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Screening and optimization of dairy and non-dairy based ingredients for development of composite biodegradable film

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Abstract

This research was conducted to assess the compatibility of natural and synthetic polymers to obtain a composite packaging film. The screening and optimization of all the ingredients were carried out singly and in combinations to evaluate the film formation. An excellent film formation was observed in which natural sourced polymers like whey protein concentrate, whey protein isolate, carrageenan, corn starch was partially replaced with synthetic polymer PVA, using glycerol as plasticizer. In another combination where only, natural polymers were used it was found that film formation occurred but developed cracks after drying. So, it was concluded that PVA being a water-soluble ingredient when combined with natural polymers exhibit excellent compatibility and easy processability by providing strong interfacial interactions by bonding.

Keywords: Screening, natural polymers, synthetic polymers, film, optimization, development

1. Introduction

Packaging of foods is an art as well as science required to prepare food items for safe transport, protection, convenience, storage or for sales elsewhere, from the point of production. Packaged food consumption has risen considerably in recent decades. With the annual growth rate of 5%, the global food packaging market in 2020 was assessed at \$1.9 trillion which is anticipated to be \$3.4 trillion by the year 2030 (Kan & Miller 2022) ^[9]. Food packaging not only aims at reducing food quality degradation but also prevents food spoilage from external conditions (oxidation, micro-organisms, moisture etc.) while transporting and marketing, thereby ensuring food quality and safety. Plastic films usage is more as they are easy to shape, stable and convenient to use. However, the disposal of plastic packaging film is not easy to recover as well as difficult to degrade. This has led to serious environmental issues. In order to tackle this issue the researchers focus has increased towards biodegradable packaging formulation. Biodegradable polymers can be classified as: Natural polymer materials (polysaccharide and protein) and synthetic polymers (polycaprolactone, polyvinyl alcohol, polylactic acid) (He *et al.*, 2025) ^[6].

Food packaging target is to ensure that throughout the supply chain the foods quality and safety are maintained. In this regard, the use of bio-degradable polymers has recently received increased attention in a wide range of applications. Despite various advantages, the use of biodegradable films for packaging of food is limited because of their poor barrier and mechanical aspects in comparison to plastic films.

So, in order to overcome these limitations, research and development of composite biodegradable packaging have risen, by utilizing natural polymers sources like sodium caseinate, whey protein in concentrate form, whey protein in isolate form, carrageenan, corn starch, glycerol for partial replacement of synthetic polymers like LDPE and PVA. Protein and polysaccharide-based films are commonly employed due to their biological compatibility, biodegradability, and versatile bioactive features (Saraiva *et al.*, 2023) ^[14]. Polysaccharides and proteins also enhance essential characteristics, such as film formation and functional durability, which are crucial in food packaging applications.

Carrageenans, are a group of water- soluble linear polysaccharides obtained from red seaweed. kappa-carrageenan (κ CA) has excellent film- forming capacity (Bhatia *et al.*, 2024) [3]. Among the synthetic polymers, polyvinyl alcohol (PVA) is an ideal choice for composite materials, as it integrates well with biopolymers to form stable and functional films (Alhussain *et al.* 2024) [1]. Whey protein is a cheese industry byproduct. Protein films have good mechanical attributes and excellent barrier to the gases (Hassan *et al.*, 2018) however, they have very low resistance to water, tend to be highly permeable to water vapor than any other biopolymers but they lack antimicrobial properties (Wihodo *et al.* 2013) [5, 19]. Keeping the above context in view, the present research was framed with the objectives of screening and optimization of dairy and non-dairy based ingredients for development of composite film, which can be applicable in ready to eat dairy and meat products.

2. Materials and Methods

2.1 Materials

Dairy based: Sodium caseinate, Whey Protein Isolate (WPI), Whey Protein Concentrate (WPC). Non-Dairy based (inclusive of natural and synthetic bio polymers): Carrageenan, Corn Starch, Low Density Poly Ethylene (LDPE), Poly Vinyl Alcohol (PVA), Poly Lactic Acid (PLA),

Polyhydroxy Alkanoate (PHA) and other required materials were purchased from the reputed firms. The magnetic stirrer cum hot plate was used for mixing the ingredients. The hot air oven was used for drying the film

2.2 Method

The screening and optimization of ingredients and process parameters were conducted for the preparation of the composite film with slight modifications using Sodium caseinate/WPI/WPC/ Carrageenan/Corn starch and glycerol as a plasticizer with partial replacement of synthetic polymers like LDPE, PVA, PLA, PHA individually or in combination (Sivakumar *et al.*, 2019; Vieyra *et al.*, 2011) [16, 18] various range of ingredients used for the preparation of film. The different levels tried were: Sodium caseinate (3-6%), WPC (4-6%), WPI (4-6%), Carrageenan (0.25-0.5%), Corn starch (4-6%), LDPE (0.5 & 1.0%), PVA (0.5 & 1%), PLA (0.5 & 1%), PHA (0.5 & 1%) along with incorporation of plasticizer glycerol in the level of 1-3%. The suitable combination and levels were selected for film preparation. While for film formation casting method was followed (fig. 1 & 2).

