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Colorimetric Response of pH Indicator Dye Solutions and Labels to Ammonia Gas Exposure for Smart Packaging Applications

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Abstract*— Frozen fishery products in the cold supply chain require fast, accurate, and real-time quality monitoring systems. Ammonia gas (NH_3) generated from protein degradation is a key spoilage indicator in these products. This study evaluates the colorimetric sensitivity of pH indicator dye solutions and colour labels upon exposure to ammonia gas (230–700 ppm) as part of efforts to develop colour-changing smart labels for food quality monitoring. Seven dye systems were screened as solutions: single dyes (BTB, MR, BCP, BPB) and combinations (AB = BTB+MR; CB = BCP+MR; DBA = BPB+MR+BTB). Based on screening results, five label formulations (BCP, MR, CB1 = BCP:MR 2:1, CB2 = BCP:MR 1:1, and BCP+PP = BCP:Phenolphthalein 2:1) were subsequently prepared on filter paper and evaluated. Parameters observed included total colour change (ΔE), individual colour coordinates (L*, a*, b*), and pH change over 60 minutes. Results showed that BCP solution had the highest ΔE value (13.81), while MR solution exhibited the greatest pH increase (up to 6.37). For label applications, MR and CB2 provided the best overall performance, combining ΔE stability (R^2 up to 0.88, MAPE <10%) with appropriate pH responsiveness. The BCP label showed the most rapid and pronounced visual colour change from the initial minutes of exposure. These findings demonstrate that the combination of ΔE sensitivity and pH response constitutes a crucial basis for selecting dyes for ammonia indicator labels in smart packaging systems. This study contributes a systematic, quantitative screening framework for dye selection, integrating colorimetric modelling and pH analysis for the development of NH_3-responsive smart labels.

***Keywords*— Indicator dye, ammonia gas, colorimetric label, ΔE, pH, smart packaging**

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I. INTRODUCTION

Frozen fishery products, such as fresh shrimp, are perishable fast-moving consumer goods (FMCG) with high consumer demand and rapid turnover. Their fragile nature necessitates packaging capable of preventing contamination, inhibiting microbial growth, and maintaining chemical and visual integrity throughout the distribution chain [1,2]. In cold supply chains, temperature control is critical, as fluctuations can accelerate product degradation.

Protein degradation in fishery products due to enzymatic and microbial activity produces basic volatile compounds, particularly ammonia (NH_3) and Total Volatile Basic Nitrogen (TVB-N). These compounds are widely recognised as primary indicators of spoilage in fishery products [3] and serve as the chemical basis for the sensing mechanism employed in this study.

Smart packaging has emerged as an innovative solution for real-time quality monitoring. Technologies such as Time-Temperature Indicators (TTI) and pH-responsive colorimetric systems can provide visual signals of product quality changes without damaging the packaging [4–10]. pH indicator dyes that respond to ammonia-induced alkaline shifts have shown particular promise for NH_3-based freshness mosnitoring. These dyes undergo protonation–deprotonation transitions that produce visible colour changes corresponding to changes in the surrounding chemical environment [7], [11–14].

Previous studies have demonstrated the feasibility of individual pH dyes such as Bromocresol Purple (BCP) and Methyl Red (MR) as NH_3 indicators; however, systematic comparisons of single and combination dye formulations—integrating both colorimetric (ΔE) modelling and pH analysis—remain limited [5,6], [15]. In particular, the trade-off between visual sensitivity (high ΔE) and model predictability (high R^2) has not been quantitatively characterised across a diverse set of dye candidates under controlled ammonia exposure conditions.

This study aims to address this gap by systematically evaluating the sensitivity of pH indicator dye solutions and colour labels to ammonia gas exposure (230–700 ppm). The specific objectives are: (1) to screen seven dye systems (four single dyes and three combinations) for their colorimetric (ΔE) and pH response in solution; (2) to evaluate five indicator label formulations applied to filter paper for their colour-change performance and mathematical model accuracy; and (3) to establish a quantitative basis for selecting optimal dye candidates for ammonia indicator labels in smart packaging applications.

II. Material and Methods

A. Materials

The materials used in this study include four single synthetic pH indicator dyes: Bromothymol Blue (BTB), Methyl Red (MR), Bromocresol Purple (BCP), and Bromophenol Blue (BPB). Three combination dye systems were also prepared: AB (BTB + MR), CB (BCP + MR), and DBA (BPB + MR + BTB). Ethanol (97%) was used as the solvent for dye solutions. Ammonium hydroxide (NH_4OH) solution was used as a simulation material to represent ammonia gas generated during shrimp spoilage.

For the label preparation stage, five formulations were applied to 5×5 cm filter paper: single dyes BCP and MR; combination dyes CB1 (BCP:MR ratio 2:1) and CB2 (BCP:MR ratio 1:1); and a BCP + Phenolphthalein (PP) combination (ratio 2:1). Beeswax and 1-tetradecanol were used as binder and substrate-coating agents to support dye immobilisation on the filter paper.

B. Preparation of Indicator Dye Solutions

Each pH indicator dye (BTB, MR, BCP, BPB) was dissolved individually at a concentration of 0.2 g per 500 mL of 95% ethanol. Combination dye solutions were prepared by mixing appropriate volumes of individual dye solutions to produce AB (BTB + MR), CB (BCP + MR), and DBA (BPB + MR + BTB) systems. All solutions were prepared freshly prior to each experiment.

