

A Review of Technologies for Detection and Measurement of Adulterants in Cereals and Cereal Products

Ashabahebwa Ambrose, Byoung-Kwan Cho*

Department of Biosystems Machinery Engineering, College of Agricultural and Life Science, Chungnam National University, 99 Daehak-ro, Yuseong-gu, Daejeon 305-764, South Korea

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Abstract

Purpose: The continued increase in the world population has triggered an increased demand for food. Cereal grains, flour, and their products constitute the staple diet for most of the world's population. This high demand for food, particularly for cereal-based products, has been exploited for commercial gain through adulteration of food materials. We provide a thorough review of the current developments and limitations of modern, nondestructive analytical techniques used for detection of adulterants in cereals and their products and compare them with conventional methods. **Results:** Adulterated food poses a serious health risks to humans, animals, and the ecosystem in general. Over the last few decades, the adulteration industry has developed fraudulent practices that often outsmart conventional methods of detection and quality control. Therefore, technological advancements to aid in detection and measurement of adulterants in food products and to ensure food quality and safety are critically important to consumers worldwide. **Conclusion:** There is a continuous demand for development of nondestructive technology to improve the accuracy and efficiency of detection, measurement, and qualification of adulterants in cereals and other food materials.

Keywords: Adulteration, Cereals, Chemometrics, Nondestructive, Spectroscopy

Introduction

Cereal grains and their products are the staple diet for most of the world's population. Natural cereals are a rich source of nutrients such as vitamins, minerals, carbohydrates, fats, oils, and protein (Brás et al., 2005). The continued increase in market demand for cereals due to increasing world population and the diverse applications of cereal products such as production of processed food and beverages and manufacture of adhesives, cosmetics, and detergents (Cozzolino, 2014) presents an opportunity for unscrupulous producers and intermediaries to adulterate food products and increase their profit margin by adding low-grade or synthetic materials to increase product bulk.

Most adulterants are similar in appearance to the original

product and are indistinguishable when combined (Lakshmi, 2012). Noonan (1981) and Schul (2000) explained that some colored adulterants such as Sudan I–IV dyes are added to food products to enhance or change their color and make them more attractive to prospective buyers.

Various reports suggest that some agents can effectively be integrated up to a significant proportion into food products and still be undetectable by physical means or conventional methods. The resulting food products have similar or improved physical attributes compared to the original products in terms of color, texture, taste, and overall acceptability (Doxastakis et al., 2002).

Indirect additives inadvertently included in cereal products such as insects and/or bone fragments, husks, and sand are another form of adulteration (Perez et al., 2003; Lankmayr et al., 2004; Pavino et al., 2010). Some insect pests attack pre- or post-harvest cereal kernels and mostly escape undetected during processing (Campbell et

*Corresponding author: Byoung-Kwan Cho

Tel: +82-42-821-6715; Fax: +82-42-823-6246

E-mail: chobk@cnu.ac.kr

al., 1976).

All adulterants decrease the quality of the final product and often undermine the integrity of the original food. Therefore, reliable tests and analyses are essential prerequisites for quality control, consumer protection, and ultimately business optimization.

Adulteration

Adulteration refers to the addition of substances of inferior quality or removal of vital component(s) from food, thus lowering its nutritional value. An adulterant is any substance that, from a legal, nutritional, or safety perspective, should not be consumed. Food fraud has developed into a sophisticated industry with high-tech investments attracted by the massive and inherent potential for economic gain. Defernez and Wilson (1995) and Haughey et al. (2013) point out that some cereal adulterants comprise natural and organic materials with less harmful effects, whereas others are synthetic or inorganic and pose serious health risks to consumers (Figure 1).

Common adulterants in cereals and cereal products

Sudan I dye

Sudan I dye is a synthetic, red-colored chemical traditionally used to color plastics and textile products. However, it has seen recent use as a food colorant, especially for culinary spices (Di Anibal et al., 2012), because of its attractive color, availability, and low cost (Di Anibal et al., 2011). According to López et al. (2013) and the International Agency for Research on Cancer (IARC), Sudan I dye is carcinogenic and genotoxic and is

therefore prohibited for use as a food colorant.

Lard

Lard is commonly found in cake product formulations, especially for animal feed, and is associated with health risks such as hypercholesterolemia and coronary disease (Syahariza et al., 2005).

Melamine

Melamine is frequently added as a protein booster since it contains a nitrogen bond similar to that in natural proteins. However, foods adulterated with melamine, notably wheat and soya bean, pose a tremendous risk to both human and animal health (Haughey et al., 2013).

