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# Reducing Aluminum Toxicity in Food Packaging Using Bio-Polymer

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## Abstract

In the era of the modern food industry, food packaging has attracted much interest because it helps to protect food from environmental factors and in maintain its the integrity. Aluminum, widely used as containers, foils and pouches in packaging of hot foods, snacks and beverages. Many studies have shown that Aluminum content is increased in brains of patients with Parkinson's (PD) and Alzheimer's diseases (AD). It has been suspected that leaching of Aluminum from the food packaging is one of the main reasons. Hence in this work, a biopolymer barrier tested for its ability to reduce Aluminum leaching into food.

Chitosan, a biopolymer, that is non-toxic and biodegradable has film forming and antimicrobial properties can be used as a barrier to prevent the leaching of aluminum into food. Hot rice (approximately. 60°C) was used as a sample for the experiment. Hot rice was wrapped in aluminum foil and chitosan coated aluminum foil for 90 minutes. Aluminum content was detected using inductively coupled plasma-optical emission spectrometry (ICP-OES) for only rice; rice in aluminum foil and rice in chitosan coated aluminum foil and found to be 2.81ppm, 3.29 ppm and 2.87 ppm respectively. The results obtained implied that chitosan acts as a barrier suggesting that chitosan coated aluminum foil can be used to reduce the leaching of aluminum to food and hence minimise the adverse effects of aluminum intake on the body.

*Keywords:* Food packaging; Aluminum leaching; Aluminum toxicity; Chitosan biopolymer; ICP-OES

## Introduction

The food grown in fields as well as processed food needs to be distributed to consumers located in faraway places. It is a necessary to protect food from environmental factors without altering its physical or chemical nature during transport. Hence food packaging becomes an evident factor. The food packaging materials must be chosen selectively based on their health safety, energy, material costs, environmental impact and waste management processes. Food packaging has applications in maintaining the nature of processed food and improving shelf life. There are three kinds of significant

environmental influences on food: physically, biologically and chemically. Chemical protection is required against light, moisture and exposure to different gases mostly oxygen. Glass and metals can be used as chemical barriers. Biological protection is required against microorganisms, insects, pests and animals. It helps in delaying ripening and senescence. Odour is usually retained within packages, preventing access to the materials and maintaining optimum internal environment. Physical protection provides protection from causing mechanical damage. It prevents damage produced during shipping and transportation such as crushing and vibrations.

Hence the exploration of materials for food packaging with the required properties is needed. Glass is odorless and chemically inert and can serve as a food packaging material. It maintains the quality of food and flavor because it is impermeable to the gases and vapors. It can withstand high temperature and is acid-resistant. The brittle nature of glass limits its applications despite its recyclability. Although paper and paperboard products are not a perfect packaging materials for long term usage, they are used in primary packaging impregnated with waxes, resins etc.

Plastics are manufactured using polycondensation reaction or polyaddition of monomer units. They can be molded into different shapes, such as sheets and offer flexibility. They are cost effective, light weight and chemically resistant. The most commonly used materials include polyolefins, polyesters, polystyrenes etc. These materials are disadvantageous due to their differential permeability to light, gases and vapors and nonbiodegradability.

Metals such as aluminum, tin free steel, laminated and metallized films and tin plates are widely used examples in food packaging. Aluminum which is commonly used in cans, foils, and MLPs is a lightweight metal derived from bauxite ore. To improve its strength, magnesium and manganese are used more often. Due to its crucial properties such as resistance to corrosion, barrier against air, temperature, moisture due to the presence of aluminum oxide coating naturally, light weight, flexibility, malleability and recyclability, and ease of separation from other materials due to its non-magnetic nature, it is used in packaging of soft-drink, sea-food and hot foods (Kirwan., 2003). Despite the wide range of applications of aluminum in the food packaging industry, studies have shown that long term intake of aluminum can cause adverse effects due to aluminum toxicity. In this study, aluminum leaching into food and subsequent aluminum toxicity were investigated by using chitosan biopolymer as a barrier.

