity indicators due to its simplicity in recycling by drying method [1, 2]. The surface of silica gel is commonly coated with a cobalt chloride because this salt can change color from blue (dried state, low RH) to pink (high RH) [3, 4]. The color of the cobalt chloride will change from dark blue to light blue to light pink to dark pink. Based on this change in cobalt chloride color, the RH can be determined with the average errors of less than 5% [5-7]. Several methods have been employed to impregnate cobalt chloride on the silica gel. One of the practical approaches is to immerse silicagel in the solution of 45 -50 % w/w cobalt chloride in water at the ambient temperature and pressure. The immersing cycles depend on the requirement of the targeted applications.

2. EXPERIMENTAL SECTION

2.1. Materials

- Equipment: Heaters, flask, drying oven, tủ sấy, thermometer (0-200^{o}C), technical balance, and analytical balance with the accuracy of 10^{-4}. 02 RH-measured devices for the comparison; clear PVC bag was used to create a testing space with a volume of 1 m^3, and colorimeter (USA).

- Chemicals: + $CoCl_2$, PA, Germany;

+ Distilled water;

+ Poliol-based desiccant, Korea;
+ Color-stablized agent S3, China;
+ Anti mold agent BR, Korea;
+ Spherical granular silicagel with average diameter of 2-3.5mm, Russia.

2.2. Experiment

- Mixing of the ingredient for the fabrication of humidity indicator: First, color-stabilized agent S3 was diluted in the distilled water with the $S3/H_2O$ weight ratio of 1/10, respectively. The solution was heated and stirred until the S3 completely dissolved (solution D1). 1% of the anti-mold agent BR and 3% of GR desiccant were gradually added to the solution D1 and stirred at 50 – 60 rpm until obtaining a homogeneous solution (D2). 30 – 55 % w/w of cobalt chloride was gradually added to the solution D2 and stirred at 50 – 60 rpm until obtaining a homogeneous solution. The filtering paper or cloths filtered the undissolved substances in the mix to obtain D3 solution. The preparation steps for the D1, D2, and D3 should be followed because of their non homogeneity of the mixture's chemicals.

- Impregnation of the indicating solution on the silicagel: before the impregnation process, the silicagel was dried at a temperature of 80-90^oC for 4 hours. Then, silicagel was completely impregnated in the solution D3 for 2 hours. The impregnated silicagel was taken out and dried at room temperature for 8 to 10 hours. The product was further dried in the oven at a temperature of 80 – 90^oC for 2 hours, then was cooled down to room temperature before carrying out the next impregnating cycle. The impregnating process was repeated until the uniform color of the granular indicators was obtained. The prepared indicator was stored in the sealed PE bag.

2.3. Properties investigation

The color changes were determined based on observation following the TCVN 2102:2008 [5] with the simulated light source having the irradiating intensity of 1000lx to 4000lx, indicator was coated on the metal surface using a clear adhesive, which does not affect the color of the indicator. The scale from 0 to 5 was utilized to evaluate the change in the color of the indicator. In which 0 is the small color differences and 5 is the high color differences. The color differences are labeled as yellow dominant: ye, green dominant: bl, red dominant: re, pink dominant: pi, blue dominant: bl, etc. However, this approach is only qualitative to determine the indicators' color differences when the RH change on the impregnated silicagel.

Testing plate and standard samples used for the color comparison have the size of 150 mm x 100 mm, which is consistent with TCVN 5670 (ISO 1514).

3. RESULTS AND DISCUSSION

3.1. Optimization of the ingredient concentrations in the indicating mixtures

Obviously, the color intensity and color changing detection of the humidity indicator significantly depend on the cobalt chloride in the mixture (TCVN 2102:2008). To select the appropriate CoCl2 cotent, several concentrations of the salt from 30 to 55% were added into the indicating mixtures with the fix of other

ingredients. The impregnating process was carried out for 4 cycles, with each cycle for 2 hours.

