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Storage quality assessment of shelled peanuts using non-destructive electronic nose combined with fuzzy logic approach

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ABSTRACT

The storage quality of shelled peanuts during storage were assessed using hybrid electronic nose (e-nose)–fuzzy logic approach, beyond conventional tests. Fuzzy logic was used to rank and screen best responsive MOS sensors (total 18) to detect global rancid odors from aged peanuts. Using e-nose data, an odor index (OI) was estimated and correlated with chemical rancidity indices (peroxide value (PV) and acid value (AV)). Multiple linear regressions (MLR) were used to predict the storage time and rancidity indices of peanuts using response data of fuzzified sensors. Fuzzy interpretation identified four sensors which best classified aged and deliberately rancid peanuts using principal component and hierarchical cluster analysis. E-nose data closely predicted the storage time of peanuts relative to chemical rancidity indices (R^2, 0.993; RMSE, 3.31 *vs.* R^2, 0.985; RMSE, 4.57) ($p > 0.05$). In addition, it predicted the rancidity indices with accuracy (PV: $R^2 = 0.995$, RMSE = 0.29; AV: $R^2 = 0.989$, RMSE = 0.19). OI of peanuts was highly correlated with PV (0.99) and AV (0.96) and estimated their discard time (basis threshold PV = O_2 at 10 mmol kg^{-1}) as 99 d (e-nose) *vs.* 97 d (conventional tests). The presented approach could be adopted as non-destructive alternative to conventional tests to assure post-harvest quality of shelled peanuts at agro-industrial settings.

1. Introduction

Peanut (*Arachis hypogaea* L.) is a legume crop taxonomically classified in *Leguminosae* family and mainly grown for its edible oil seeds. It is highly valued industrial oil crop grown in tropical and sub-tropical regions of the world. It is widely consumed as an economical supplement to counter malnutrition owing to its excellent nutritional profile (*ca.* 26, 48, and 3% protein, oil, and fibres, respectively) (Sarvamangala et al., 2011). Peanut is a trade crop with multiple platform applications in pharmaceutical and culinary industries as an important ingredient to therapeutic foods, peanut oil, peanut butter, peanut flour, and peanut based confections. The oilcake residue from peanut oil extraction is used as an animal feed and soil fertilizer to enrich nitrogen content.

The post-harvest quality of shelled peanuts is subject of utmost attention as it is highly susceptible to rancidity during storage and processing owing to high lipid content. The traditional methods to detect peanuts rancidity rely heavily on the chemical tests namely peroxide value (PV) and acid value (AV), and sensory evaluation by trained panels which is time and solvent consuming and often suffers panel's subjectivity issues (Zheng et al., 2009). The advanced analytical instrumentations namely gas chromatography–mass spectrometry (Liu et al., 2013), high performance liquid chromatography (Hepsag et al., 2014), and hybrid immuno–chromatographic methods (Zhang et al., 2011) can effectively resolve these issues. However, they suffer disadvantages of being expensive, laborious, and environmentally taxing due to the requirement of trained labor and organic solvents. The internal quality changes in peanuts require time to reflect on its surface; thus, making it hard to detect the real-time storage quality of the peanuts.

Recently, electronic nose (e-nose) has gained wide popularity in analytical space owing to its rapid and non-destructive ways to identify the global aroma profile (Esteves et al., 2014; Upadhyay et al., 2017a). It consists of an array of sensors which mimics the human nose in recognizing the complex odor associated with food products and integrated statistical data processing tools. The odor volatiles pass through the array of sensors, and their response signals are recorded as fingerprints for analysis. E-nose has been successfully applied in diverse fields with a particular focus on agro-food industry (Capone et al., 2013; Lippolis et al., 2014; Pacioni et al., 2014). Though, e-nose has been previously investigated to link the pattern of aroma related changes in peanuts during roasting operations (Osborn et al., 2001; Jensen et al., 2005); there are limited studies which described the e-nose based detection of rancidity indices in peanuts during storage (Wei et al., 2015; Xu et al., 2017). It is important to highlight that array of