Natural adulterants

Edible wheat is classified into two categories: durum wheat and bread wheat (Cocchi et al., 2006; Zapotoczny, 2011). These two varieties have different physical, chemical, and nutritional properties, and their commercial values are consequently distinct. High-quality durum wheat is therefore often adulterated with the cheaper, low-quality bread wheat to increase bulk and profit margins.

Others

Other cereal flour adulterants include, but are not limited to, mungbean powder in spirulina, cornstarch in lotus root powder, and lupin bean in wheat flour (Pompei et al., 1985; Parisot et al., 2001; Magni et al., 2005; Scarafoni et al., 2009).

The negative effects of adulteration require consumers to be cautious about the commercially available products. However, consumer cautiousness is often insufficient to



Source: <https://www.google.co.kr/search?q=adulterated+bread&newwindow> (accessed July 2014).

Figure 1. Illustration of bad taste and health risks of adulterated food.

prevent the purchase and consumption of adulterated products because they do not have the resources (technical knowledge and equipment) to ascertain product authenticity. An organization for standards or an authentication authority for each geographical area must take responsibility on behalf of the public to test and analyze the authenticity of the food products presented to the consumers.

Several methods using tools ranging from low capability instruments to sophisticated high-tech equipment have evolved to detect and measure adulterants to ascertain food quality (Hernández et al., 2014). However, the adulterers continually circumvent accepted standards of authentication by developing new methods of adulteration.

Conventional methods used to detect adulterants in cereal food products

Gas chromatography

Gas chromatographic techniques can explicitly separate samples and identify components that can be vaporized without being degraded. Separation of molecular components is based on differences in vapor pressure at the boiling point created by the stationary liquid phase and mobile gas phase within the chromatograph. Gas chromatography is useful for detection and measurement of synthetic cereal adulterants, e.g., β -adrenergic agonists in animal feeds and Sudan I dyes in culinary spices (Reid et al., 2004; Caban et al., 2011). It is also used to quantify and classify metabolites in cereal plants such as maize, rice, wheat, barley, rye, and oats (Khakimov et al., 2014). Chemometric analysis is necessary for extraction of the molecular signatures of adulterants present in the sample (Alves et al., 2003; Reid et al., 2004). This adds to the labor, expertise, and time required for analysis and is the most common drawback for this technique (Cao et al., 2013). The combination of gas chromatography and mass spectrometry (GC-MS) potentially reduces the time of analysis, but its application is greatly limited by the high cost of equipment and operation (Reid et al., 2004).

Enzyme-linked immunosorbent assay (ELISA)

ELISA, an antibody-based analytical technique available since the 1970s, evolved as a replacement for radioimmunoassays and has been used in detection of adulterants and contaminants in various food materials (Robertson et al., 1986; Lin and Cousin, 1987; Notermans et al., 1988; Tsai and Cousin, 1990). The biochemical reaction between the antibody and enzyme substrate introduced in the test

generates a signal, which is visualized as a color change in the substrate. The intensity of the color directly correlates with the concentration of the analyte in the sample. Tsai and Yu (1999) used ELISA to detect aflatoxigenic molds in naturally contaminated corn, rice, wheat, and peanut samples. The result of their study indicated that ELISA measurements were highly correlated, with coefficient of determination (R^2) values of 0.94, 0.93, 0.96, and 0.86 for corn, rice, wheat, and peanuts, respectively. More recently, ELISA has been incorporated into other techniques to enhance the performance of target detection. For example, Cao et al. (2013) used liquid chromatography-tandem mass spectrometry (LC-MS/MS) to verify the analytical model they developed to detect phenylethanolamine-A using ELISA. Although ELISA has high, it is limited by prevalent background artifacts that reduce its net-specific signal output.

Polymerase chain reaction (PCR)

PCR is the most commonly used analytic method for detection of adulterants in food. PCR is a DNA-based technique that is highly sensitive to molecular formation and is ideal for natural products with intact DNA. Any form of disequilibrium in the DNA structure due to loss, distortion, or the presence of inhibitors may negatively affect the results. DNA of food materials, especially cereals, undergoes physical and chemical changes from oxidation reactions and mechanical shearing during processing and/or extraction (Cankar et al., 2006). This renders PCR less effective in detection of adulterants in cereal flours and their products. DNA is quantified and classified using established standards from a DNA quantification kit with a fluorescent dye, bisbenzimidazole H 33258, which binds to the minor groove of dsDNA (Moe et al., 1994). This technique requires specialized personnel and is laborious, expensive, and time-consuming compared to modern analysis techniques such as mass or vibrational spectroscopy.