Table 1. *The difference in the color following the change in humidity with various $CoCl_2$ concentrations (TCVN 2102:2008).*

% kl $CoCl_2$	Color differences (DH)			Color differences (scale unit)		
	RH<40%	RH=40-60%	RH>60%	RH<40%	RH=40-60 %	RH>60%
30	DH: 1bl	DH:0 bl	DH: 1pi	1	0	1
35	DH: 2 bl	DH: 0 bl	DH: 2 pi	2	0	2
40	DH: 3 bl	DH: 1 bl	DH: 3 pi	3	1	3
45	DH: 4 bl	DH: 2 bl	DH: 4 pi	4	2	4
50	DH: 5 bl	DH: 3 bl	DH: 5 pi	5	3	5
55	DH: 5 bl	DH: 3 bl	DH: 5 pi	5	3	5

It has been well known that the change in the color of $CoCl_2$ is from blue to pink due to the change of humidity. When the RH < 60%, the $CoCl_2$ is blue and the $CoCl_2$ is pink when the RH > 60%. The pink color of the $CoCl_2$ is ascribed to the formation of $CoCl_2.6H_2O$. However, in the small interval of RH, the color of $CoCl_2$ is also different [6].

$$\text{Dried } CoCl_2 \text{ (blue)} \xrightarrow{\text{(moisture)}} CoCl_2.6H_2O \text{ (pink)}$$

When the percentage of the $CoCl_2$ in the indicating mixture (including $CoCl_2$, desiccant, anti-mold agent, and color-stabilizing agent) increase from 30 to 50%, the color differences increase from 0 to 5, which means that when the concentration of $CoCl_2$ increase, the color of the indicator also increases from light blue (DH:2bl) to dark blue (DH:5bl) with RH < 40%. With the RH from 40 to 60%, the color of $CoCl_2$ changes from no color (DH:0bl) to blue (DH:3bl). And finally, with the RH > 60%, the color of $CoCl_2$ change from light pink (DH:1pi) to dark pink (DH:5pi). When the $CoCl_2$ concentration was further increased, the change in color and color differences are negligible. Thus, the optimized $CoCl_2$ concentration was selected to be from 45 to 50%.

The optimized concentrations of the color-stabilizing agent, antimold agent, and desiccant were selected to be 1, 1, and 3 %w/w, respectively.

3.2. Effect of the impregnation cycles on the property of the indicator

The color differences (DC): more color is assigned as (+), and less color is assigned as (-). The differences in the brightness of the color (DL): more brightness is assigned as (+), and less brightness is assigned as (-). DC and DL of the impregnated silicagel depend on the number of the impregnating cycles. In this work, the several cycle numbers of the impregnation process with the same conditions (same indicating mixtures, impregnating for 2 hours, dried at 80 – 90^{o}C in 4 hours) were carried out. The method to measure the color differences is based on the TCVN 2102:2008 and measured at RH of 40% with the standard color of 3 (light blue).

Table 2. *The differences in color and brightness from the various numbers of the impregnating cycles.*

Impregnating cycles	Color differences (DC)	Brightness differences (DL)
1	DC: -3	DL: +3
2	DC: -2	DL: +2
3	DC: -1	DL: +1
4	DC: 0	DL: 0
5	DC: +1	DL: -1
6	DC: +2	DL: -2

It has been demonstrated that the change in the color of the humidity indicator not only depends on the $CoCl_2$ contents in the mixture but also depends on the $CoCl_2$ contents on the surface of the silicagel, which is controlled by the impregnation cycles. When the impregnation cycles increase from 1 to 6, the DC rises from minus 3 (DC:-3) to positive 2 (DC:+2). The color of the indicator reaches the standard color of 3 after impregnating for 4 cycles. The color of the indicator increases along with the increase of the impregnating cycles. If only considering the accuracy of the indicator at the RH of less than 40%, then the increase in the indicator's is beneficial. However, this will cause misleading with the color of the indicator at the other RHs. On the other hand, the DL brightness decreases from light blue to dark blue, which means that the colors of the indicator increase from DL:+3 to DL:-2 and meet with the standard's brightness after 4 cycles of impregnation. From the aforementioned results, it can be concluded that the 4-cycles of the impregnation is the most suitable to impregnate $CoCl_2$ on the silicagel surface.