#### Aluminum as a Food Packaging Material

Aluminum foil is prepared by converting aluminum metal into very thin sheets by rolling which is immediately followed by annealing so that it is folded tightly. It is available in a wide range of thickness where thinner layers are used to wrap foods such as rolls and thicker layers are used for trays. It provides the same barrier properties to air, odor, light and moisture as aluminum. It also has a dead fold property which means that once fixed, the Aluminum foil takes the shape of a container. It can combine with many components such as paper, plastic and adhesives etc. (Kirwan., 2003; Kerry, 2012).

In addition to their many advantages aluminum provides several disadvantages: It is not suitable for cooking foods above 150°C since it loses its strength and other characteristics. Foil can also be punctured and torn out easily. This causes vapor and water permeability (Kerry, 2012).

Habian et al. (2011) in their historical research on the feasibility of using aluminum as a food packaging reported that increased aluminum content related to several diseases such as Alzheimer's disease and Parkinson's disease, renal failure, dementia and bone disease etc. It is stated that aluminum enters the brain by binding to the protein called transferrin which is an iron carrier. The majority of aluminum entering body binds to the transferrin receptor rich region of the brain which is also vulnerable to Alzheimer's disease. The authors mentioned that aluminum cookwares when used to cook foods rich in acid can increase metal leaching. Epoxy resins and plastics were used as barriers to prevent aluminum leaching. However it can cause epoxy resin or bisphenol A poisoning in the body (Habian., 2011).

Humans can be exposed various sources of aluminum including the diet, personal care products and aluminum based antiperspirant salts through dermal application. The increase of aluminum in type 1 human breast cyst fluids was analyzed to further investigate whether intake has any biological impact. Chronic exposure to aluminum inhibited Na<sup>+</sup>/K<sup>+</sup> ATPase activity. Aditionally it was found that the presence of aluminum can lead to iron dyshomeostatis. Sources other than personal care products and underarm cosmetics and aluminum in food and water were found to be major sources of exposure to breast cancer. Further investigation into the link between aluminum intake and the development of breast cancer are warranted to determine whether aluminum intake plays any role in combination with other environmental compounds (Darbre., 2011).

Turhan et al. (2006) conducted experiments to detect aluminum content in different kinds of meat such as beef, water buffaloes, mutton, turkeys and chickens wrapped in aluminum foil. Meat samples were heated in an electric oven at different conditions such as 150°C for 60min, 200°C for 40 min and 250°C for 20 min. Both the raw and cooked samples (3g) were taken and dried for 4 h at 125°C and heated at 500°C for 6-8 h until ash was obtained. The acid digested samples were cooled, filtered and stored in glass jars for analysis via AAS at 309.3 nm. It was evident that the maximum increase in aluminum content was in cooked food kept at the highest temperature. Therefore, they concluded that temperature over time acts as a major factor in leaching. Although there were traces of leaching, they state that this leaching is tolerable since the provisional tolerable daily intake of aluminum according to the WHO is 1mg /kg body weight per week. However, long term accumulation of aluminum in the body may cause severe problems (Turhan., 2006).

Low pH, temperature and salt content in food are the factors that affect the extent of aluminum into food. Fermo et al. (2020) studied aluminum content in meat and fish that are cooked in pans, wrapped in aluminum foil and wrapped in aluminum foil along with seasoning materials. The plants were cooked for 1 hour at 180°C. These samples as well as the raw samples were digested in 1:3 HN03: HCl on a hot plate for 1hr and then diluted. The samples were analyzed using ICP-OES and the surface of the foil was studied using SEM-EDS. The results showed that the aluminum content was greater in the fish and meat wrapped in aluminum and seasoning material than in the raw material and the other two samples however for beef the aluminum content was greater only in the wrapped food. The fat content may play a role in this process (Fermo., 2020). Inan-Eroglu et al. (2018) wanted to assess the aluminum content leached into different types of meat(beef, chicken and mutton) that were heated at different conditions as 150°C for 60 min, 200°C for 40 min, 250°C for 20 min and different types of foils. One is normal aluminum foil whereas the other is aluminum foil the side of which is paper. They acid digested food using HNO, in a microwave digester and diluted it to analyze Al content using ICP-MS. They concluded that leaching was more greater in aluminum foil than in paper. They also observed that aluminum leaching is dependent on fat content, pH and cooking temperature and time irrespective of the type of foil used (Inan-Eroglu.m 2018). In their studies, Dordevic et al. (2019) used different types of meats (beef, chicken and mutton) and Ertl et al. (2018) used marinated and nonmarinated samples and found that aluminum leaching was not as significant as the daily viable limit of consumption (Dordevic., 2019; Ertl., 2018). From different studies conducted using a wide range of food samples, it is evident that long term intake of aluminum leached due to usage in food packaging can lead to accumulation and potentially contribute to associated diseases.

