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ORIGINAL PAPER

A paper-based colourimetric sensor for sodium sulfite detection in beverages

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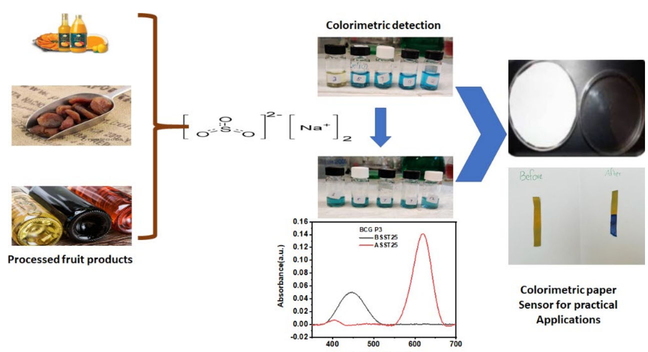
Abstract

Sulfite is a common food additive that prevents oxidation from damaging food nutrients, and it has long been used in the food industry as a bleaching agent. It can harm the human body if taken wrongly or excessively. In this study, three dyes (cresol red, chlorophenol red, and bromocresol green) were explored to analyze the presence of sodium sulfite (SS) in an inexpensive, disposable paper sensor with a lower visible detection limit of 0.05 M. This visual paper sensor detects sodium sulfite with high selectivity and sensitivity at room temperature. An IoT-based sensor was also developed to practically apply the developed method, which is rapid and low-cost and can replace heavy-duty instruments. Both these sensors can substantially impact scenarios such as food quality monitoring and detecting sodium sulfite in medicinal items.

Graphical Abstract



Keywords Sodium sulfite · Toxicity · Food quality monitoring · Colorimetry · Food ageing

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Introduction

Food quality control has grown in importance, and it is now a major issue in our daily lives due to the growing need for high-quality, sanitary food products [1]. Consumer demand for moderately maintained, hardly processed, cooked, and ready-to-eat "fresher" meals and food industry globalization and transportation from processing centers make food quality management a considerable concern [2].

Many additives are used in food to maintain its colour, taste, and other aspects. Sulfite is most typically used to produce sulfur dioxide or inorganic sulfite that can produce sulfur dioxide [3]. Sulfite is a common preservative found in foods and pharmaceuticals. Five sulfites are absorbed and delivered to all organs, including the brain, via the gastrointestinal tube: sodium metabisulfite, sodium hydrogen sulfite, potassium metabisulfite, sodium sulfite, and potassium sulfite. Preservatives containing sodium sulfite are often employed in processed fruit products such as dried fruits, juices, and wine. They are both effective antioxidants that decrease browning reactions and increase ascorbic acid retention in specific fruit products and inhibitors of specific bacteria, making them helpful in the winemaking business. They are used to suppress malolactic bacteria, acetic acid bacteria, and spoilage yeasts and moulds in the juice and juice beverage industries [4].

Clinical studies have shown that sulfite can harm the gastrointestinal system and liver if consumed excessively, causing symptoms such as diarrhoea, dyspnea, nausea, and decreased haemoglobin and red blood cells [5]. According to research, sulfite is a major factor associated with the development and progression of liver disease. Sulfite oxidase activity is high in the liver, kidney, and heart. The liver is the organ most prone to drug-induced toxicity, most likely since the primary metabolic site for most medicines. According to international food Standards, additional sulfites must be mentioned on the label of a packaged food if they are present in concentrations of 10 mg/kg or above. This enables customers to avoid sulfites that are sensitive to them.

Established techniques for detecting physical, chemical, microbiological, and human sensory evaluations of food freshness have proven unsatisfactory in this environment. They must be updated, if not replaced, by rapid, low-cost, non-destructive approaches [6–9].

Colourimetric sensors are very appealing since they can change colour due to a response to volatile chemicals released in packaged food items. They are the easiest, most practical, instrument-free technique of freshness monitoring directly by the human eye, whether directly on the packaging or with an on-package sticker. Colourimetric sensors detect colour shifts generated by intermolecular interactions between chromophores and analytes [10–13]. Colourimetric sensing methods, such as paper-based sensors, on the other hand, are easy to use and produce results fast, requiring no specific training to explain them [14]. Jalal et al. developed sensitive and simple colourimetric methods for visual detection and quantitative determination of semicarbazide in flour products using colourimetric reagents [15]. This method can be either employed as sensors for detecting a trace amount of SEM as low as 10 and 0.5 ppm by bare eye observation within 4 – 2 min for 1st and 2nd methods, respectively or as determination methods UV–vis spectrophotometry in the range from 8 to 180 ppm and 0.5–30 ppm for the 1st and 2nd proposed methods respectively. Mohamed et al. developed a method for the colourimetric detection of volatile organic compounds for the shelf-life monitoring of milk [16]. Colour changes correlated with bacterial growth and associated production of aldehydes and ketones, making this sensor useful for real-time monitoring of milk spoilage and potentially for monitoring shelf life for other high protein and higher fat foods.