3.3. Design of the coloring bars for the humidity indicator

Based on the degree of the color differences, the color difference (DH), DC, and brightness differences DL, a color-indicating table was obtained to determine the relative humidity of the air.

Table 3. *Color-indicating bars vs. RH.*

RH, (%)	Color differences	DH	DC	DL
<30	5	DH:5bl	DC:+3	DL:-3
30	5	DH:5bl	DC:+3	DL:-3
40	4	DH:4bl	DC:+2	DL:-2
50	3	DH:3bl	DC:+1	DL:-1
60	4	DH:4pi	DC:+2	DL:-2
70	5	DH:5pi	DC:+3	DL:-3
>70	5	DH:5pi	DC:+3	DL:-3

The basic to construct the color bar for RH indicator is based on the theory that when the RH is relatively low or high, the color difference is more significant, and the brightness is reduced because of the darker color obtained under high humidity. Thus, the RH < 30% or RH > 70% will result the highest color differences as indicated in TCVN 2102:2008. When the RH increases from 30 to 40%, the indicator has a dark blue color, with DC reduces from DH:5bl to DH:4bl, and the DL gradually increases as the fainting of the light blue color. Further increase of the RH from 40 to 50%, the light blue is fainter, challenging to differentiate in color with the DU of only about DH:3bl. When the RH increases from 50 to 60%, the indicator's blue color starts changing to light pink, in this RH range, the color difference is relatively difficult due to the change of blue to light pink (DH:3bl to DH:4pi). However, at this RH range, DC and DH can not reach DC:0 and DL:0 because the color and brightness remain, in this RH range, the color differences only change from blue to pink. When the RH increases from 60 to 70%, DC gradually increases from DC:+2 to DC:+3; the brightness also reduces due to the darkening of the pink color (DL:-2 to DL:-3). At the RH of higher than 70%, the highest colors and lowest brightness are obtained. It can be concluded that the initial color of the indicator is blue and change to the pink color in the high RH condition, depending on the RH range to differentiate the color change.

4. CONCLUSION

- The humidity indicator was fabricated from 45 – 50% w/w of $CoCl_2$, 1 %w/w color-stabilizing S3 agent, 1 %w/w antimold BR agent, and 3 %w/w GR desiccant;

- The humidity indicator granular was fabricated by impregnating silicagel beads (2 to 3.5 mm) in the indicating mixtures and repeated for 4 impregnating cycles. In each cycle, impregnating time for2 hours, natural drying for 8 to 10 hours, finally dried in the oven at a temperature of 80 to 90^{o}C for 4 hours;

- The moisture-indicating table can operate in the RH range of from 30 to 70% with several parameters such as color differences (from 0 to 5), the color differences (blue-bl and pink-pi), brightness differences;

- Moisture-indicating granular can be utilized to measure humidity distribution (multi-point) and measure humidity in a small space, which is difficult to observe.

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TÓM TẮT

NGHIÊN CỨU CHẾ TẠO CHẤT CHỈ THỊ ẨM DẠNG HẠT

Bài báo đề cập tới việc nghiên cứu chế tạo chất chỉ thị ẩm dạng hạt nhằm xác định định tính phân bố độ ẩm tương đối của môi trường không khí hay trong giới hạn không gian hẹp: túi, thùng chứa, hòm hộp, nhà kín,… phục vụ cho công tác bảo quản niêm cất vũ khí, khí tài quân sự; bảo quản thực phẩm, hạt giống trong nông nghiệp; kiểm soát quá trình lên men, sinh trưởng của cây trồng trong công nghệ phát triển sinh học,… Chất chỉ thị ẩm được chế tạo trên cơ sở clorua côban, chất hút ẩm, chất ổn định màu được mang trên silicagen dạng hạt. Khoảng xác định độ ẩm tương đối RH từ 30 đến 70 %, được phát hiện nhờ sự thay đổi màu sắc của chất chỉ thị ẩm chứa trên chất mang silicagen.

Từ khoá: Chất chỉ thị ẩm; Clorua côban; Silicagen dạng hạt.

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