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Review Article

Examining the Detection of Urea and Melamine as Adulterants in Milk and Milk Products

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The modern research is required to identify adulterants in milk, as it is now being falsified in more complex ways. The study was conducted in response to contemporary concerns about natural milk being tampered with in order to boost its marketability. This study is developed as a "adulterant-based" survey that specifically evaluates melamine and urea in order to provide a clear assessment of qualitative detection methods exclusive to the specified adulterants. Numerous surveys of research on traditional detection approaches have been paired with an in-depth examination of contemporary advancement. This section discusses the basic notion of purposeful adulterant addition, its influence on human health, and the detection tools available to counteract the mass threat to human life.

Keywords: Adulteration, Detection, Milk, Melamine, Urea

1 Introduction

Milk fat is an important nutritional component that has an impact on the economics, nutrition, physical and chemical aspects of milk and milk-derived products. Because of its great nutritional value, it has been widely consumed by humans for centuries, particularly during childhood. It has a high nutritional value in its natural state and provides modest level of protein, fat, carbs, vitamins and minerals in an easily digestible form[1]. Regretfully, it is easily tampered with all across the world. Adulterants have been employed in the past, and sometimes even harmful drugs have been used. It was common in the United Kingdom until the Victorian era where colouring was done to cheese using lead. From the bygone times till the current decade, adulteration in milk have been ruining the human health and sometimes even causing death. Melamine, urea, chlorine, starch, hydrogen peroxide, detergent, pesticides and preservatives are the most common adulterants found in milk [2] which is shown in Figure 1. Milk is usually diluted with water in rural and urban regions to increase the volume which reduces the amount of protein in the diluted milk. To overcome this, a substantial amount of melamine and urea is added as an adulterant to boost the fake protein content casually[3, 4]. Research reports evidences that intake of melamine causes death and renal failure whereas urea affects heart, kidney, liver and causes indigestion, acidity and ulcers[5]. Melamine has a permitted limit of maximum 2.5 $mgkg^{-1}$ in imported foods, notably those containing powdered milk from china, and 1.0 $mgkg^{-1}$ in new-born formula, according to the European Commission and the US FDA[6]. Similarly, the maximum amount of urea allowed in milk is about 11.547 M[7]**.** In 2008, SANLU, one of china's largest dairy production companies, added 20.298M of melamine to milk products (WHO/FAO study, 2008)[8], which was 1706 times the WHO limit. Almost 294000 new-borns, 13 adults, and 51,900 children were hospitalised and affected by kidney difficulties as a result of the incident. After an outbreak of urinary stones in babies and children who consumed melamine-tainted milk in china and the finding of contaminated pet food in the United States, the impacts of melamine on human and animal health garnered considerable attention[9]. Demand and supply gaps, perishable nature of milk, poor customer purchasing power, and a lack of effective detection tests are all possible causes. Hence proper detection of the various adulterants in the milk is an increasing concern to avoid life fatalities[10], including Gas Chromatography Mass Spectrometry (GC-MS), Liquid Chromatography Mass Spectrometry (LC-MS), High Performance Liquid Chromatography (HPLC), Capillary Electrophoresis (CE) and Surface Enhanced Raman Spectroscopy (SERS). Similarly, conductance

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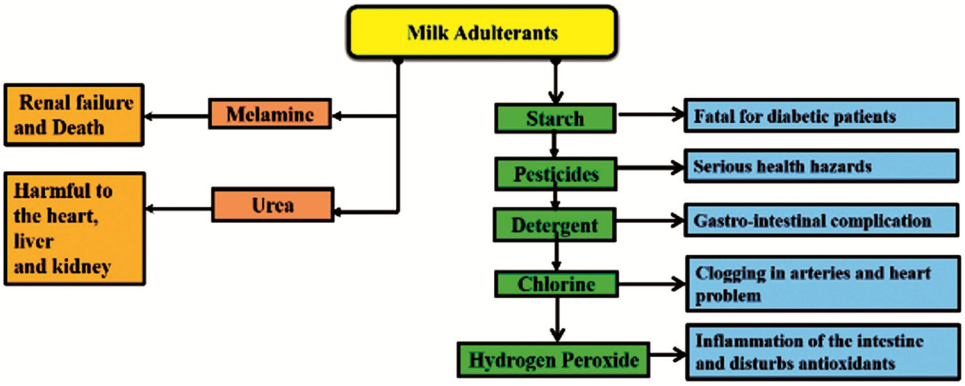

Fig. 1 — Common adulterants found in milk and its effects on human body.

measures, potentiometric measurements, Flow injection analysis and Spectrophotometric detection methods have all been described for the determination of urea[11, 12]. This critical review emphasis various melamine and urea detection methods that have been developed thus for. A few other adulterants and their detection methodologies as well as recent technological breakthrough, performance characteristics and future prospects were also meticulously reported.