Furthermore, the colourimetric response is well-suited to interpret rapid detection, especially in resource-constrained settings or underdeveloped countries [17]. Filter paper has become increasingly useful in developing sensors in recent years due to its wicking capacity [18]. The Whatman® cellulose series is especially popular, with the filter paper types distinguished by porosity, particle retention, and flow rate. Many groups used Whatman® filter paper No. 1, a standard-grade filter paper with medium retention and flow rate, in their research [19]. A cost-effective and sensitive IoT (Internet of Things) colourimetric sensor prototype was developed to implement these dyes systems for practical and real-time application. Using dyes as sensing elements, the prototype exhibits distinct RGB (Red, Green, Blue) values when exposed to test solutions, with a response time of only 2 s.

This research provides a simple and speedy colourimetric detection for dynamic identification, which is provided via an integrated method that relies on dye systems of sodium sulfite and a unique mechanistic understanding.

Materials and methods

Chemicals and equipment

Sodium sulfite anhydrous was purchased from BDH Ltd. Pool England. Bromocresol green was purchased from Sigma Aldrich along with chlorophenol red, cresol red, sodium nitrite, potassium bisulfite, monosodium glutamate, and sorbic acid. The tests were carried out using double distilled water supplied by the Millipore Milli-Q instrument. The reagents employed in the investigation were all of the analytical quality. The characterization was done using a UV-Vis spectrophotometer (Biochrom Ltd, Cambridge, United Kingdom) with a 300–750 nm scanning range.

Arudino UNO, Adafruit LCD shield (both from Adafruit Industries, New York, NY, USA), LEDs (peak around 606 nm), resistors, TSL230R light-to-frequency sensor, protoboard, conductors (Cat 5 cable), and Black ABS or PLA filament were used to make the sensors. To protect sensing elements, a QIDI 3D printer (Zhejiang QIDI Technology Co., Ltd., Ruian, China) was used to print the case.

Methods

Preparation of reagents and analysis

0.003 M cresol red, chlorophenol red, and bromocresol green solutions were made and utilized throughout the experiment. The effect of pH of dye solutions was investigated in acidic (3, 5), neutral, and basic (9, 12) mediums. All pH adjusted, 10 mL of dye solution was treated with 1 mL of 0.1 M SS solution. Colour changes and matching response times were noted. The concentration and temperature effects of these test solutions in dyes were examined using visible colour change at the detected pH. The dyes' detection limits were explored using 0.05-1 M concentrations of SS. The detection limit of an individual analytical procedure is the lowest amount of analyte in a sample which can be detected but not necessarily quantitated as an exact value. Several approaches for determining the detection limit are possible. Based on the Standard Deviation of the Response and the Slope, LOD can be calculated as.

$$LOD = 3\sigma/m \tag{1}$$

Where m is the calibration plot's slope, and σ is the intercept's standard deviation [16, 20–23].

The effect of the temperature was investigated at 25 °C, 40 °C, 60 °C, 75 °C, and 90 °C and the selectivity of dye solutions to sodium sulfite was evaluated by interferences assessment exposing dye solutions to several food additives, including sodium nitrites (SN), potassium bisulfite (PBS), monosodium glutamate (MSG) and sorbic acid (SA) at a concentration of 1 M for all test analytes.

The selectivity was validated by measuring the relative change in wavelength (Δλ) from UV-Vis analysis, as calculated by Eq. 2 (see below).

$$\Delta\lambda = (\lambda_X - \lambda_0)/\lambda_0 \times 100 \tag{2}$$

λ_X represents the precise wavelength of maximum absorption in the presence of the analyte, and λ_0 represents the wavelength of peak absorbance in the absence of the analyte [20–22].

Whatman Paper Functionalization for SS Detection

Colourimetry detected sodium sulfite irreversibly by coating Whatman filter paper with 0.003 M dye at room temperature. The filter paper was kept in the pH 3 dye solution on a petri plate for 1 h. Afterwards, it was dried overnight under ambient conditions. The various notations used are shown in Table 1.