3. Method for preparation of Composite film

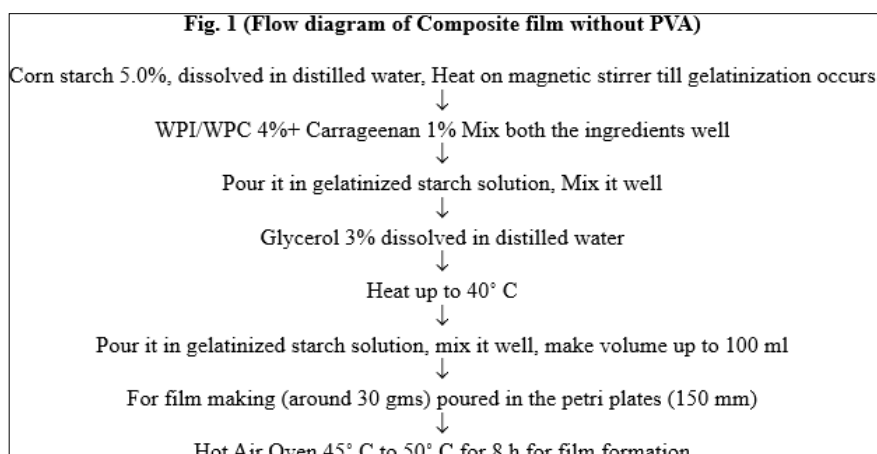


Fig 1: Flow diagram of Composite film without PVA

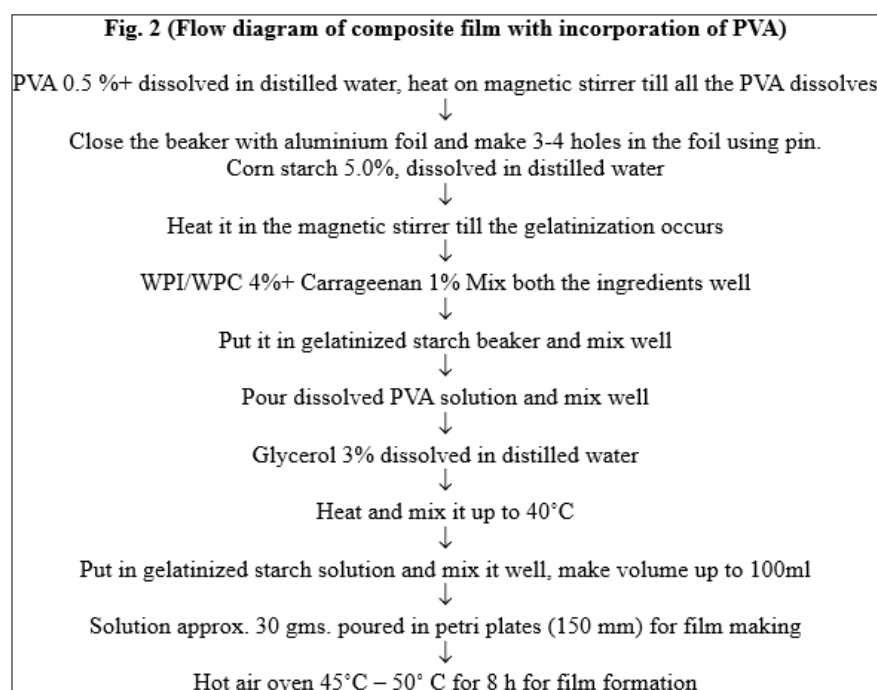


Fig 2: Flow diagram of composite film with incorporation of PVA

4. Results and Discussion

Different ingredients were used for the screening and

optimization for the development of composite biodegradable film. The observations are presented in Table 1 below

