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Overall Migration and Quality Evaluation of Tomato Ketchup and Guava Jelly Packed in Biodegradable Films during Storage

B.N. Jadhav[1], G. Immanuel[2] and A. Chattree[3]

[1,2] Department of Processing and Food Engineering, Sam Higginbottom University of Agriculture, Technology and Sciences, 211 007, U.P., India
[3]Department of Chemistry, Sam Higginbottom University of Agriculture, Technology and Sciences, Prayagraj 211 007, U.P., India

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ABSTRACT

This study focuses on the effect of factors like the nature of the food, the type, time, and temperature of contact on overall migration (OM) from packaging material to food. The OM from the biodegradable film in contact with food products such as tomato ketchup and guava Jelly were investigated for six months using simulant 'B' as per IS 9845:1988. Under the room temperature storage condition for six months, the selected food products were checked for quality and stability. The maximum OM during the migration study was 4.24 mg/dm^2 for tomato ketchup and 3.48 mg/dm^2 for guava jelly, both of which were within the standards-permitted limits. Total Soluble Solids (TSS) and pH gradually increased whereas ascorbic acid was reduced in tomato ketchup during storage. The total sugar of jelly increased during the storage study from 46.92 to 66.10 %. The findings revealed that the OM rate was well within the specifications of the Bureau of Indian Standards (BIS) while the quality parameters of TSS, pH, and total sugar were altered after and during the three months of storage study. So, using this biodegradable film, tomato ketchup and guava jelly can be packaged safely for up to three months.

***Key words:** Biodegradable films, Food safety, Overall migration, Storage study*

Introduction

The food packaging sector is constantly working to meet consumer needs. The development of packaging materials that are suitable for food is facilitated by this need. For food packaging, polyethylene (PE), polystyrene (PS), and polyethylene terephthalate (PET) are the most often used plastic materials. A high number of chemical substances have been found in foodstuffs including additives, mycotoxins, and flavorings. Migration occurs when food and packaging materials come into direct contact, acting as a source of chemicals and other components (Arvanitoyannis and Kotsanopoulos, 2014). The quality and safety of the food are changed by this mass transfer. Food contact materials (FCM) shall not transfer their contents to foods at levels that could endanger human health or result in an undesirable change in the composition of food, according to a European Union (EU) regulation 1935/2004 (Silva *et al*., 2007). Plasticizers, monomers and oligomers, solvents are positively listed additives that

([1]Research Scholar, [2]Associate Prof., [3]Associate Prof.)

may migrate into foods by direct contact or through the free space in the interior of the package (Arvanitoyannis and Kotsanopoulos, 2014). This drives researchers and the plastic packaging sector to make sustainable biodegradable films. All packaging materials of plastic origin must clear the prescribed overall migration limit of 60 mg/kg or 10mg/dm^2 once tested according to IS 9845 using food simulant with no visible color migration, according to the Food Safety and Standards (Packaging) regulations, 2018. During the storage and processing condition oxidation and isomerization of lycopene takes place in the case of tomato ketchup (Kumar *et al*., 2015). However, under particular storage conditions, such as high temperatures and exposure to air and light jellies become unstable.

In this study, the simulant D (3% acetic acid) was used to accurately measure the overall migration levels into the food simulant as permitted by law, IS 9845: 1998. Estimating migration and evaluating the product quality of biodegradable films used in FCM during storage were the key goals. The results are addressed in terms of how various factors impact the migration rate.

Materials and Methods

Biodegradable films

For overall migration testing, a biodegradable film composed of LDPE and modified corn starch (MCS) was used. The virgin LDPE film was considered blank. The biodegradable film thickness ranged from 80 to 155 µ depending on the percentage of MCS content.

Sampling

The products chosen for testing were tomato ketchup and Guava jelly. Both the products were bought from the university cooperative store, weighed, and sealed with a heat impulse sealing machine (Model No. 300, Make: Sevana, Kochi, India), and kept at room temperature for six-months migration and storage study. Five samples of each product were prepared and analyzed. All the treatments were done in triplicate. The research was conducted at the Department of Processing and Food Engineering, SHUATS, Prayagraj, India.

Chemicals and standard solutions

All the reagents were of analytical quality. Rankem (Thane, India) supplied 3% acetic acid, Fehling's solution A and B.

Methods

Preparation of specimen for OM study

The film samples were rolled in concentric rings in the form of a coil with a surface area of 100 cm^2 on both sides. To completely immerse the sample, 100 ml of simulant was taken.