Table 1 List of notations

No.	Description	Notations
1	Cresol red	CR
2	Chlorophenol red	CPR
3	Bromocresol green	BCG
4	Sodium sulfite (molar)	SS(z)
5	Before adding analyte	B
6	After adding analyte	A
7	pH (specific pH)	P(x)
8	Temperature (°C),	T(y)

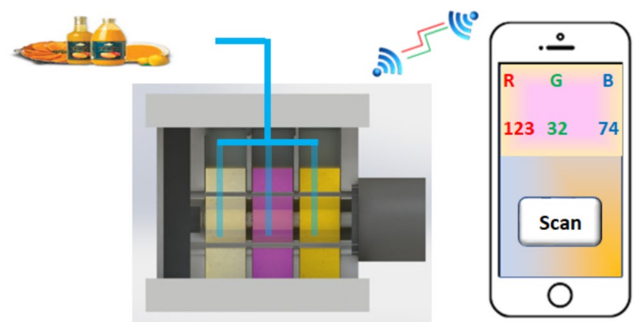

Fig. 1 Schematic of developed sensor prototype

Fabrication of sensor prototype

Figure 1 shows a schematic representation of a sensitive IoT-based colourimetric sensor prototype developed to apply this novel technique. For the setup, three cuvettes were connected in series, and three different dye solutions were used as sensing elements in these cuvettes. A vital light source with four lights (red, blue, white, and green) was placed at one end, and at the other end, a colour detector was placed. An external switch was used to control the illumination of the light source by an Arduino nano controller [20, 22].

Results and discussion

pH Effect

The pH effect of 0.1 M SS at room temperature was investigated on dye solutions in acidic, basic, and neutral mediums. Data were collected before and after the test solutions were added to the dyes at various pH levels in each dye. All dyes successfully provided visible colour changes in a fraction of a second [24].

In the case of bromocresol green, a visible colour change was observed in pH 3 after adding 0.1 M sodium sulfite (Fig. 2a). The colour changed from greenish yellow to light blue. The appearance of a new absorption band centered at

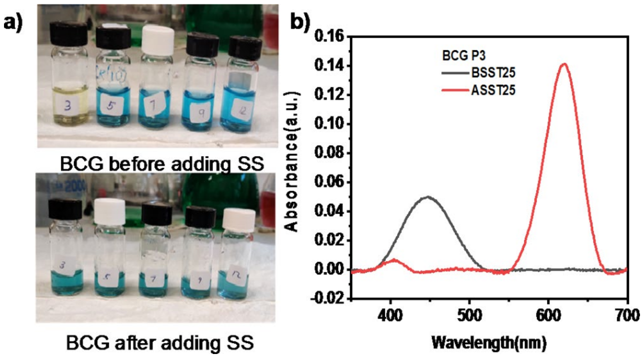

Fig. 2 Detection of SS in BCG (**a**) BCG dye solution during analysis. (**b**) Absorbance spectra of pH 3 dye solution

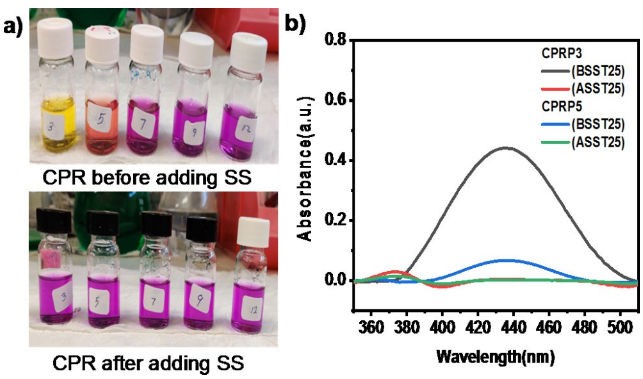

Fig. 3 Detection of SS in CPR (**a**) CPR dye during analysis (**b**) Absorbance spectra of pH3 and pH5 dye solution

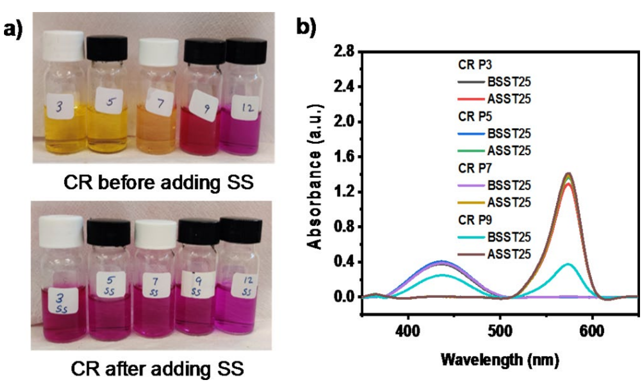

Fig. 4 Detection of SS in CR (**a**) CR dye solution during analysis. (**b**) Absorbance spectra of pH 3, 5, 7, and 9 dye solution

620 nm from 448 nm in UV-Vis absorbance spectra incorporated this colour change. (Fig. 2b).