Electronic nose

The electronic nose is an instrument that detects odors and flavors that uses an array of semi-selective chemical sensors and pattern-recognition systems derived from mammalian olfactory sequencing for detection of (usually volatile) adulterants in flour and similar food products (Olguín et al., 2014; Zhang and Tian, 2014). The data computing system generates a fingerprint for the interaction between the scent from the target sample and the response

from the chemical sensors that is compared to standard reference signals (Chen et al., 2013; Jiang et al., 2014). This technique has been applied in several settings including medicine, manufacturing, product standardization, and food authentication (Arshak et al., 2004, Chen et al., 2013; Lippolis et al., 2014). The high potential of the electronic nose technology is attributed to its considerable accuracy, non-invasiveness, and rapid detection. However, applications have been limited due to the requirement for highly trained and skilled operators and its sensitivity to temperature, humidity, and non-target gases, which often reduce the accuracy of test results. Intensive research and development in this area could harness the full potential of the electronic nose for qualification and quantification of adulterants in the future.

Generally, conventional methods can be subjective, invasive, non-specific, and time-consuming, and most require large and expensive equipment and fail to disregard intrinsic artifacts. Recent modern analytical techniques have considerably addressed some of these shortcomings to improve detection and measurement of adulterants in food products, especially cereals.

Modern analytical techniques used for detection of cereal adulterants

Food safety control in the 21st century requires approaches with high accuracy, rapid turnaround, and flexibility that allow dynamic and sophisticated analyses to counter the existing, high-tech adulteration techniques that have so far surpassed conventional methods. Modern methods and equipment are encoded with sophisticated mathematical formulas and computer-aided algorithms to trace specific chemical, physical, or textural properties based on the identification of one or more molecules characteristic of a particular adulterant (Scarafoni et al., 2009). We discuss the most common methods used in detection and measurement of adulterants in cereals below.

Fourier transform infrared (FTIR) spectroscopy

FTIR is a method of spectral measurement performed with long-wave infrared radiation that records absorbance in a time domain and converts it into a frequency domain using the Fourier transform algorithm. The sizes of the frequency peaks in the spectrum are proportional to the amounts of chemical constituents present in the sample (Baravkar and Kale, 2011). Syahariza et al. (2005) used FTIR to detect lard adulteration in cake formulations, and

the resultant model indicated a high potential for identification of adulterants with $R^2 = 0.979$. Sivam et al. (2012) successfully used FTIR to identify and classify two types of wheat bread with differing amounts of pectin. The absorption bands indicated the functional groups present and their molecular conformations based on their characteristic frequencies (Sivam et al., 2012). This technique has a high potential to identify adulterants in cereals more reliably than conventional infrared spectroscopy.

Near-infrared spectroscopy (NIRS)

NIRS a visual technique for food quality measurement and is the most widely used technique for food authentication. Over the last decade, NIRS has gained popularity in the grain processing industry to assist with standardization in several parts of the world (Pierna et al., 2012; Salgó and Gergely, 2012; Albanell et al., 2012). According to Williams (1975), the Canadian Grain Commission adopted NIRS as their official analytic method for proteins and detection of adulterants in wheat in the 1970s. Other reputable agencies have since adopted the technique for food authentication including the Federal Grain Inspection Service of the United States in 1980 and the American Association of Cereal Chemists (AACC) in 1989 (Osborne, 2000). NIR radiation is part of the electromagnetic spectrum that covers wavelengths in the range 780-2500 nm. The incident ray is directed to the target sample and is modified through interaction with the sample due to absorption or transmission scattering that are influenced by physical and/or chemical disturbances at specific wavelengths in the NIR spectrum. Chemometric formulations with multivariate statistical analyses are performed on NIR spectra by PCA or PLS regression to correlate the NIR spectrum with the quality attributes of interest (Martín et al., 2014; López et al., 2014). The β -coefficients in PLS regression indicate the importance of particular wavelengths used in calibration (Che Man et al., 1999). Response prediction is calculated by multiplying the absorbance at each wavelength by the corresponding β -coefficient and taking the sum over all wavelengths considered. The NIR spectra mainly correspond to C-H, O-H, and N-H vibrations (Wu et al., 2011). The potential of the NIR technique stems from its inherent operational characteristics: it is nondestructive, rapid, and requires little or no sample processing. Recent improvements in software and hardware such as FTNIR and high-speed NIR spectrophotometers have increased performance efficiency.