Procedure

At the test temperature, the cylindrical jar with a capacity of 500 ml was filled with 100 ml of preheated simulant. The test specimen (5cm×10cm×2 sides) was completely immersed in the simulant and shielded by a glass plate. The jar with the sample was immersed in the simulant for 30 min at 38 ^oC. The samples were removed with the help of a glass rod at the end of the test period. The samples were washed with a small amount of fresh simulant and combined with the extractants. By evaporating the extracted simulant by a hot plate, the volume was reduced to 10-15 ml. The concentrate was then transferred to a clean tared stainless-steel dish and washed three times with a small amount of fresh simulant before evaporating the simulant and dried in an oven at 100 °C. The residue was cooled in a desiccator for 30 minutes before being weighed to the nearest 0.1 mg until it reached a constant weight. The extractive was calculated in mg/dm^2. Without the sample, the blank was also performed. This demonstration was carried out by IS 9845:1988. The amount of extractive (Ex) was calculated by equation (1)

$$Ex(mg/dm^2)= \frac{M \times 100}{A} \quad .. (1)$$

Where

M = mass of residue in mg minus blank value, and

A = total surface area in cm^2 exposed in each replicate

Product quality assessment during storage

Packaged and stored tomato ketchup was evaluated for (i) TSS (ii) pH (iii) Ascorbic acid content. The total sugar content of guava jelly was also tested. It was determined for 30 days intervals for six months.

Determination of Total Soluble Solids

The refractive index of the test solution at 20 ^oC was measured by IS 13815: 1993 / ISO 2173: 1978. The prepared ketchup sample drop was placed on the prism, and an observation was made while it was illuminated. A dark line indicator measuring TSS in °Brix could be seen on the visible scale.

Determination of pH

The ketchup sample was homogenized, then an electrode was submerged in it, and adequate contact between the probe and the sample was ensured. When the meter reading was stable, a reading was taken. The test sample was subjected to three separate measurements; the extreme readings ought not to vary by more than 0.15 pH units. The arithmetic mean of the three readings was used to calculate the result.

Determination of Ascorbic acid

Determination of ascorbic acid was done by the (AOAC, 967.21) official method. In a 100 ml volumetric flask, added 50 g ketchup, 25 ml 3% metaphosphoric acid as a stabilizing agent, and diluted. Pipetted 10 ml in a small flask and 2.5 ml acetone was added to a small flask. Titrated for 15 seconds with 2,6-dichloro indophenol solution, or until a faint pink color remains.

The ascorbic acid content was determined by equation (2)

$$\text{Vitamin C (mg/100g)} = 20\,(V)\,(C) \qquad ..\,(2)$$

Where, V= ml indophenols solution, C = Vitamin C per ml indophenol solution

Determination of Total Sugar of Guava Jelly

Determination of total sugar was done by the AOAC 17th edition, 2000 method. The percentage of total sugar was calculated by the equation (3)

$$\text{Total sugar (\%)} = \frac{\text{Factor} \times 100}{\text{Titre}} \qquad ..\,(3)$$

Statistical analysis

All treatments were carried out in triplicates. SAS 9.1 was used to analyze variance (ANOVA) on the experimental data (SAS Institute Inc., Cary, NC, USA), which revealed the significance of the study. A significant difference between treatments was determined using Tukey's method at a level of significance ($p < 0.05$). All the data were presented as mean± standard deviation.

Results and Discussion

Food compatibility

Overall migration values for LDPE (T_0) and LDPE-modified corn starch (T_1, T_2, T_3, and T_4) film in 3% acetic acid simulant ranged from 0.12 to 4.24 mg/dm² for tomato ketchup and 0.10 to 3.48 mg/dm² for guava jelly, all well within the 10 mg/dm² limit, indicating suitability for food packaging applications, as shown in Tables 1 & 2. Migration is influenced by a variety of factors that can have a direct impact on the extent of migrant and rate. The substrate with the highest fat content, contact time, and temperature had the highest level of migration into food as mentioned by Triantafyllou *et al.* (2007). Higher migration rates are linked to thinner packages according to Nerin *et al.* (2007). The T_4 film samples, which were thicker in size, had a lower migration rate than the other treatments (T_1, T_2, and T_3). Experimental data showed that the rate of migration increased with storage time. When the packed ketchup made contact with the film, a leaching phenomenon occurred. In such cases, the migration phenomenon