Similarly, for chlorophenol red, a visible colour change was noticed in pH 3 and pH 5 solutions after adding 0.1 M sodium sulfite (Fig. 3a). The colour changed from yellow to magenta in the pH 3 solution and red to magenta in the pH 5 solution. In UV-Vis absorbance spectra, it was confirmed by the appearance of a new absorption band centered at 372 nm from 435 nm for the pH 3 solution and a change in peak from 430 nm to 375 nm for the pH 5 solution. (Fig. 3b).

After adding 0.1 M sodium sulfite to pH 3, pH 5, pH 7, and pH 9 solutions of cresol red, a perceptible colour change was observed. (Fig. 4a). The colour shifted from yellow to magenta in pH 3, 5, and 7, whereas in pH 9, the colour changed from red to magenta. It was confirmed by a new absorption band centered at 574 nm from 434 nm for pH 3,5 and 7 solutions and a shift in peak position from 436 nm to 365 nm for pH 9 solution in UV-Vis absorbance spectra. (Fig. 4b).

Concentration effect

The UV spectrometric analysis was carried out to examine the dyes' sensitivity to the test solution by adjusting the test analyte concentration from 0.05 to 1 M in particular reactive pH of the dyes at ambient temperature [23]. Analysis was carried out in pH 3 solutions for all dye solutions (Fig. 5a, c and e). The dye solutions successfully provided a visible colour change in the concentration range of 0.05-1 M of sodium sulfite solution. Figure 5b and d, and 5f demonstrate the matching calibration plot for estimating the dye limit of detection (LOD) for SS sensing. The calibration curve was created by taking the peak absorbance of each dye at various concentrations of specific test analytes into account. BCG had an absorbance peak at 624 nm, CPR at 575 nm, and CR at 573 nm. Linear fitting from 0.05 M to 1 M was used to determine the LOD using the Eq. (1).

The linear fitting estimated a LOD of BCG as ~0.021 M, $R^2=0.958$. Similarly, the L.O.D. of C.P.R. was ~0.018 M with $R^2=0.97$. In CR, the predicted LOD for various sodium sulfite concentrations ranging from 0.05 to 1 M was 0.014 M, $R^2=0.98$.

According to the sensitivity investigation, all of these dyes have a high sensitivity to SS, with a linear detection limit as low as 0.014 M in the indicated concentration range. This concentration investigation revealed that these dyes are extremely sensitive to SS, even at low concentrations. Figure 5a shows that absorbance increases when concentration increases according to the Lambert-Beer rule.

Temperature effect

Smart monitoring, management, and control of interior settings is one potential sensor application area. A series of tests under various temperatures in this study using common sources and simultaneous measurements were performed [21].

To evaluate the effect of temperature on the sensing environment, measurement was carried out at five different temperatures 25 °C, 40 °C, 60 °C, 75 °C, and 90 °C. No statistically significant correlation between temperature and low-cost sensor output was found for any sensor evaluated