2 Materials and Methods

2.1 Melamine and Urea detection-Recent survey

Various approaches were used to determine the amount of melamine in milk some of the latest reports are briefed as follows. Quang Hieu-Tran[13] reported study that uses Liquid Chromatography tandem Mass Spectrometry (LC-MS/MS) to identify melamine (MEL) in milk and dairy products. This was reported to have fast, selective and sensitive results when checked for adulterant in the milk sample. This approach was used to examine 20 different varieties of milk available on the Vietnamese market which showed nil presence of melamine. Filazi[14] reported HPLC method for the detection of melamine in the milk and dairy products. After that, 300 samples of milk and dairy products purchased from major Turkish merchants were analysed using the approach. Melamine was not discovered in infant formulae or pasteurised UHT milk, although it was found in 2% of cheese, 8% of milk powder and 44% of yoghurt samples at concentration of 121, 694, 146 and 294 98 g/kg, respectively. The suggested approach proved sensitive, reliable and accurate allowing for the identification of melamine residues in dairy products at levels as 105 to 340 g/kg. Sijia[15] used the SERS approach for the study of different concentrations of melamine in milk. This method uses dual-mode readout sensing device that successfully distinguishes between actual and false-positive melamine in milk signals. The combination of colorimetric and SERS methods in this study reported not only allows for quick preliminary screening of melamine with the naked eye, but also considerably decreases false-positive signals in SERS due to surface selection rules. Limin[16] employed the SERS approach to identify melamine in milk. For quick detection of melamine in milk, a simple Ag nanocube (NC) array substrate was created. This method identified melamine as low as 0.01 ppm in reference solutions and 0.5 ppm in real milk samples. U.B.Trivedi[17] developed a potentiometric biosensor for determining urea in milk which uses urease's enzymatic breakdown of urea. This broken urea is determined potentiometric ally utilising a variety of transducers. These studies have shown that the product has a reasonable storage stability and shelf life. The proposed urea biosensor system is reported to have a limit of detection of 2.5 x 105 mol/L. Martina Baumgartner[18] evaluated flow injection analysis for urea measurement in sheep and cow milk. The evaluated technique was accurate and precise, with a sample capacity of 55 samples per hours but was reported to have increased cost thus avoiding it for routine analysis.

2.2 Adulterants Detection Methods

Apart from melamine and urea, milk contains a variety of adulterants as stated before. Below is the light thrown on these varieties for better idea behind the complete milk adulteration process. Shujun Dong[19] worked on raw dairy cow milk adulterated with chlorinated paraffins. Raw cow milk samples were analysed and found to have short and medium chain chlorinated paraffin concentration. The CP given through animal feed was found to have great

impact in the milk produced by it through this investigation. Mohsen Moozami Goodarz[20] used black carrot anthocyanins in starch to create an easy-to-use methodology to check the freshness/spoilage of pasteurised milk in this study. The colour shift for various pH varied samples demonstrated on this study had a strong link with the physiochemical and microbiological changes in the milk during spoiling, implying that the indicator can distinguish between fresh, medium fresh and ruined milk. Manoel.J.A[21] proposed a colorimetric spot test and smart phone based photometry as a new method for detecting hydrogen peroxide in milk. Digital images are taken immediately in disposable micro-tubes under controlled illumination conditions without sample pre-treatment, while the acid medium required for complex formation also serves as a protein precipitation solution. Adulterant detection takes about 2 minutes, and a linear response was achieved with a high confidence level and a 1.7 mg L^{-1} limit of detection. Abid Hussain[22] used Surface Enhanced Raman Spectroscopy for the detection of preservatives such as sodium thiocyanate (STC) and benzoic acid (BA) in milk. The proposed substrate was sensitive, and the approach only necessitated a few basic sample prep steps.