Table 1. Food compatability of LDPE-MCS fims containing ketchup

Month	Overall migration (mg/dm²)				
	T_0	T_1	T_2	T_3	T_4
1	$0.12^d±0.015$	$0.61^a±0.015$	$0.59^{ab}±0.01$	$0.57^b±0.015$	$0.52^c±0.015$
2	$0.12^e±0.015$	$1.18^a±0.02$	$1.11^b±0.015$	$0.97^c±0.020$	$0.92^d±0.015$
3	$0.16^e±0.01$	$1.29^a±0.015$	$1.21^b± 0.01$	$1.15^c± 0.015$	$1.12^d±0.015$
4	$0.17^d±0.01$	$1.96^a±0.01$	$1.86^b±0.015$	$1.85^b±0.020$	$1.71^c±0.015$
5	$0.17^c±0.020$	$2.84^a±0.025$	$2.83^a±0.020$	$2.74^b±0.02$	$2.73^b±0.025$
6	$0.23^e±0.026$	$4.24^a±0.026$	$4.08^b±0.03$	$3.99^c±0.02$	$3.93^d±0.026$

All values shown are mean± standard deviations.
Various letters inside the columns clearly show statistically significant ($p < 0.05$) differences between samples.
Data with the same letter (a-e) within a row are not statistically different at a ($p < 0.05$) level.

occurs as a result of migrant diffusion, dissolution, and dispersion, or diffusion into food. As suggested by Lee *et al.* (2008), the system most commonly associated with this type of migration is the migration from plastic packages to liquid foodstuffs or moist solid products that come into direct contact with the packaging material. Jordan *et al.* (2003) revealed the characterization of aromatic profile in guava puree and essence. Arvanitoyannis and Kotsanopoulos, (2014) stated that in a volatile system, migration can be performed without contact between the package and the food, it may be affected by the contact. Under these conditions, the volatile substances migrate following diffusion or evaporation of migrant, desorption, and adsorption onto the products. The environment temperature was raised during the sixth-month migration study, which may have affected the overall migration rate.

Effect on Total Soluble Solids

The maximum TSS values of tomato ketchup observed at room temperature after a 6-month storage study was 31.04 (T_0), 32.04 (T_1), 34.05 (T_2), 36.01 (T_3), and 40.06 °Brix (T_4) as shown in Table 3. There is a possibility that acid hydrolysis of polysaccharides, particularly gums and pectin, may cause an increase in TSS with longer storage times (Luh and Woodroof, 1975). The experimental analysis demonstrated that tomato ketchup packed in T_1 and T_2 biodegradable films performed pretty well up to 3 months of storage at room temperature with TSS of 27.06 and 28.09°Brix, respectively which is based on national standards (BIS & FSS Regulations). Safdar *et al.* (2010) reported increased TSS contents of tomato paste during storage of 240 days at different temperatures.

Effect on pH

Among the majority of important variables to consider when evaluating tomato quality is pH, because acidity impacts the thermal processing parameters required to manufacture safe products (Kumar *et al.*,

Table 2. Food Compatibility of LDPE-MCS Films containing Jelly

Month	Overall migration (mg/dm^2)				
	T_0	T_1	T_2	T_3	T_4
1	0.10^b±0.015	0.35^a±0.015	0.34^a± 0.02	0.35^a±0.025	0.32^a±0.036
2	0.13^c±0.020	0.39^a±0.015	0.35^b±0.005	0.36^b±0.005	0.33^b±0.005
3	0.14^c±0.01	0.42^a±0.01	0.37^b±0.015	0.37^b±0.005	0.35^b±0.001
4	0.15^d±0.01	0.83^a±0.015	0.79^a±0.03	0.72^b±0.02	0.65^c±0.026
5	0.16^d±0.01	1.64^a±0.02	1.55^b±0.035	1.54^b±0.02	1.42^c±0.025
6	0.23^e±0.020	3.48^a±0.025	3.35^b±0.020	3.22^c±0.020	2.89^d±0.02

All values shown are mean± standard deviations.
If there are different letters in the columns, it means that the differences between the samples are statistically significant ($p < 0.05$).
Data with the same letter (a-e) within a row are not statistically different at a ($p < 0.05$) level.