#### Chitosan: A Potential Biopolymer for Food Packaging

Active packaging is a type of packaging which includes the interaction between environment, package and product which is aimed at increasing shelf life and maintaining shelf life and quality of the product. Environmental concerns about packaging materials lead to usage of renewable resources as an alternative source. Biopolymers have been extensively studied in last decades which incorporate special properties like antifungal, antioxidants and antimicrobial activity (Mitelut., 2015; Bégin., 1999). They act as a barrier against gases and vapours. Some of them are cellulose, chitin, lipid and protein based polymers. Chitosan stands next to cellulose in abundance. They are obtained by de-acetylation of chitin which is majorly available from shells of shrimp, crab, sea weeds etc. Since they are the by- product of sea-food industries, the cost of production is economically feasible. They are biodegradable, non-toxic and have excellent strength and elongation properties (Singh., 2015). Antimicrobial property of chitosan depends on external factors like target organism, pH and also internal factors like de-acetylation, molecular weight etc. the long positive charge present in chitosan due to the presence of amino acids, interact with negative charges in cell surface of target organisms thereby causing disruption and leakage

of proteins and contents of cell. Since the number of protonated amino acid increases with degree of de-acetylation, this acts as factor affecting antimicrobial property (Mitelut., 2015). Different types of chitosan films can be prepared using different methodologies. Chitosan can be dissolved using acetic acid and made as film. It can be blended with agar/cellulose/starch so that strength of material is increased. Many additives like potassium sorbate and nisin or edible oils like garlic oil can be used to increase antimicrobial property of film. The antimicrobial tests are generally conducted using agar diffusion method where certain amount of inoculums of organisms like Escherichia coli, bacillus cereus, listeria monocytogenes and staphylococcus aureus are seeded on it. The films are made into small discs and placed on media and they observed zone of inhibition (Tripathi., 2008; El-Hefian., 2012). Bégin et al. (1999) dissolved chitosan in 2% (w/v) acid like formic, citric, acetic and lactic acids along with HCl. They found out that the film thickness was more in ctric acid and lactic acid as compared to acetic acid containing films. Although they are thicker, they are brittle. Hence acetic acid is preferred while forming films (Bégin., 1999). Singh et al. (2015) carried out optimization of process of chitosan film formation using different concentrations of chitosan level, glycerol level and drying temperature in order to get different responses like thickness, solubility, density, water vapor permeability etc. they used chitosan 1.5- 2% and dissolved in 1%(w/v) acetic acid and glycerol 0.5-1% which acts as plasticizer. They were stirred on hot plate magnetic stirrer at 90°C and hen dried at varied temperatures. They found out that 2% w/v chitosan, 0.75% glycerol and drying temperature of 40°C for 48hrs were optimum conditions for development of good edible film (Singh., 2015).

## **Materials and Methods**

Aluminum foil (thickness 0.0023mm) and Chitosan (low MW extrapure, 10-150m.Pas, 90% DA) and Glycerol (glycerin, anhydrous, extrapure AR, MW 92.09) purchased from Sisco Research Laboratories Pvt. Ltd. Acetic acid glacial 99- 100% and Nitric Acid (min. 69% GR) purchased from Merck Specialities Pvt. Ltd.

#### Chitosan Coated aluminum Foil Preparation

Chitosan coated aluminum foil was prepared by coating chitosan solution on aluminum foil wrapped on the 15x15cm glass plate evenly. Chitosan solution was prepared by dissolving Chitosan (2% w/v) in aqueous solution containing acetic acid (1% v/v) and glycerol (0.75% v/v) as a plasticizer and heated in hot plate magnetic stirrer at  $90^{\circ}$  C for 10 minutes. The solution was cooled to room temperature. The prepared chitosan solution was coated onto aluminum foil using Hand Lay-Up method and incubated at  $50^{\circ}$ C for 48 hours (Singh., 2015).