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sensors in e-nose system adds to its versatility in identifying the global fingerprint of volatile compounds. Interestingly, Chatterjee et al. (2014) stated that the presence of multiple sensors in e-nose often creates poor sample discrimination and suggested the statistical approaches to filtering the most responsive sensors to analyze the odor fingerprints of food samples. Our group recently investigated the application of fuzzy logic as an appropriate filtering method to screen the e-nose data in oil frying (Upadhyay et al., 2017a). The analogy was derived from the application of fuzzy logic in sensory tests to rank the linguistic data of sensory panels (Jaya and Das, 2003; Routray and Mishra, 2012). Our best literature survey indicates that this is the first attempt to integrate fuzzy logic interpretation to data extracted from e-nose analysis to predict the storage quality of shelled peanuts. It is presumed that the present investigation could contribute to the existing knowledge gap in this domain and would allow the benefits of e-nose based quality assessment to be reaped at post-harvest scale of agro-food industries.

In this work, we investigated (1) ability of hybrid e-nose–fuzzy logic approach to filter (ranking and screening) the best responsive sensors which could be used to monitor the conventional chemical rancidity indices (PV and AV) in shelled peanuts, and (2) predict the storage time and chemical rancidity indices of shelled peanuts by data extracted from fuzzy screened sensors using multiple linear regressions (MLR). Odor Index (OI) was also calculated by data processing software integrated with e-nose data and later correlated with chemical rancidity indices to estimate the discard time of aged peanuts. The hybrid e-nose–fuzzy logic approach envisaged to be a non-destructive way to monitor the storage quality of shelled peanuts.

2. Materials and methods

2.1. Materials

A locally available variety of red color peanut (*Arachis hypogaea* L. var. GG 2) were procured from the local market of Kharagpur (West Bengal, India). Only fresh kernels that were uniform in size were shelled and used for trials. The analytical grade reagents and solvents were purchased from Merck (Mumbai, India).

2.2. Peanuts grouping and storage

Before storage trials, the shelled peanuts were distributed into two sets namely training peanuts (T) and storage peanuts (P). To acquaint the e-nose against volatile global fingerprint of rancid peanuts, it was trained using a duplicate set of deliberately rancid peanuts (T1–T5). For inducing the rancidity development in peanuts, each subset of T (*ca.* 200 g) were deliberately aged in a rancidity accelerating chamber equipped with UV light at high temperature (60 °C) and relative humidity (RH) (75%) for one (T1), two (T2), three (T3), four (T4) and five (T5) days. Post aging treatment, the rancid peanuts were relocated from rancidity chamber to an incubator maintained at 27 °C and 40% humidity. PV of each subset was periodically tested to ensure the threshold discard limit (O_2 at 10 mmol kg^{-1}) was exceeded. Once the threshold mark crossed, they were stored frozen before being analyzed for rancid odors by e-nose training. Unlike T set, the duplicate sets of P (*ca.* 2 kg) namely calibration (CP) and validation (VP) were incubated at 27 °C and 40% RH. A small amount (*ca.* 20 g) of peanuts were periodically withdrawn (3 and 5 d for CP and VP, respectively) for conventional quality measurements (PV and AV) and continued until the accumulated rancidity levels surpassed the threshold PV ($> O_2$ at 10 mmol kg^{-1}) (O_2 at 1 mEq. = O_2 at 0.5 mmol). Post chemical tests, the peanut samples were stored at −4 °C to arrest the chemical changes at the time of withdrawal before e-nose analysis.