Table 3. TSS of Tomato Ketchup Packed in Biodegradable Films

Month	TSS (°Brix)				
	T_0	T_1	T_2	T_3	T_4
0	25.22^a±0	25.22^a±0.01	25.22^a±0.005	25.22^a±0.01	25.22^a±0.01
1	26.04^c±0.06	26.11^c±0.015	27.04^b±0.04	27.29^b±0.33	29.11^a±0.11
2	26.07^e±0.06	27.09^d±0.08	28.07^c±0.06	28.64^b±0.036	30.01^a±0.032
3	27.06^e±0.05	28.09^d±0.08	29.07^c±0.07	30.03^b±0.04	32.07^a±0.068
4	28.08^e±0.08	29.06^d±0.05	30.06^c±0.052	30.65^b±0.045	32.05^a±0.05
5	30.06^c±0.05	31.04^c±0.04	33.04^b±0.04	34.06^{ab}±0.05	34.72^a±1.10
6	31.04^e±0.05	32.04^d±0.04	34.05^c±0.043	36.01^b±0.01	40.06^a±0.05

All values shown are mean± standard deviations.
If there are different letters in the columns, it means that the differences between the samples are statistically significant ($p < 0.05$).
Data with the same letter (a-e) within a row are not statistically different at a ($p < 0.05$) level.

2015). During room temperature storage an increasing pH trend was observed for all the samples packed in various treatments (T_0 and T_1, T_2, T_3, and T_4) as in Table 4. The highest pH values recorded at room temperature after a 6-month storage study were 3.72 (T_0), 3.80 (T_1), 3.89 (T_2), 4.15 (T_3), and 4.30 (T_4), as reported. The rising trend in the pH values of ketchup packed in T_3 and T_4 biodegradable films exceeded the standard pH value limit of 3.80 - 4.0. These films were made up of 15% and 20% MCS coupled with LDPE, which had shown an inclination for increased water activity, indicating that they were unsuitable for ketchup packaging. The experimental analysis demonstrated that the pH value increased as a result of the storage state. The pH of all samples packaged in biodegradable films of various treatments is directly connected to the acidity. The acidic pH levels obtained throughout the experimental study are important that influence shelf-life quality by confining the microbiota to acid-tolerant microorganisms (Bracket, 1994).

Effect on Ascorbic Acid

Ascorbic acid is susceptible to oxidation when exposed to oxygen. Table 5 shows the impact of storage on ketchup' ascorbic acid content. During room temperature storage, it was observed that the amount of ascorbic acid in the samples packed in different treatments (T_0 and T_1 to T_4) declined as the storage period increased. The maximum amount of ascorbic acid ketchup during storage was found to be 35.76 (T_0), 35.23 (T_1), 33.46 (T_2), 31.43 (T_3), and 29.46 mg/100g (T_4) after the first-month storage study. Koh *et al.* (2012**)** revealed ascorbic acid decreases with prolonged storage, with only 13% remaining at 12 months. The degradation of ascorbic acid may be influenced by oxygen. This could be due to ascorbic acid being oxidized to dehydroascorbic acid, which is then degraded to 2,3-

Table 4. pH of Tomato Ketchup Packed in Biodegradable Films

	pH				
Month	T_0	T_1	T_2	T_3	T_4
0	3.48 ^{a}±00	3.48 ^{a}±0.01	3.48 ^{a}±0.005	3.48 ^{a}±0.01	3.48 ^{a}±0.01
1	3.5^c±0.01	3.51^c±0.01	3.57^b±0.015	3.59^b±0.01	3.66^a±0.020
2	3.50^d±0.011	3.53^d±0.01	3.61^c±0.011	3.65^b±0.01	3.7^a±0.01
3	3.57^e±0.005	3.61^d±0.01	3.67^c±0.01	3.7^b±0.01	4.05^a±0.007
4	3.63^e±0.01	3.69^d±0.005	3.74±0.01	4.02^b±0.01	4.10^a±0.02
5	3.65^e±0.005	3.73^d±0.01	3.79^c±0.01	4.10^b±0.01	4.19^a±0.02
6	3.72^e±0.015	3.80^d±0.005	3.89^c±0.01	4.15^b±0.005	4.30^a±0.01

All values shown are mean± standard deviations.
If there are different letters in the columns, it means that the differences between the samples are statistically significant ($p< 0.05$).
Data with the same letter (a-e) within a row are not statistically different at a ($p < 0.05$) level.