C. Preparation of Indicator Labels

Based on the screening results from the solution-phase evaluation, dye formulations demonstrating significant colorimetric responses were selected for label preparation. Five formulations were applied to 5×5 cm filter paper using a brushing method: BCP, MR, CB1 (BCP:MR = 2:1), CB2 (BCP:MR = 1:1), and BCP+PP (BCP:Phenolphthalein = 2:1). Phenolphthalein was incorporated in the BCP+PP formulation to assess whether a higher-pH-range indicator could extend the detection window. Prepared labels were stored at 4°C for 7 days prior to testing to simulate cold-chain storage conditions for shrimp packaging.

D. Ammonia Gas Sensitivity Analysis

The ammonia gas sensitivity of dye solutions was tested at a concentration of 230 ppm in sealed airtight jars. NH_4OH solution was allowed to evaporate inside the sealed jar to generate NH_3 vapour. Colour changes in the dye solution were observed at 10-minute intervals from 0 to 60 minutes.

For indicator label sensitivity analysis, 75 µL of NH_4OH solution was dripped onto tissue paper placed at the bottom of an airtight jar. The prepared colour label was placed above a support wire to prevent direct contact with the liquid ammonia solution. Label sensitivity was evaluated over a concentration range of 230–700 ppm for 60 minutes, with measurements taken at 0, 10, 20, 30, 40, 50, and 60 minutes. All experiments were conducted in triplicate.

E. Colour Change Analysis

Colour changes of both solutions and labels were measured using a HunterLab Colorflex EZ spectrophotometer calibrated against a white reference tile. Measurements were expressed in the CIELAB colour space (CIE 1976), comprising L* (lightness, 0 = black, 100 = white), a* (redness/greenness), and b* (yellowness/blueness). Total colour difference was calculated as:

$$\Delta E = \sqrt{[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]} \quad \text{eq (1)}$$

F. Mathematical Modelling

The relationship between ammonia gas exposure time and colour change (ΔE) was modelled using regression equations. Both linear and quadratic polynomial models were evaluated using Microsoft Excel. Model performance was assessed using the coefficient of determination (R^2), Root Mean Square Error (RMSE), and Mean Absolute Percentage Error (MAPE).

G. pH Analysis

The pH of indicator dye solutions was measured directly using a calibrated pH meter at each time point. For label pH analysis, a 2×2 cm portion of exposed label was cut and dissolved in 15 mL of distilled water overnight to allow complete dye dissolution. The pH of the resulting solution was then measured.

H. Statistic Analysis

A two-factor completely randomised design (CRD) was used. For solution testing, the first factor was dye type (7 levels: BTB, MR, BCP, BPB, AB, CB, DBA) and the second factor was exposure time (7 levels: 0, 10, 20, 30, 40, 50, and 60 minutes). For label testing, the first factor was label formulation (5 levels: BCP, MR, CB1, CB2, BCP+PP) and the second factor was exposure time (7 levels, as above). Each treatment combination was replicated three times (n = 3). Analysis of variance (ANOVA) was conducted using IBM SPSS Statistics, followed

by Tukey's Honest Significant Difference (HSD) post hoc test at α = 5% for treatments showing significant effects.

III. RESULTS

I. Colour Coordinates (L, a*, b*) of Dye Solutions*

Changes in the a*, b*, and L* colour coordinates of dye solutions exposed to ammonia gas are presented in **Figures 1, 2**, and **3**, respectively. The a* parameter represents the red-green axis: positive values indicate red, negative values indicate green. CB and BTB dye solutions initially exhibited the highest a* values (strongly red), which decreased sharply after 70–80 minutes of exposure (10–20 minutes code intervals). BCP and MR solutions maintained moderate, relatively stable a* values, while BPB showed the lowest and most stable a* value throughout the test period, reflecting minimal red/green colour character.

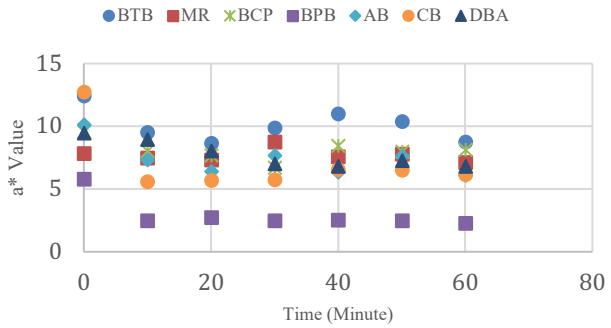

Fig 1. Change in the value of a* over time

The b* parameter indicates yellow-blue tendencies: positive values reflect yellow, negative values indicate blue. BCP solution exhibited a high and fluctuating b* value, consistent with its known yellow-to-purple colour transition. BTB solution showed a moderate b* value. Conversely, BPB and AB solutions exhibited very low b* values (approximately 0.6–2), indicating blue-dominant colour character and limited responsiveness to ammonia exposure.