2.3. E-nose assessment

The fresh and rancid odor (headspace volatiles) of T and P set of peanuts was evaluated by Fox 4000 e-nose system (Alpha MOS, Toulouse, France) consisting of a fully automated HS 100 auto-sampler (Alpha MOS, Toulouse, France) and an array of 18 metal oxide semiconductor (MOS) sensors. These sensors are classified as L-type (LY2/LG, LY2/G, LY2/AA, LY2/GH, LY2/gCTI, LY2/gCT; short chain fatty acids and aldehydes), P-type (P10/1, P10/2, P40/1, P30/1, P30/2, P40/2, PA2; aliphatic non-polar compounds), and T-type (T30/1, T70/2, T40/2, T40/1, TA/2; polar alcoholic and chlorinated compounds) that are responsive to wide range of volatile compounds (Oliveros et al., 2002). E-nose offers several advantages over sensory analysis: (1) It eliminates the subjectivity in sensory tests, (2) minimizes sensory saturation of members during prolonged exposure to an odor, (3) operates similarly to the human nose by analyzing complex mixtures of volatiles, and (4) requires no sample pre-treatment and gives quick results.

The operating procedure was as follows: 5 g shelled peanuts were transferred to a 20 mL glass vial, fitted with Teflon septum, for thermo-incubation (60 °C for 30 min) to generate and equilibrate the headspace volatiles. The accumulated volatiles were injected into e-nose system, assisted by an auto-sampler, using purified air stream (3.5×10^4 Pa) at a flow rate of 150 mL min^{-1}. The sensors responses were recorded as a change in resistance ($\Delta R/R$) about base values for 120 s followed by a recovery period (420 s) to allow the sensors to return to the baseline resistance. For each sensor, an absolute value of maximum $\Delta R/R$ was extracted. It is important to mention here that the instrument was first trained to get acquainted with volatile odor fingerprints of rancid peanuts using T set of peanuts (T1–T5), as described by Upadhyay et al. (2017b). Later, the P set of peanuts comprising CP (P0–P132) were analyzed for rancid odors and a combined odor map was generated to visualize their clustering with T sets. The VP set followed next for e-nose measurements. The data extracted by e-nose was used to rank the sensors using fuzzy logic analysis.

2.4. Fuzzy logic analysis

For fuzzy logic assisted ranking of e-nose data, an analogy was drawn from conventional sensory panel evaluation, and e-nose data ($\Delta R/R$) were regarded equivalent to hedonic scores (Upadhyay et al., 2017b). The fuzzy logic analysis is based on assigning certain weight or importance to specific attributes in food products (Jaya and Das, 2003). An assumption was made to assign an equal statistical weight to fresh and rancid peanuts. It is because the response of all the MOS sensors against odor fingerprints of peanuts have equal impact on their selection and estimation of discard time. Each sensor was assigned with different response scale factor (X_1, X_2, X_3, X_4, and X_5) which is a scalar quantity and referred to peanut sets (T (T1–T5) and P (P0–P132)) for which signal response ($\Delta R/R$) ranges: 0–0.25 (not sensitive, X_1), 0.25–0.5 (fairly sensitive, X_2), 0.5–1 (medium sensitive, X_3), 1.0–2.0 (good sensitive, X_4), and more than 2.0 (excellent sensitive, X_5) (Table 1) (Upadhyay et al., 2017b). For each sensor, the triangular fuzzy number (TFN) denoted triangular membership distribution function and fuzzy triplet set (*a*–*c*) (refer supplementary figure, Fig. S1). The triplet (*a*–*c*) was represented by a triangle, where *a* represented the fuzzy membership value of 1, *b* units left to *a* is one vertex, *c* units right to *a* is another vertex. The triplet set for sensors was calculated by Eqs. ((1)–(3)).

$$a = (0 \times X1 + 25 \times X2 + 50 \times X3 + 75 \times X4 + 100 \times X5)/50 \quad (1)$$

$$b = (0 \times X1 + 25 \times X2 + 25 \times X3 + 25 \times X4 + 25 \times X5)/50 \quad (2)$$

$$c = (25 \times X1 + 25 \times X2 + 25 \times X3 + 25 \times X4 + 0 \times X5)/50 \quad (3)$$

where, X_1, X_2, X_3, X_4, and X_5 denoted the response scale factors.

The defuzzification of overall sensor signal response scores (0–100) from triplets was done by calculating centroid value of corresponding triplets by Eq. (4). The defuzzified values of sensors were used to rank and screen them. Using the data extracted from fuzzy screened sensors,