#### Sample Preparation

Rice (40 g) taken as sample was cooked upto approximately  $60^{\circ}$ C. Food samples were wrapped in only aluminum foil which served as control and wrapped in prepared material as well, left still for 2 hours in room temperature. 2g of wrapped samples along with only rice were digested with 65% HNO $_3$ . For the food digestion nitric acid is used as all the nitric salts are soluble in water and  $H^2$ SO $_4$  or HCl are not used since they produce sulphate or chloride salts which are not soluble in water. The samples after digestion were centrifuged at 5000 rpm for 10 minutes. Using Whatman filter paper-1 filtrate is obtained which was analysed for Al content using ICP-OES (Analytical Research & Metallurgical Laboratories Pvt. Ltd).

## **Results and Discussion**

Chitosan solution was prepared by using acetic acid instead of lactic acid, which is structurally more rigid with reduced tendency to deform (Pavoni., 2019). Chitosan solution coated on aluminum foil by hand lay-up method was spread uniformly and found to be non-porous to significant extent as evident in figure 1.



**Figure 1:** Chitosan solution uniformly coated on Aluminum foil, incubated at 50°C for 48h.

The thickness of aluminum foil and Chitosan coated on aluminum foil measured with micrometer were found to be 0.023 and 0.024 mm respectively.

Upon wrapping the hot food sample (60°C) in the chitosan coated aluminum foil, the coated chitosan layer was found to stay intact. No peeling of chitosan layer from aluminum foil was observed. However, temperature of food is an evident factor that contributes to extent of peeling of chitosan layer.

Aluminum content in each sample including only rice; rice wrapped in aluminum foil and rice wrapped in prepared material analysed using APHA/ICP-OES were found to be 2.81, 3.29 and 2.87 ppm respectively as mentioned in Table 1.

Sample	Test Parameter	Unit	Test Result
Only Rice	Aluminum Content	Ppm	2.81
Rice in Al foil			3.29
Rice in chitosan coated Al foil			2.87

Table 1: Chemical Analysis of Samples for Al content by APHA/ICP-OES method (ARML Pvt. Ltd.).

The chitosan biopolymer layer acted as a barrier and significantly reduced leaching of aluminum into food upto 0.42 ppm. Turhan et al. (2006) found out that there is 89-378% increase in aluminum content in red meats and 76-215% increase in poultry after cooking which is comparable with our results where leaching is more in aluminum wrapped food than raw food and prepared material (Turhan, 2006). Although there are various studies conducted on aluminum leaching into food from aluminum foil, this study is progressive in terms of including a biopolymer barrier to prevent leaching.

## **Conclusion and Future Scope**

Food packaging is an ever-growing market in modern food industry as it plays a major role in protecting integrity of the food. Over the centuries, wide range of materials has been explored for food packaging considering abundance, safety and environmental impacts. aluminum as a substrate for food packaging has become popular due to its durability, low transportation cost and recyclability. Its barrier function against the migration of moisture, oxygen and other gases, and volatile aroma, as well as against the impact of light is generally higher than any plastic laminate material (Junianto., 2021). Chitosan, a polysaccharide originated from deacetylation of chitin is a potential food packaging material due to its particular physicochemical features, biodegradability, non-toxicity, biocompatibility, good film-forming properties, chemical stability, and high reactivity (Lamberti., 2007).

Aluminum being a widely used packaging material in food industries, its long term uptake due to leaching may cause accumulation and contribute to associated diseases. Hence, in our work, we emphasized on utilizing binding property of chitosan onto aluminum foil to act as a barrier preventing leaching into food. The reading obtained by the ICP-OES proved that there is significant reduction in the leaching of aluminum into the food when chitosan is coated on the aluminum foil. However, the result may vary depending on the food sample used, temperature and pH of food, thickness and porosity of chitosan film, thickness of aluminum foil, duration of food contact with the film and contamination during conduction and testing. Leaching can be further reduced by using adhesives along with chitosan solution to enhance binding to aluminum foil. This work could be further upscaled as a potential packaging material and can be used along with aluminum foil wide scale.

### **Conflict of Interest**

The Authors declare as No Conflict of Interest.

#### Supporting information

No supporting information.

#### **Funding Declaration**

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