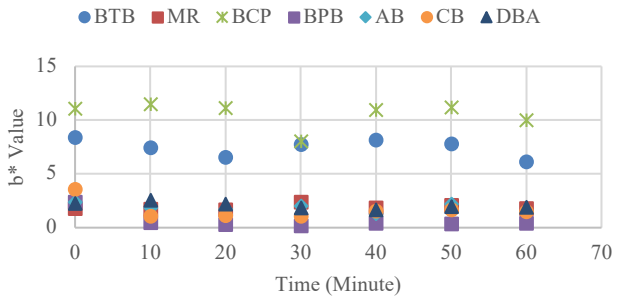

Fig 2. Change in b* value over time

The L* parameter reflects brightness (0 = black, 100 = white). Most dye solutions exhibited a decrease in L* value over time, indicating colour darkening upon reaction with ammonia gas. BCP dye maintained the highest and most stable L* values (10.22–11.94), while BPB and DBA solutions showed relatively low L* values (approximately 5–7), reflecting a tendency towards dark colour from the onset of exposure.

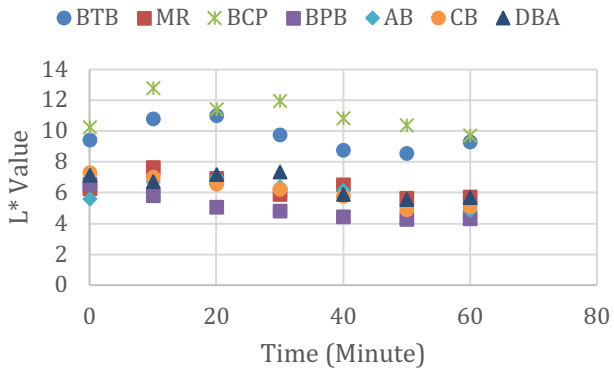

Fig 3. Change in the value of L* over time

J. Total Colour Change (ΔE) of Dye Solutions

Table 1 presents the ΔE values of dye solutions exposed to approximately 230 ppm ammonia gas over 60 minutes. BCP solution exhibited the highest maximum ΔE value (13.81 at 10 minutes), indicating the greatest visual colour change upon ammonia exposure. BTB solution showed consistently high ΔE values (>11 at all time points) and a gradual, stable decrease. MR solution had a moderate ΔE range (8.83–9.85), while BPB showed the lowest values, indicating limited visual sensitivity to ammonia.

Among combination dyes, CB (BCP + MR) showed a relatively high initial ΔE (11.26) that decreased to 8.27 by 60 minutes, reflecting the combined influence of BCP's high initial sensitivity and MR's stabilising effect. DBA (BPB + MR + BTB) followed a similar declining pattern. AB (BTB + MR) showed moderate, gradually declining ΔE values.

TABLE 1
ΔE value of dye solution over time

Time	BTB	MR	BCP	BPB
0	12.66±0.00^{a}	9.26±0.03^{g}	12.66±0.01^{a}	9.01±0.00^{i}
10	12.47±0.01^{a}	9.85±0.01^{c}	13.81±0.01^{a}	8.03±0.02^{n}
20	12.34±0.00^{a}	9.46±0.01^{e}	12.89±0.01^{a}	7.53±0.02^{n}
30	12.05±0.00^{a}	9.38±0.00^{f}	12.71±0.03^{a}	7.32±0.01^{o}
40	11.88±0.01^{a}	9.31±0.00^{g}	12.70±0.01^{a}	7.07±0.00^{o}
50	11.55±0.01^{b}	8.99±0.00^{i}	12.41±0.01^{a}	6.96±0.02^{p}
60	11.36±0.01^{b}	8.83±0.02^{j}	11.93±0.03^{a}	6.94±0.01^{p}

Time	AB	CB	DBA
0	9.71±0.01^{d}	11.26±0.00^{b}	10.08±0.00^{c}
10	9.37±0.01^{f}	9.17±0.01^{h}	9.78±0.01^{d}
20	9.33±0.01^{g}	8.92±0.01^{i}	9.76±0.00^{d}
30	9.28±0.00^{g}	8.75±0.01^{k}	9.61±0.00^{e}
40	8.84±0.00^{j}	8.72±0.02^{k}	8.84±0.04^{j}
50	8.74±0.02^{k}	8.29±0.00^{l}	8.81±0.01^{j}
60	8.15±0.01^{m}	8.27±0.01^{m}	8.74±0.03^{k}

Note: Different superscript letters within the same row indicate significant differences at α = 5%.

K. Mathematical Models for Dye Solutions

Table 2 presents the mathematical models fitted to ΔE data for each dye solution. BTB, AB, and DBA solutions were best

described by linear regression models, reflecting consistent, steady colour changes over time. BTB showed near-perfect fit ($R^2 = 0.99$), confirming highly predictable colour kinetics. MR, BCP, BPB, and CB solutions followed quadratic polynomial patterns, indicating a change in the rate of colour response during exposure. MR's model fit was moderate ($R^2 = 0.75$), while BPB achieved high fit ($R^2 = 0.98$). BCP showed the lowest R^2 (0.63) and moderate MAPE (1.99%), reflecting its inherent high sensitivity and colour fluctuations in a narrow pH range. CB showed the highest MAPE (3.44%), consistent with the combined, non-linear responses of its constituent dyes.