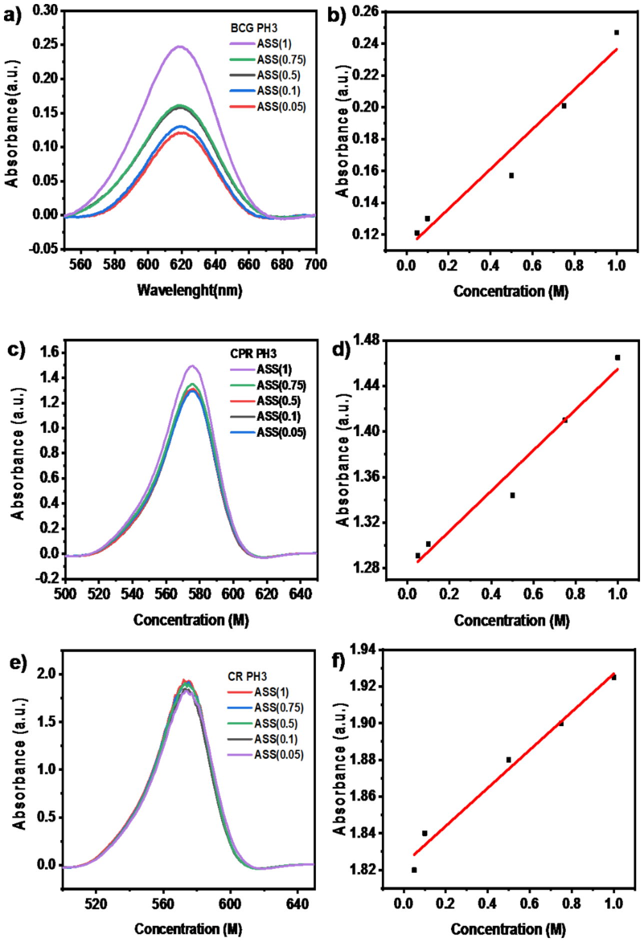

Fig. 5 Dyes sensitivity towards SS detection. (**a**, **c**, **e**) UV-Vis absorption curve with change in SS concentration (**b**, **d**, **f**) calibration plot of BCG dye, CPR dye, and CR dye, respectively

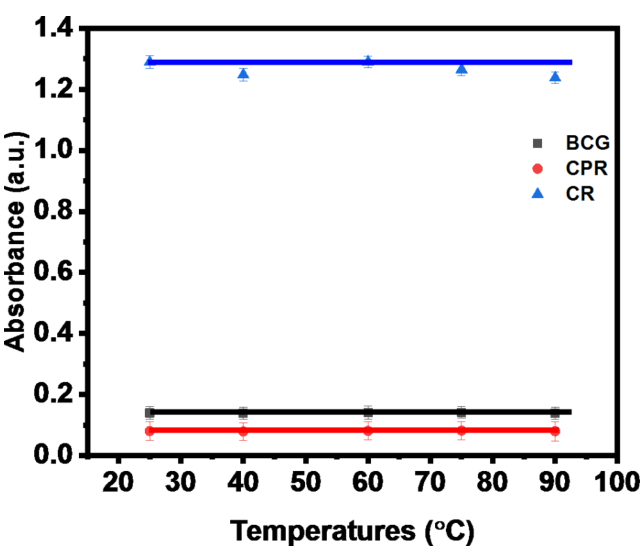

Fig. 6 Temperature effect of 0.05 M of bromocresol green, chlorophenol red, and cresol red dye solutions

in the range of 25 °C to 90 °C, indicating that this variable is likely not a significant factor in the deterioration of sensor performance (Fig. 6).

Interference studies

Processed foods are frequently treated with additives, and compounds that add colour, enhance flavour or extend shelf life. Most meals require keeping them fresh and nutritionally balanced. Small amounts of chemicals may be safe to consume, but health concerns pile up if you eat many processed foods. A processed-food diet has been related to chronic ailments such as obesity, high blood pressure, heart disease, and cancer.

Here, the analysis was carried out in pH 3 solutions for all dyes. The dye solutions recognized sodium sulfite in all cases (Fig. 7). According to the data, all dye solutions demonstrated extremely high selectivity for sodium sulfite.

Real time-assessment

For the real-time assessment of the proposed method, we evaluated the performance of colourimetric paper sensors in detecting sodium sulfite in beverages. Whatman No. 1 filter paper was functionalized by soaking it in pH adjusted 3 M dye solutions for one hour and then drying it at room temperature overnight. These filter sheets were exposed to sodium sulfite-containing beverages (0.079 M) to observe the colour change.

Because of the fast adsorption of dye solutions through its porous structure, the hydrophilic nature of the Whatman No 1 filter paper makes it a suitable tool; in fact, the use of paper as a platform for sensing devices offers significant advantages in terms of affordability and availability of functionalization processes [23]. It is extremely cost-effective because only a few chemicals are required to activate the paper device.

For the analysis, the developed paper sensors were cut into strips, which were used for further assessment. Analysis was carried out in fruit juices that contain sodium sulfite. These paper strips were dipped in this juice and observed for colour change. These strips were successful in detecting sodium sulfite (Fig. 8). In the case of bromocresol green, the colour changed from greenish yellow to blue after dipping in fruit juice. Similarly, the colour shift from dark yellow to magenta for chlorophenol red, and in cresol red, the colour shift was observed from yellow to light purple.