Hyperspectral imaging (HSI)

HSI utilizes the NIR wavelength range to simultaneously acquire both spectral and spatial information about the target sample (Ruiz-Altisent, 2010; Iqbal et al., 2014). Spatial information provides a virtual view into the composition of cereals and their products that enables a chemical map of the sample to be extracted from a hypercube. The capability of HSI to provide information about the distribution of chemical and physical disorders makes it relatively superior to conventional NIRS, which only provides information at the scanned point. Dimensional disturbances in the acquired hyperspectral data can be resolved using multivariate statistical image analysis tools. Several statistical image analysis parameters such as the maximum, minimum, mean, median, standard deviation, variance, and other histogram features can be extracted from images at specific wavelengths (Singh, 2009). Smail et al. (2006) and Singh et al. (2007) successfully used this technique to detect fungi in wheat grains and sprouted wheat kernels, respectively. Linear discriminant analysis (LDA), quadratic discriminant analysis (QDA), and Mahalanobis analysis are the most essential supervised classifier algorithms for extracting useful analytical information about the different composition classes in a sample.

Nuclear magnetic resonance (NMR)

NMR spectroscopy is based on the absorption of electromagnetic radiation in the radio-frequency region of the spectrum that changes the orientation of spinning nuclei in a magnetic field (Reid et al., 2006). This electromagnetic technique requires sensors with highly sophisticated mathematical models and computer software algorithms that can be harnessed to relate physical or chemical properties and quality attributes of the product in consideration. The combination of PLS-DA and NMR has been successfully used to detect adulterants in several foodstuffs including cereals and cereal products (Boffo et al., 2009; Marcone et al., 2013). PLS-DA is a widely used multivariate technique that uses few latent variables, takes into account the correlation between the variables, filters the noise, and leads to good predictive performance of the model. NMR analyses contain relatively less bias compared to those of other techniques such as GC-MS, LC-MS, and CE-MS, and NMR is the most widely used technique in food metabolomics tests, especially for cereal-based food materials.

UV-vis spectroscopy

UV-vis employs wavelengths in the visible range of the electromagnetic spectrum between 400-800 nm. It is principally used for classification based on color differences or other visual sensorial characteristics. González et al. (2005) pointed out that UV-vis is essential for detection of colored adulterants in food matrices. However, there is limited information indicating serious advancements in the application of UV-vis spectroscopy for detection of adulterants in cereal food products as few studies have been conducted in this area compared to other spectroscopic techniques. This technique is limited to detection of visual attributes, which is a disadvantage as most adulterants have more pronounced chemical characteristics that can easily be detected and interpreted by other spectroscopic techniques combined with chemometric analysis.

Raman spectroscopy

Vibrational Raman spectroscopy is a proficient analytical method used for fingerprinting and investigating molecular distributions in a target sample. In the recent past, Raman spectroscopy has been widely used in detection and quantification of adulterants in food matrices including cereals because of its ability to identify a wealth of chemical information about the composition of a sample. Several forms of Raman spectroscopy including surface enhanced Raman spectroscopy (SERS), coherent anti-stroke Raman scattering (CARS), and resonance Raman spectroscopy have been developed and are applied in different situations. Di Anibal et al. (2011) successfully used SERS as a screening tool for Sudan I dye in culinary spices. Fluorescence caused by impurities and redundant chemicals in the sample is the most common drawback for the weak Raman signal but can be minimized by selecting an appropriate wavelength for a particular sample and by applying various multivariate statistical methods for signal pre-processing and data analysis. Raman spectroscopy is suitable for micro-scale measurements but is time intensive for measurements of large samples. Recent innovation of "Line Scan Raman Spectroscopy (LSRS)," which can extract spectral data from several samples arranged in a line with a single shot, and non-target Raman spectroscopy (NTRS), which extracts data at several points, can potentially reduce the operation time and enhance performance efficiency.

Conclusions

The development of nondestructive technologies for detection and measurement of adulterants in food matrices has progressed remarkably over the years. Aesthetic and ergonomic improvements have enhanced functionality, reliability, and general performance efficiency. Fourier transform-based techniques such as FTIR and FTNIR and improved Raman spectroscopy approaches have increased performance confidence in multiple areas of food quality management, especially for cereals. However, widespread application of most non-invasive spectroscopy-based techniques is still limited by the high cost of equipment, which is prohibitive for many potential users. Current and future development of light and portable equipment with remote sensing features will reduce the number and size of system components and hence the costs of equipment and operation. Continuous modification of the software algorithms for existing techniques such as FTNIR, FTIR, and SERS is needed to reduce the analysis time and enhance prediction efficiency. Similar advances in software and hardware are essential for accurate detection, measurement, and qualification of adulterants in cereals and other food materials.

Conflict of Interest

The authors have no conflicting financial or other interests.

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