TABLE 2
Mathematical equation for dye solutions

Dye	Mathematical Equation	R^2	RMSE	MAPE (%)
BTB	Regresi Linier	0.99	0.04	0.31
MR	Regresi Polynomial Kuadratik	0.75	0.15	1.23
BCP	Regresi Polynomial Kuadratik	0.63	0.33	1.99
BPB	Regresi Polynomial Kuadratik	0.98	0.10	1.14
AB	Regresi Linier	0.91	0.15	1.34
CB	Regresi Polynomial Kuadratik	0.86	0.37	3.44
DBA	Regresi Linier	0.90	0.16	1.47

Dye	Equation
BTB	$\Delta E = -0.0221t + 12.71$
MR	$\Delta E = -0.0004t^2 + 0.0127t + 9.4335$
BCP	$\Delta E = -0.0006t^2 + 0.0152t + 13.004$
BPB	$\Delta E = 0.0008t^2 - 0.0787t + 8.8873$
AB	$\Delta E = -0.0229t + 9.7485$
CB	$\Delta E = 0.0012t^2 - 0.1083t + 10.801$
DBA	$\Delta E = -0.0246t + 10.111$

L. Colour Coordinates of Indicator Labels

Figures 4, 5, and **6** present the changes in a*, b*, and L* values for indicator colour labels exposed to ammonia gas. For the a* parameter, MR and BCP single-dye labels maintained positive a* values throughout the test period, indicating stable red hue. CB1 and CB2 combination labels experienced a sharp decline in a* values, reflecting a colour shift from red toward green or neutral as pH increased. The BCP+PP combination showed a delayed a* decrease, occurring after 30–40 minutes of exposure.

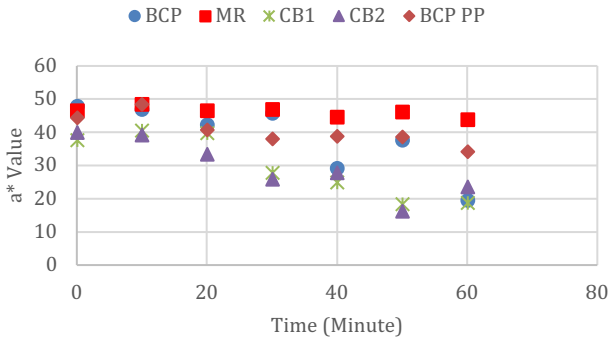

Fig 4. Changes in the value of a* dye label over time

BCP labels showed a high initial b* value that decreased sharply at 30 minutes, consistent with its known colour transition from yellow to purple at elevated pH. CB1 and CB2 labels showed more gradual b* decreases. MR labels showed a slight increase in b* value, reflecting a shift toward orange/yellow, consistent with its colour transition from red to yellow at pH 4.4–6.2.

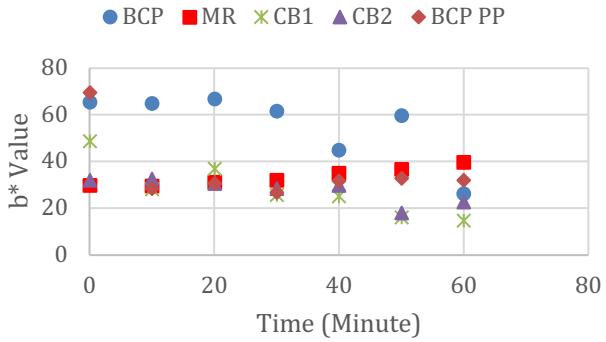

Fig 5. Changes in the b* value of dye labels over time

BCP and BCP+PP labels maintained high L* values initially (indicating bright colours), with decreases occurring between 40–60 minutes. MR labels showed moderate, stable L* values throughout exposure. CB1 and CB2 labels showed more rapid L* decreases, indicating earlier onset of colour darkening.

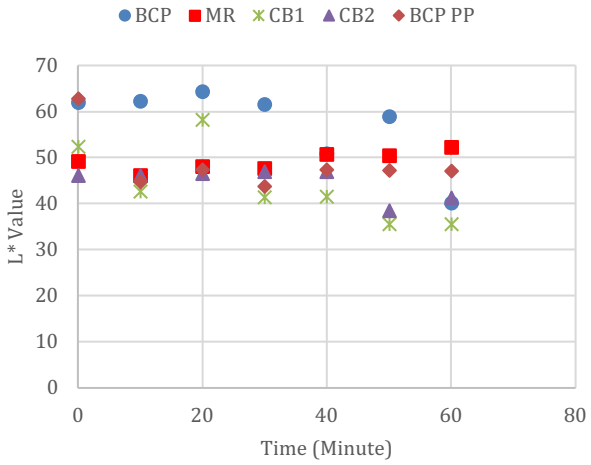

Fig 6. Changes in the L* value of dye labels over time

M. Total Colour Change (ΔE) of Indicator Labels

Table 3 shows the ΔE values for each colour label over 60 minutes. BCP label showed the highest initial ΔE (78.79 ± 1.33) and remained elevated until minute 20, after which it declined sharply to 39.33 ± 3.56 by minute 60. BCP+PP showed a similarly high initial ΔE (77.88 ± 1.75) but with irregular fluctuations, including a marked drop at minute 30 followed by

a partial recovery at minute 40. MR label showed a stable ΔE range (60.80–63.94), indicating minimal but consistent colour change throughout the test. CB1 and CB2 labels showed significant decreases in ΔE over time, with CB1 declining from 61.33 to 33.74 and CB2 showing more moderate initial values but ending at 41.43 by minute 60.

