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Prediction of theaflavin and thearubigin content in black tea using a voltammetric electronic tongue

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1. Introduction

Quality of tea has a distinct effect on the human palate and sensory organs. The chemical basis of tea quality is the concentration and composition of different chemical constituents present in tea that produce the desired effect on these organs. The contribution of major bio-chemical compounds to the taste of tea [1] is presented in Table 1.

Although individual chemical components present in tea have distinct and individual taste and flavor, the overall quality of tea is due to the multidimensional contribution of these constituent chemical compounds. The tea leaves after plucking are processed through different stages during which the compounds present in green leaves are transformed and finally the delicious end product with floral taste and aromatic flavor is obtained. For the CTC (cut-tear-curl) variety of black tea, these processing stages are called withering, cut-tear-curl, fermentation and drying. After the withering process, where the moisture content is reduced to a large extent in a controlled manner, the tea leaves are cut, torn and curled so that the enzymes responsible for producing better taste are released from vacuolar bodies in the cells to act on leaf polyphenols. The CTC stage is followed by the fermentation stage, where enzymic oxidation of polyphenols takes place. It is

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ABSTRACT

The two most important chemical groups that decide the liquor characteristics of black CTC (cut, torn and curled) tea are theaflavins (TF) and thearubigins (TR). Hence, a quick estimation of concentration of these compounds can significantly contribute to the evaluation process for the quality of finished tea in an objective manner. In this paper, a scheme for rapid measurement of concentration of TF and TR is described using a voltammetric electronic tongue with five working electrodes made of noble metals. The results indicate good correlation of electronic tongue predictions with the actual concentrations obtained using ultraviolet–visible spectrophotometer.

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during this stage, the taste or flavor of tea increases. The last stage of tea processing is drying, where the moisture content of tea leaves is reduced to 2-3%.

During the fermentation stage, oxidation of polyphenols is brought about by certain oxidase enzymes present in tea shoots to produce two major groups of pigments known as theaflavin and thearubigin [2]. Theaflavins normally constitute about 0.5-2% of dry weight depending on the processing parameters of tea, while thearubigins constitute about 6-18% of dry weight. Theaflavins impart briskness and brightness while thearubugins contribute to the mouth feel (thickness) and color [3]. Spectrophotometric and human sensory panel studies suggest that TF content correlates positively with liquor brightness [4]. The TR content has, however been found to contribute for the mouth-feel sensation of tea liquor, but at the same time relates negatively with liquor brightness and taste. Though the concentrations of other chemicals contribute to the quality of finished tea, the concentrations of TF and TR have much greater influence on the brightness, briskness, depth of color, strength, mouth-feel and overall quality of made tea [2,3,5-8].

The ratio of TF and TR is considered as a quality parameter by the tea scientists [9] and the estimation of these two groups of compounds in tea gives a reasonable idea about the quality of tea. This parameter, i.e. the ratio of TF/TR in a certain sample does not contain any uncertainty or ambiguity as compared to the quality scores assigned by the expert human panel. The reproducibility and consistency of the quality scores assigned by the tea tasters could not be guaranteed as they are subjective, strongly affected by mental state

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Table 1

Contributions of major biochemical compounds to the "taste" of tea.

Compounds	Taste
Polyphenol	Astringent
Amino acids	Brothy
Caffeine	Bitter
Theaflavins	Astringent
Thearubigin	Ashy and slight astringent

of taster and prone to sensory adaptation. The analytical methods like gas chromatography (GC), spectrophotometry and high performance liquid chromatography (HPLC) can also give quantitative idea about the composition of tea liquor or aroma but are expensive in terms of time and money and also require expert handling. Thus there is a need towards finding an instrumental, objective and inexpensive means for tea quality evaluation. Towards this direction, are proposed some electronic instruments like electronic nose [10,11] and electronic tongue [12]. These systems mimic the senses of smell and taste of the human sensory panel and lead to rapid quality standardization of finished tea. These measurement systems have been used to grade the quality of finished tea [11,12] and detect optimum fermentation time of tea processing [13]. But, the outputs of these instruments calibrated by quality scores of the human tasters are also susceptible to uncertainties and limitations. An approach which might help users to gather improved quality-perception of tea from these instruments is to train them to predict concentrations of biochemical constituents responsible for quality. The generation of target data in this case the concentration of chemical compounds, can be determined by standard instrumental techniques and will be free of organoleptic limitations.

In this pursuit, a method is described, where a voltammetric electronic tongue is employed to estimate the concentrations of TF and TR. Voltammetric techniques with LAPV (large amplitude pulse voltammetry) waveform generate large amount of information with low detection limit [14,15]. This can be very helpful, when the objective is to look for quantification of chemical species dominating the taste of tea. Literature survey reveals that an electronic tongue can evaluate the levels of tea astringency [16,17]. It can differentiate among various grades of tea [12,15,18] and has been predominantly used for classification of food products [19–26]. An electronic tongue is also employed for compositional analysis [27–29].

The methodology to estimate the content of TF and TR for an idea about tea quality is not new. Initial reports for estimation of TF and TR using spectrophotometric measurements are available in [30-33]. Estimation for TF and their fractions using gas liquid chromatography are reported in [34]. HPLC instruments are extensively used for extraction and separation of TF and TR fractions [35]. Studies on the detection of TF, TR using HPLC with photo diode array (HPLC-PDA) are also reported in [36]. Near infrared reflectance spectroscopy has been used for estimation of TF in [37]. Wright et al. proposed the use of capillary electrophoresis [38,39] for analysis of TF. Highspeed countercurrent chromatography (HSCCC) has been applied for the separation of TF, TR and catechins in [40–42]. Use of micellar electrokinetic capillary electrophoresis (MEKC) method is reported for detection of theaflavins in tea [43]. All the above methods employ costly instruments with complex procedures for estimation and detection of these compounds. But it is expected that the proposed methodology using an electronic tongue shall be novel, rapid and a cost effective alternative.

In the present work, the estimation task is taken up as a function approximation problem. The regression models are developed using partial least squares (PLS), support vector regressors (SVR) and multilayer feed-forward neural networks. The calibration of these models has been carried out using TF and TR content determined through UV–vis spectrophotometry. The operational flowchart for the whole process is shown in Fig. 1.



Fig. 1. Operational flowchart.

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