Table 5. Ascorbic Acid of Tomato Ketchup Packed in Biodegradable Films

	Ascorbic Acid (mg/100g)				
Month	T_0	T_1	T_2	T_3	T_4
0	36.12^a±0	36.12^a±0.01	36.12^a±0.005	36.12^a±0.01	36.12^a±0.01
1	35.76^a±0.58	35.23^a±0.20	33.46^b±0.50	31.43^c±0.40	29.46^d±0.50
2	26.16^a±0.15	24.46^b±0.45	23.16^c±0.20	21.03^d±0.05	19.33^e±0.41
3	21.2^a±0.2	18.96^b±0.05	18.5^b±0.45	16.4^c±0.36	13.23^d±0.20
4	16.06^a±0.11	16.33^a±0.30	15.13^b±0.15	13.23^c±0.25	11.03^d±0.057
5	15.13^a±0.23	14.2^b±0.26	12.23^c±0.20	11.16^d±0.15	10.03^e±0.05
6	13.13^a±0.15	13.4^a±0.36	11.1^b±0.1	10.03^c±0.05	10.36^c±0.35

All values shown are mean± standard deviations.
If there are different letters in the columns, it means that the differences between the samples are statistically significant ($p< 0.05$).
Data with the same letter (a-e) within a row are not statistically different at a ($p < 0.05$) level.

Table 6. Total Sugar of Jelly Packed in Biodegradable Films

Month	Total Sugar (%)				
	T_0	T_1	T_2	T_3	T_4
0	46.62 ^{a}±0	46.62 ^{a}±0.01	46.62 ^{a}±0.005	46.62 ^{a}±0.01	46.62 ^{a}±0.015
1	46.92^{d}±0.01	46.94^{d}±0.01	46.98^{c}±0.01	47.06^{b}±0.02	47.1^{a}±0.01
2	48.12^{e}±0.01	48.19^{d}±0.01	48.29^{c}±0.02	48.91^{b}±0.02	49.10^{a}±0.005
3	56.90^{d}±0.01	56.96^{c}±0.01	57.12^{b}±0.02	57.29^{a}±0.02	57.32^{a}±0.02
4	59.91^{e}±0.01	60.17^{d}±0.01	60.35^{c}±0.01	60.48^{b}±0.011	60.52^{a}±0.015
5	63.11^{e}±0.011	63.28^{d}±0.01	63.92^{c}±0.02	64.11^{b}±0.015	64.34^{a}±0.01
6	65.16^{e}±0.011	65.46^{d}±0.01	65.70^{c}±0.01	65.91^{b}±0.09	66.10^{a}±0.01

All values shown are mean± standard deviations.
If there are different letters in the columns, it means that the differences between the samples are statistically significant ($p < 0.05$).
Data with the same letter (a-e) within a row are not statistically different at a ($p < 0.05$) level.

diketogulonic acid, and eventually to furfural compounds (Kuchi *et al.,* 2014). However, ascorbic acid is an unstable compound under certain storage conditions, such as high temperatures and exposure to air and light. It has been reported that numerous breakdown reactive chemicals developed as a result of vitamin C degradation, and these compounds may mix with amino acids, resulting in the creation of dark pigments (Duru *et al.,* 2011).

Effect on Total Sugar of Jelly

Sugars are the most important elements of fruit products; they are necessary for the flavor of the food products and act as natural food preservatives (Pavlova *et al.,* 2013). The total sugar of jelly increased during the six-month room temperature storage study from 46.92 to 65.16% (T_0), 46.94 to 65.46% (T_1), 46.98 to 65.70% (T_2), 47.06 to 65.91% (T_3), 47.10 to 66.10 % (T_4) tabulated in Table 6. Similar results were reported by Kuchi *et al.,* (2014) for guava jelly bars. The total sugar content of guava jam increased from 63.39 to 63.56 % during storage durations of 0 and 60 days, respectively in agreement with Kanwal *et al.,* (2017) for guava jam. The conversion of starch and other insoluble carbohydrates into sugars is responsible for the rise in total sugars (Kanwal *et al.,* 2017).

Conclusion

In terms of overall migration, biodegradable films can be used as food packaging material. Migration data were analyzed, and they demonstrated compatibility with food products while adhering to legislation and food safety standards. During a product storage study, it was revealed that biodegradable films (T_1, and T_2) can be used as a packaging material for tomato ketchup, while guava jelly was found to be in good condition for up to three months. It is possible to conclude that biodegradable films T_1 and T_2 could be used as food packaging material for the aforementioned food products.

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Conflict of Interest

Hereby, the author declares that in the current study there is no conflict of interest.

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