TABLE 3
ΔE values of dye solution over time

Time	BCP	MR	CB1	CB2	BCP PP
0	78.79±1.33^{a}	62.10±2.26^{f}	61.33±1.27^{f}	55.17±1.17^{g}	77.88±1.75^{b}
10	78.37±0.87^{a}	60.80±2.38^{g}	52.34±6.26^{h}	54.76±1.71^{h}	71.81±1.70^{c}
20	77.95±1.12^{b}	61.12±0.42^{f}	62.40±23.74^{f}	51.67±2.75^{h}	78.41±0.50^{a}
30	76.52±0.41^{c}	61.27±1.14^{f}	43.87±7.11^{j}	48.23±4.00^{i}	39.55±4.39^{j}
40	55.30±3.54^{g}	62.56±0.82^{f}	42.48±4.03^{j}	49.19±1.09^{i}	74.99±1.42^{c}
50	68.94±3.38^{d}	63.61±0.28^{e}	34.04±6.74^{k}	35.85±0.72^{k}	35.70±5.59^{k}
60	39.33±3.56^{i}	63.94±1.69^{e}	33.74±2.63^{k}	41.43±0.25^{j}	43.96±9.65^{j}

Note: Different superscript letters within the same row indicate significant differences at α = 5%.

N. Mathematical Models for Indicator Labels

Table 4 presents the mathematical models for ΔE of each colour label. All labels were best described by quadratic polynomial models, reflecting a non-linear colour change pattern. MR label showed the highest model performance (R^2 = 0.88, MAPE = 0.62%), confirming highly stable and predictable colour change. BCP, CB1, and CB2 labels showed acceptable model accuracy (MAPE <10%, R^2 = 0.78–0.82). BCP+PP showed the lowest model performance (R^2 = 0.50, MAPE = 19.55%), reflecting its inconsistent and unpredictable colour changes.

TABLE 4
Mathematical equation of dye labels

Dye	Mathematical equation	R^2	RMSE	MAPE (%)
BCP	Regresi Polynomial Kuadratik	0.78	6.64	8.02
MR	Regresi Polynomial Kuadratik	0.88	0.41	0.62
CB1	Regresi Polynomial Kuadratik	0.82	4.62	7.27
CB2	Regresi Polynomial Kuadratik	0.79	3.01	5.24
BCP PP	Regresi Polynomial Kuadratik	0.50	12.73	19.55

Dye	Equation
BCP	$\Delta E = -0{,}0137t^2 + 0{,}252t + 78{,}156$
MR	$\Delta E = 0{,}0017t^2 - 0{,}0555t + 61{,}687$
CB1	$\Delta E = -0{,}0018t^2 - 0{,}3921t + 61{,}216$
CB2	$\Delta E = -0{,}0015t^2 - 0{,}2022t + 56{,}035$
BCP PP	$\Delta E = -0{,}0007t^2 - 0{,}5923t + 78{,}926$

O. pH Values of Dye Solutions

Table 5 presents the pH values of dye solutions exposed to 230 ppm ammonia gas. MR solution exhibited the highest and most consistent pH increase across all time points, reaching a maximum of 6.37 ± 0.02 at 40 minutes. This is consistent with MR's known pH transition range of 4.4–6.2. AB and CB combination dyes also showed significant pH increases (maximum 6.03 and 6.04, respectively), reflecting the dominant influence of MR in these mixtures. BTB and BCP solutions showed moderate pH increases (approximately 3.0–3.4), while BPB reached up to pH 4.02. DBA showed the slowest and most moderate pH increase (up to 4.18) despite containing MR, suggesting that the presence of BPB and BTB partially suppresses the overall pH response.

TABLE 5
pH values of the dye solution after exposure to ammonia gas

Time	BTB	MR	BCP	BPB
0	2,57±0,00^{j}	5,13±0,06^{c}	2,77±0,01^{j}	3,42±0,06^{i}
10	3,36±0,07^{h}	5,84±0,02^{a}	3,31±0,00^{h}	3,74±0,04^{g}
20	2,93±0,01^{j}	6,30±0,05^{a}	3,38±0,02^{h}	3,95±0,09^{f}
30	3,23±0,08^{i}	6,34±0,03^{a}	3,42±0,00^{h}	4,02±0,02^{f}
40	3,42±0,07^{h}	6,37±0,02^{a}	3,06±0,11^{i}	4,00,±0,03^{f}
50	3,38±0,05^{h}	6,17±0,04^{b}	3,35±0,02^{h}	3,93±0,05^{f}
60	3,29±0,03^{i}	5,86±0,01^{b}	3,23±0,03^{i}	3,74±0,01^{g}

Time	AB	CB	DBA
0	3,56±0,01^{h}	3,53±0,02^{h}	3,33±0,06^{i}
10	5,66±0,01^{b}	5,89±0,01^{a}	3,88±0,07^{f}
20	6,03 ±0,04^{b}	6,04±0,05^{b}	3,68±0,01^{g}
30	5,9±0,02^{b}	5,89±0,04^{b}	3,59±0,06^{g}
40	5,97±0,01^{b}	5,66±0,06^{c}	4,18±0,02^{e}
50	5,7±0,07^{c}	5,67±0,01^{c}	4,08±0,02^{e}
60	5,93±0,03^{b}	5,88±0,01^{b}	4,04±0,05^{e}

Note: Different superscript letters within the same row indicate significant differences at α = 5%.