2.3 Latest technological advancements

A team (IISc, Bangalore, India) has created a low-cost portable device that can swiftly detect melamine in milk and milk products, where conventional methods are time-consuming and typically require expensive and complicated equipment as well as highly educated personnel[23]. The researchers utilised the novel gadget to test a range of melamine concentrations and were able to detect melamine levels as low as 0.1 parts per million (ppm) in water and milk, well below the permissible limit of 1 ppm. Another research report from the same institute reported that the way milk evaporates can reveal the presence of adulterants[24]. This study showed that by analysing deposition patterns following evaporation, "low-cost and effective approach" for detecting the presence of urea and water in milk can be done. In diluted milk, this approach was claimed to detect water concentration of up to 30% and urea amounts as low as 0.4%. The researchers claim that the technique does not require the use of a laboratory or other specialised skills, and that it may be easily adapted for use in rural and remote places.

2.4 Future prospects

Despite the reported literatures on the detection and sensing technologies of melamine and urea in the milk and milk products, additional research and development is required because concerns such as sensor poisoning and lack of specificity with the sensors used in electronic nasal equipment still exists. Electronic nose technology could have a wide range of industrial applications, including milk certification and quality assurance, if these problems can be overcome. Future improvements in analytical techniques are anticipated to focus not just on improving existing analytical procedures, but also on automating sample preparation activities that are difficult to automate. The future of milk authentication procedures must eliminate time-consuming sample preparation protocols. The development of analytical systems that combine strong analytical instruments with data processing software appears to be the way of the future.

3 Results and Discussion

3.1 Adulterant sensor's specification

Different performance parameters like sensitivity, linearity, resolution, span, response time are used to analyse milk adulteration and to test the level of adulterants added in milk. Some of the important parameters shown in Fig. 2 are discussed in brief as follows.

Sensitivity(S) is defined as the ratio of output obtained foe input given for the sensor which is given using the Eq (1)

$$S\% = \frac{output\ obtained}{input\ given} \times 100 \quad \ldots(1)$$

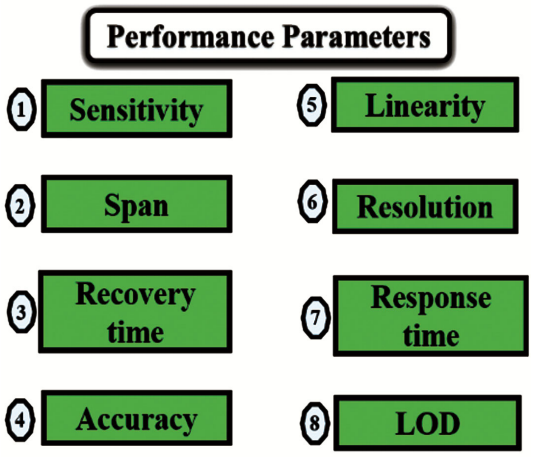

Fig. 2 — Various performance parameters of milk adulterant sensor.

Linearity of the adulterant sensor is the measure of consistency over range of values measured.

Limit of Detection (LOD) is defined as the lowest detection limit up to which a sensor can detect the adulterant present in the milk.

Resolution is briefed as the tiniest change that can be quantified

Accuracy of a measurement refers to how close it is to the true value being measured. Range of accuracy is the sensor's maximum range of sensitivity.

Span is calculated using the difference between the scale's minimum and maximum of the particular range of sensor.

After removing the measured variable in steps, the time is taken for a sensor to return to its baseline value. This is termed as recovery time.

The response time of a sensor output can be described as the time it's taken for it to change from its prior condition which is called as response time.

4 Conclusion

Although financial gain is one of the main drivers of milk adulteration, a lack of supply due to the world's growing population has also contributed. Because of a lack of competent monitoring and law enforcement, the problem is especially seven in emerging and underdeveloped countries. Existing common detection procedures are not always practical or available in these nations, making it difficult to handle the various forms of milk fraud. This necessitates a collaborative effort between specific groups and regulatory agencies in the development, implementation, and dissemination of improved milk adulteration detection tools. Furthermore, understanding and access to information may play a crucial role in addressing this difficulty in specific locations. Simple consumer detection technologies, as well as state-of-the-art authority tactics, can put an end to the problem for the victims, who include millions of children in developing countries.

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