A portable prototype device with full detection capabilities for SS was fabricated. The sensor prototype displayed distinct RGB values when exposed to test solutions using dyes as sensing elements. The first method used three dyes

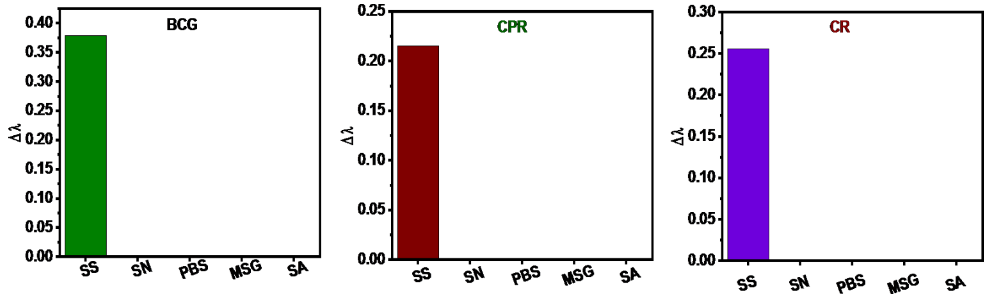

Fig. 7 Selectivity analysis of 1 M of sodium sulfite, sodium nitrites, potassium bisulfite, monosodium glutamate and sorbic acid in different dye solutions

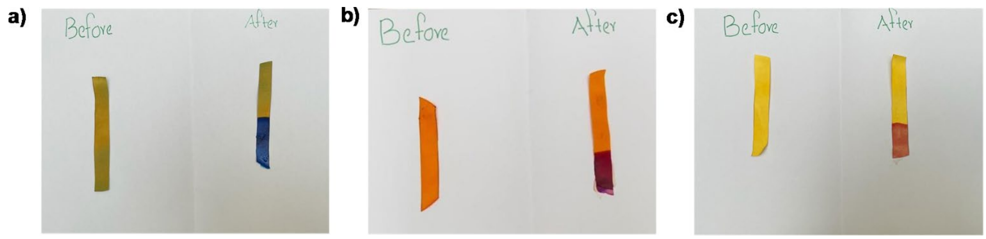

Fig. 8 Real-time assessment of fruit juice for sodium sulfite detection using different dye solutions (**a**) Bromocresol green (**b**) Chlorophenol red (**c**) Cresol red

Table 2 Analysis of dye mixture using sensor prototype at 0.5 M concentration

Sl. No	Dye Mixture	RGB values
1	CR(Nil) CPR (Nil) BCG (Nil)	265,195,10
2	CR(SS) CPR (Nil) BCG (Nil)	245,107,80
3	CR(Nil) CPR (SS) BCG (Nil)	255,95,25
4	CR(Nil) CPR (Nil) BCG (SS)	245,107,60
5	CR(SS) CPR (SS) BCG (Nil)	235,103,110
6	CR(SS) CPR (Nil) BCG (SS)	260,210,115
7	CR(Nil) CPR (SS) BCG (SS)	235,118,85
8	CR(SS) CPR (SS) BCG (SS)	275,215,96

to detect SS at a concentration of 0.5 M, and the RGB values obtained are shown in Table 2.

We can predict the system of dye solutions that detected SS based on the RGB chart. A similar RGB chart can be created for different test solution concentrations. These studies demonstrated that we could create a unique RGB chart for each dye solution. Another advantage is that based on the RGB values, this sensor prototype could detect the presence and concentration of each test molecule.

Conclusion

In this study, we elucidated a colour change visual paper sensor for sodium sulfite that can detect it quickly at room temperature with great selectivity and sensitivity. The breakthrough employs efficient dyes such as bromocresol green, chlorophenol red, and cresol red, which have never previously been used for successful sodium sulfite detection. The novelty of this study is that it uses dye properties related to their lack of stability to sodium sulfite to make it functional as a visual sensor.

For practical applications, Whatman No 1 filter paper was used as the dye-coated paper sensors are simple to use and can visually detect a very low sodium sulfite concentration (0.05 M) simply by changing colour. Along with the paper sensor, the developed IoT sensor also helps detect sodium sulphite in a fraction of a second. And this prototype and paper sensor can be used to detect other food additives by incorporating efficient dyes.

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Author contributions SD carried out Investigation and Data curation. JKA and MSTS did validation. MSS did the original draft preparation. MG, SM, and KKK helped in Writing- Reviewing and Editing. KKS took part in Supervision Conceptualization, Methodology. All authors reviewed the manuscript.

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Data Availability The data that support the findings of this study are available upon request.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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