P. pH Values of Indicator Labels

Table 6 presents the pH values of colour labels exposed to ammonia gas (230–700 ppm). BCP labels showed a gradual pH increase from 4.49 to 4.98, remaining in the acidic range throughout. MR labels were relatively stable (pH 5.23–5.33). CB1 labels increased from 5.18 to 5.46, and CB2 labels from 5.47 to 5.57. BCP+PP labels showed a slight decrease from 5.31 to 5.16, suggesting instability in the immobilised PP component.

TABLE 6
pH values of the dye label after exposure to ammonia gas

Time	BCP	MR	CB1	CB2	BCP PP
0	4.49±0.16^{k}	5.23±0.01^{f}	5.18±0.08^{g}	5.47±0.04^{b}	5.31±0.03^{e}
10	4.67±0.06^{k}	5.25±0.03^{f}	5.32±0.04^{e}	5.47±0.04^{h}	5.09±0.04^{i}
20	4.90±0.04^{i}	5.29±0.06^{f}	5.44±0.04^{c}	5.35±0.08^{d}	5.02±0.06^{i}
30	4.73±0.04^{k}	5.27±0.02^{f}	5.36±0.03^{d}	5.38±0.03^{c}	5.10±0.04^{i}
40	4.86±0.07^{i}	5.27±0.01^{f}	5.42±0.05^{c}	5.35±0.03^{d}	4.96±0.05^{i}
50	4.88±0.05^{i}	5.30±0.03^{f}	5.45±0.08^{b}	5.43±0.03^{b}	5.08±0.02^{i}
60	4.98±0.02^{i}	5.33±0.02^{e}	5.46±0.04^{b}	5.57±0.10^{a}	5.16±0.07^{h}

Note: Different superscript letters within the same row indicate significant differences at α = 5%.

IV. DISCUSSION

Q. Mechanisms of Colour Change: Solution vs Label Performance

The colour change observed in both dye solutions and immobilised labels upon exposure to ammonia gas is primarily driven by the protonation–deprotonation equilibrium of the pH indicator dyes. Ammonia (NH_3), being a basic gas, dissolves in the aqueous phase of the dye system to form ammonium hydroxide (NH_4OH), raising the local pH. This pH increase triggers the deprotonation of the indicator molecule, causing a

shift in its electronic conjugation system and consequently a change in the absorbed wavelengths of visible light—manifesting as a visually detectable colour change [7], [11].

For BCP, the colour transition occurs from yellow (protonated form, pH < 5.2) to purple (deprotonated form, pH > 6.8). The high ΔE values observed in BCP solution (up to 13.81) confirm its strong chromogenic response at relatively low pH changes, attributable to the large difference in molar absorptivity between its protonated and deprotonated forms. For MR, the colour transitions from red to yellow across its pH range of 4.4–6.2, consistent with the gradual pH increases observed (up to 6.37 in solution).

A critical observation is the significant difference in pH and ΔE response between the solution phase and the immobilised label phase. In solution, dyes were freely mobile, allowing rapid equilibration with NH_3 vapour. In contrast, when immobilised on filter paper with binder (beeswax and 1-tetradecanol), dye molecules are physically constrained and their interaction with the surrounding pH environment is mediated by the binder matrix. This physical constraint slows the protonation–deprotonation response, resulting in smaller pH shifts and different ΔE magnitudes in labels compared to solutions [7]. The binder matrix also introduces steric and diffusion effects that alter the apparent sensitivity of the dye. This explains why BCP, which exhibited the highest ΔE in solution (13.81), showed a different kinetic profile in label form—with high initial ΔE but steeper decline—while MR, which showed a moderate solution ΔE, demonstrated superior label stability.

R. Trade-off Between Visual Sensitivity (ΔE) and Model Predictability (R^2)

The central finding of this study is the inherent trade-off between two key performance criteria for ammonia indicator label selection: visual colour change sensitivity (quantified by ΔE) and model predictability (quantified by R^2 and MAPE).

BCP exhibited the highest visual sensitivity in both solution (ΔE up to 13.81) and label (initial ΔE > 78) forms. However, BCP's mathematical model in solution showed low predictability (R^2 = 0.63), reflecting the highly non-linear and concentration-sensitive nature of its colour response. This behaviour is characteristic of indicators with narrow pH transition ranges, where small changes in ammonia concentration can cause disproportionately large colour shifts, making the response difficult to model consistently [7], [11].

In contrast, MR demonstrated a more moderate but highly stable and predictable colour response. In label form, MR achieved the highest model accuracy (R^2 = 0.88, MAPE = 0.62%), indicating that its colour change can be reliably predicted as a function of exposure time. This predictability is highly desirable for Time-Temperature Indicator (TTI) applications, where a defined, mathematically predictable colour change is required to correlate with product shelf life [5], [6].