Table 1: Composite film formation trials

S. No.	Formulations	Glycerol (1%-3%)	Optimized	Remarks
1	a) LDPE (0.5-5.0%) b) LDPE (0.5 & 5.0%) + PVA (0.5 & 1%) c) LDPE (0.5& 5.0%) + PLA (0.5 & 1%)		Singly / All combinations	No film formation
2	a) PVA (0.5 & 1%)		All combinations	
3	a) Carrageenan (0.25- 2%)			
4	a) PLA (0.5 & 1%)			
5	a) PHA (0.5 &1%)			
6	a) Corn starch (4-6%)		5% corn starch, 2% Glycerol	Film formed
7	a) Sodium Caseinate (3-6%)		4% sodium caseinate, 2% Glycerol	
8	a) WPI (4-6%)/WPC (4-6%)/ Mixture of WPC&WPI (2% each)		Singly /All combinations	
9	a) LDPE (0.5 & 5.0%) PVA (0.5 &1%) PLA (0.5 &1%) PHA (0.5 & 1%) Corn starch (4-6%) WPI (4-6%)/ WPC (4-6%) Carrageenan (0.25-2%)/ Sodium Caseinate (3-6%)		PVA 0.5%, PLA 0.5%, PHA 0.5%, Corn Starch 5%, Carrageenan 1%, WPI &WPC 2% each, Sodium caseinate, 4%, glycerol 3%	Film not formed
10	a) PVA (0.5 &1%) Corn starch (2.5 & 5.0%) WPI (4-6%) WPC (4-6%) (Mixture of WPC & WPI) Carrageenan (0.5 & 2%)		PVA 0.5%, Corn Starch 5%, Carrageenan 1%, WPI & WPC (2% each) Glycerol 3%	Very good film formation
11	a) Corn starch (2.5 & 5.0%) WPI (4-6.0%) WPC (4-6%) (Mixture of WPC & WPI) Carrageenan (0.5 to 2%)	Corn starch 5% Carrageenan 1% WPI &WPC (2% each) Glycerol: 3%	Good film formation	

The film formation was assessed visually through physical appearance. The results revealed that the film formation was excellent with addition of PVA (fig. 3) to natural biopolymers (PVA 0.5%, Corn Starch 5%, Carrageenan 1%, WPI & WPC 2% each and Glycerol 3%) having smooth appearance which shows that the polymers were well dispersed and compatible. This film was easy to peel and did not torn. Adding the corn starch caused gelatinization, this as it used the excess water, i.e. granule disruption occurred in excess water. The glycerol (plasticizer) increased the solubility of film, lightness and water absorption giving the film a compact structure (Singh *et al.*, 2022) [15]. According to study of Yeşilyurt *et al.*, (2024) [20], the films with PVA/κCA had a smoother and homogeneous structure. Hosseini *et al.* (2024) [12] suggested that starch and PVA, exhibit polyoleic nature which results in high hydrophilicity, increasing the solubility of composite in the water. A good film formation can also be attributed to addition of WPI and WPC, as the solution heats protein denaturation occurs which increases the interaction between these chains and water molecules increasing their miscibility, this denaturation also increases the interaction between proteins and other polymers which helps in blend formation (Lara *et al.* 2019) [11]. A good film (fig. 4) was obtained using natural biopolymers without PVA (Corn starch 5%, Carrageenan 1%, WPI &WPC 2% each and Glycerol: 3%) but after drying it gave whitish and hazy appearance. Blooming and blushing optical defects were detected by Laohakunjit & Noomhorm (2004) [10] while conducting the study of the effect of plasticizers on rice starch based films Muscat *et al.* (2012)

[12] said that such defects occur when a plasticizer with low-molecular-weight tends to migrate on surface as a result of over plasticization and it was concluded that when the concentration of plasticizer is more than the polymer's capacity it leads to segregation of phase (exudation) causing expulsion of the compounds like glycerol or sorbitol. While it was found that no film formation occurred when LDPE, PVA are mixed, this observation can be linked to the immiscibility of these ingredients which resulted to a cracky film. In a study conducted by Ismail *et al.* (2010) [8] it was found that when a crosslinker (maleic acid) is not added the LLDPE/PVA blends exhibit holes as PVA forms agglomerates that detaches PVA from LLDPE. The observations are in accordance with other studies also where the LDPE/PVA blends form voids due to PVA agglomeration due absence of a coupling agent/ crosslinker (silane) Ameer *et al.* (2018) [2]. The formulations of some synthetic and natural polymers like PVA, PLA, PHA and carrageenan when tried for film formation singly led to no film formation. Carrageenan which is a polysaccharide when mixed with other biopolymers (PVA, corn starch etc) gave better film formation, but when tried singly fails to form the packaging material due to their hydrophilic nature. Carrageenan consists of alternating α-(1→4)-3,6-anhydro-D-galactose and β-(1→3)-D-galactose units having varying degrees of sulfation (Pal *et al.* 2018) [13] its sulfate content influences solubility and functionality. κ-carrageenan is a type of carrageenan with the lowest sulfate content (≈20% w/w) due to these properties it produces firm and brittle gels (Dellatre *et al.* 2011) [4].

Touswan *et al* (2026) ^[17] in a review concluded that polysaccharides when modified chemically introduces functional and hydrophobic groups which enhances their physico-chemical properties that makes them suitable as sustainable packaging materials.

5. Conclusion

In overall investigation of the composite films made from dairy and non-dairy ingredients, the results of the composite film made of PVA, corn starch, carrageenan, and mixture of WPI and WPC showed that the resulting composite had excellent film formation as compared to film without PVA which shows that presence of PVA significantly improves the miscibility of the solution creating a good network formation between PVA and other components of the film.

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