These contrasting profiles define two distinct indicator application categories: BCP is better suited as an immediate visual alert indicator (rapid, high-contrast colour change from the initial minutes of ammonia exposure), while MR is more appropriate for quantitative monitoring or TTI applications requiring stable, predictable colour change [4], [5]. CB2 (BCP:MR = 1:1) offered an intermediate profile, combining moderate visual sensitivity with acceptable model accuracy (R^2 = 0.79, MAPE = 5.24%), making it a versatile candidate for applications requiring both visual impact and some quantitative monitoring capability.

S. Comparison with Previous Studies

The findings of this study are consistent with the broader literature on pH-based ammonia indicators. Kim et al. [3] reported that BCP-based indicators effectively detected ammonia from fish spoilage, with colour transitions visible at pH values >5.2—consistent with our observation of significant BCP ΔE changes at pH 2.77–3.42 (in solution) and 4.49–4.98 (in label form), suggesting that even sub-transition-range pH changes can generate detectable colour shifts. Luo et al. [7] comprehensively reviewed colorimetric indicators for food freshness monitoring and highlighted the importance of selecting dyes with appropriate pH transition ranges for the target spoilage metabolite. Our results corroborate their conclusion that MR, with its transition range covering the typical spoilage-associated pH region, is a robust candidate for NH_3 indicator applications.

Compared to single-dye systems reported in previous studies, the combination dyes evaluated here (CB1, CB2) offer a potential advantage in broadening the effective pH detection range, as the two constituent dyes (BCP and MR) have overlapping but distinct transition ranges. However, the combination formulations in this study showed less consistent model performance than single-dye MR, suggesting that further optimisation of the mixing ratio and binder composition is warranted [5,6].

T. Novelty and Scientific Contribution

The primary scientific contribution of this study lies in its systematic, quantitative framework for dye screening and selection, integrating two complementary performance metrics—colorimetric sensitivity (ΔE) and mathematical model accuracy (R^2, MAPE, RMSE)—across both solution and immobilised label phases. While individual pH dyes such as BCP and MR have been studied separately for ammonia detection, this study provides a direct, standardised comparative evaluation of seven dye systems under identical experimental conditions, enabling objective trade-off analysis.

Furthermore, the modified method for label preparation (immobilisation on filter paper with beeswax and 1-tetradecanol binder, stored at 4°C for 7 days prior to testing) advances methodological reproducibility for future smart label development studies. The integration of ΔE modelling with pH

analysis in a unified framework provides a more complete characterisation of indicator performance than either parameter alone.

U. Methodological Selection Criteria and Limitations

The transition from seven solution-phase dye systems to five label formulations was guided by the following criteria: (1) $\Delta E > 9$ at any time point in solution, indicating sufficient visual response; (2) ANOVA significance ($p < 0.05$) for both ΔE and pH effects; and (3) practical considerations including dye solubility and compatibility with the filter paper–beeswax substrate. BTB and BPB were not carried forward as single-dye labels because BTB's colour change (yellow to blue) required a wider pH range than was consistently achieved under our test conditions, and BPB showed insufficient ΔE response. DBA was excluded as a label candidate because its three-component system introduced compounding variability.

Phenolphthalein (PP) was incorporated in the BCP+PP formulation as an exploratory candidate to assess whether a higher-pH-range indicator (pH transition 8.2–10.0) could extend detection to higher ammonia concentrations. The results confirmed that PP's transition range was outside the pH window generated by our ammonia exposure conditions, accounting for its inconsistent performance.

A key limitation of this study is that experiments were conducted at a fixed temperature (ambient conditions during testing) without systematic temperature variation. Since real cold-chain applications involve temperature fluctuations, future studies should evaluate the temperature dependence of dye response to ensure indicator stability across the cold-chain temperature range. Additionally, the effect of humidity and competing volatile compounds (e.g., trimethylamine, which also accumulates during shrimp spoilage) on indicator performance should be investigated.

V. CONCLUSION

This study systematically evaluated the colorimetric (ΔE) and pH response of seven pH indicator dye systems in solution and five formulations in label form upon exposure to ammonia gas, providing a quantitative framework for dye selection in smart packaging applications.

In the solution phase, BCP exhibited the highest visual sensitivity (ΔE up to 13.81), while MR showed the greatest pH responsiveness (up to 6.37). BTB, AB, and DBA demonstrated stable, linear colour responses well suited to prediction modelling. In the label phase, MR provided the best overall performance, combining moderate visual sensitivity with superior mathematical model accuracy ($R^2 = 0.88$, MAPE = 0.62%). BCP showed the highest initial visual impact but with declining predictability over time. CB2 offered an intermediate profile suitable for applications requiring both visual responsiveness and moderate predictability.

The central finding of this study is the trade-off between visual sensitivity and model predictability in dye selection: BCP is recommended as an immediate-alert indicator for rapid quality change detection, while MR and CB2 are recommended for stable, predictable ammonia monitoring in TTI-type applications. Future work should investigate temperature-dependent dye response, the effect of competing volatiles, and scale-up feasibility for food packaging integration.

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CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest to disclose.

USE OF ARTIFICIAL INTELLIGENCE (AI) TOOLS STATEMENT

We used Grammarly (Grammarly Inc., 2025) to improve the clarity and grammar of the manuscript. The authors reviewed and approved all changes.

REFERENCES

[1] R. Montanari, "Cold chain tracking: a managerial perspective," *Trends Food Sci. Technol.*, vol. 19, no. 8, pp. 425–431, 2008, doi: 10.1016/j.tifs.2008.03.009.

[2] M. Thirupathi Vasuki, V. Kadirvel, and G. Pejavara Narayana, "Smart packaging—An overview of concepts and applications in various food industries," *Food Bioeng.*, vol. 2, no. 1, pp. 25–41, 2023, doi: 10.1002/fbe2.12038.

[3] D. Y. Kim, S. W. Park, and H. S. Shin, "Fish Freshness Indicator for Sensing Fish Quality during Storage," *Foods*, vol. 12, no. 9, 2023, doi: 10.3390/foods12091801.

[4] Y. W. Hasnedi, "Pengembangan kemasan cerdas (smart packaging) dengan sensor berbahan dasar chitosan-asetat, polivinil alkohol, dan pewarna indikator bromothymol blue sebagai pendeteksi kebusukan fillet ikan nila," Institut Pertanian Bogor, 2009.

[5] E. Warsiki and Riris Octaviasari, "Identification of media and indicator liquid as a recorder smart label," in *International Conference on Adaptive and Intelligent Agroindustry (ICAIA)*, 2015, pp. 169–173. doi: 978-1-4673-7404-0.

[6] E. S. Thamrin, E. Warsiki, and T. Djatna, "Model asosiasi perubahan warna pada indikator kemasan cerdas dan perubahan mutu produk susu," *J. Teknol. Ind. Pertan.*, vol. 27, no. 1, pp. 96–102, 2017, doi: 10.24961/j.tek.ind.pert.2017.27.1.96.

[7] X. Luo, A. Zaitoon, and L. T. Lim, "A review on colorimetric indicators for monitoring product freshness in intelligent food packaging: Indicator dyes, preparation methods, and applications," *Compr. Rev. Food Sci. Food*

Saf., vol. 21, no. 3, pp. 2489–2519, 2022, doi: 10.1111/1541-4337.12942.

[8] N. Shukla, A. K. Tripathi, A. Tiwari, S. Pophaly, K. Sharma, and S. Sahu, "From Farm to Table: Advancements in Smart Packaging for Dairy and Food Products," *Biol. Forum-An Int. J.*, vol. 16, no. 4, p. 33, 2024.

[9] A. R. Bhatlawande, P. U. Ghatge, G. U. Shinde, and R. K. A. Sagar, "Unlocking the future of smart food packaging : biosensors , IoT , and nano materials," *Food Sci. Biotechnol.*, vol. 33, no. 5, pp. 1075–1091, 2024, doi: 10.1007/s10068-023-01486-9.

[10] T. Mkhari, J. O. Adeyemi, and O. A. Fawole, "Recent Advances in the Fabrication of Intelligent Packaging for Food Preservation: A Review," *Processes*, vol. 13, no. 2, pp. 1–49, 2025, doi: 10.3390/pr13020539.

[11] P. H. N. Crosby and A. N. Netravali, "Green Thermochromic Materials : A Brief Review," *Adv. Sustain. Syst.*, vol. 2200208, pp. 1–12, 2022, doi: 10.1002/adsu.202200208.

[12] A. Hakami, S. S. Srinivasan, P. K. Biswas, A. Krishnegowda, and S. L. Wallen, "Review on thermochromic materials : development , characterization , and applications Graphical abstract Enhanced energy efficiency of buildings with smart coatings of microencapsulat, Review on thermochromic materials : development , characterization ," *J. Coatings Technol. Res.*, vol. 19, no. 2, pp. 377–402, 2022, doi: 10.1007/s11998-021-00558-x.

[13] U. Altaf, V. Kanojia, and A. Rouf, "Novel packaging technology for food industry," *J. Pharmacogn. Phytochem.*, vol. 7, no. 1, pp. 1618–1625, 2018, [Online]. Available: http://www.phytojournal.com/archives/2018/vol7issue1/PartV/7-1-328-377.pdf

[14] E. S. Thamrin, K. I. 2022. K. B. P. T. T. L. D. S. P. P. P. I. J. I. dan T. P. 11(4):635–643. Warsiki, EndangThamrin ES, Warsiki E, Bindar Y, Y. Bindar, and I. A. Kartika, "Karakterisasi Bahan Pewarna Tinta Termokromik Leuco Dye System Pada Produk Pempek Ikan," *J. Ilmu dan Teknol. Pangan*, vol. 11, no. 4, pp. 635–643, 2022.

[15] R. Riyanto, I. Hermana, and S. Wibowo, "Karakteristik Plastik Indikator sebagai Tanda Peringatan Dini Tingkat Kesegaran Ikan dalam Kemasan Plastik," *J. Pascapanen dan Bioteknol. Kelaut. dan Perikan.*, vol. 9, no. 2, p. 153, 2014, doi: 10.15578/jpbkp.v9i2.108.