



Development Of Edible Starch-Based Paper

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Abstract

The study is aimed to develop an edible film from maize and rice flour. The film is prepared using corn starch, CMC, MC, LG, and glycerol in various combinations. The 15% glycerol showed excellent binding and elasticity qualities. Broken rice flour inclusion resulted in a significant reduction in fractures and stickiness. The edible paper made from starch (3%), rice flour (22%), sodium carboxymethyl cellulose (0.5%), microcrystalline cellulose (0.5%), glycerol (20%), liquid glucose (5%), and xanthan gum (1%) had good physical and structural qualities. The thickness of the formed paper was $138 \pm 2 \mu\text{m}$, with a solubility of 89-92 percent. Edible paper is slightly diverse in character, according to SEM data. The edible paper is amorphous in nature, according to the X-ray diffraction analysis.

Keywords: Corn starch, edible paper, glycerol, rice flour, xanthan gum.

INTRODUCTION

Investigation into Food packaging is critical for developing unique packaging solutions not only for preserving food but also for marketing products at a higher value. This emphasises the freshness of the product, health issues, and food product quality traceability (Keränen et al., 2021). Packaging plays an important part in food safety since it protects and functions as a barrier against microorganisms from outside sources (Kraśniewska et al., 2020). Packaging is the most significant aspect of food processing, preservation, storage, and distribution, and it also improves a person's living standard (Han et al., 2021).

The packaging market is expected to increase at a CAGR of 3.92% between 2022 and 2027. The market is expected to grow by USD 223.96 billion. Cans, glass bottles, paper bags, and plastics are among the most extensively used packaging materials in the food sector (Calva-

Estrada et al., 2019). Consumer choices have a big influence on the evolution of different sorts or types of food packaging materials (Boesen et al., 2019).

Apart from these packaging materials the local vendors and food serving caterers are seen to use newspaper and/or related materials. These paper-based materials are coming directly in contact with food (Deshwal et al., 2019). The transfer of ink from a newspaper to food may have an impact on the dish's quality and safety. It eventually has an impact on a person's health (Jadhav et al., 2020). Because of the tight link between oily food and newspapers, there is a greater possibility of components being transferred through the oil used to cook food, which acts as a conduit for ink constituent transmission (Baele et al., 2020). The Food Safety and Standards Authority of India (FSSAI), which is part of the Ministry of Health and Family Welfare, has proposed packaging legislation that prohibits the use of newspaper or recycled plastic for packaging, conveying, or distributing food (Mehdi et al., 2019).

When a food product is wasted, the packaging is also discarded, adding to the environmental burden. In this context, it appears that reducing food and packaging waste implies using more packaging rather than fewer, or replacing oil-based resources with renewable resources (Brandao et al., 2021). Aside from reducing the negative burden of packaging resources and waste management, sustainable food packaging enhances its positive usage benefit, which is the decrease of food losses and waste (Wang et al., 2021). This is accomplished primarily through meeting food criteria to protect food quality and safety across the supply chain, with a focus on distribution and consumption. Considering the product and its packaging as a whole system is thus essential for optimising the overall sustainability of food/packaging systems (Chen et al., 2020).

A study on the development of edible packaging material was conducted to reduce the danger of food poisoning caused by the usage of newspaper / printed paper. The goal of this study is to create an edible packaging material that may be

used as a food serving material.

MATERIALS AND METHODS

The raw materials such as starch, rice flour was purchased from local shop in Bangalore, all the chemicals and other ingredients required for the formation of film, analysis of raw material, and analysis of final product were obtained from the Department of Food Technology, Jain University, Bangalore, India.

PREPARATION OF EMULSION

Food-grade sodium carboxymethyl cellulose was prepared by dissolving 0.5 g in 100 ml of water followed by boiling to get clear solution. Beforehand, a 1% xanthan gum was prepared through the application of heat. The edible paper was prepared using the approach described by Robert in 2015. Corn starch (5%) was combined with 50 mL of water and followed by heating for 15 minutes. After adding the 0.5 percent CMC, 1% xanthan gum and 0.5 percent MCC, the mixture was heated for another 5 minutes. To retain the suppleness of the packaging material, 5ml of glycerol has been added to the solution. The solution was cooled before being placed into the petri dish and dried for 24 hours at 50°C.

FILM CHARACTERIZATION

THICKNESS

A hand-held micrometer was used to measure the thickness of the created paper material. The sample was placed between the spindle and anvil of the micrometer, and the distance was reduced by adjusting the ratchet until the paper was fixed/ clamped, at which point no further adjustments were required. A total of ten readings were obtained at various locations on the paper material and the average value was calculated. Micrometer thickness is a measurement of how thick something is (Arfat et al., 2017).

FILM DENSITY

By dividing the weight of the sample by the weight of the paper volume, the density of the created paper was estimated. The area of the paper was multiplied by the thickness of the paper to calculate the volume of the paper (Sun et al., 2017).

$$\text{Density} = \frac{\text{Wt. of the sample (g)}}{\text{Volume of the paper (cm}^3\text{)}}$$

$$\text{volume of the paper} = \text{area of the paper (cm}^2\text{)} \times \text{thickness of the paper (cm)}$$

The starting weight of the sample was used to determine the moisture content of the created paper, and the samples were held in an oven at 90 °C until the constant weight of the sample was reached. It is then placed in desiccators to chill before being weighed (Deshwal et al., 2019).

Moisture content (%) =

$$\frac{\text{Initial mass of paper} - \text{Final mass of the paper}}$$

$$\times 100$$

SOLUBILITY OF EDIBLE PAPER

The solubility of the edible paper was determined using the Thawien, 2006 method with minimal changes, and by cutting the paper into three replicates of 4 X 4 cm size (Bourtoom et al., 2006). The cut pieces of paper were weighed before being placed in a beaker with 15ml of distilled water. The solution was then filtered and the residue was collected using Whatman #1 filter paper while it was agitated continuously at 22 °C. In a hot air dryer, the leftovers were dried (Kim et al., 2015).

$$\text{Solubility} = \frac{\text{Initial wt. of the sample (g)} - \text{Wt. of undissolved residues of the paper (g)}}{\text{Initial wt. of the sample (g)}}$$

WATER UPTAKE OF FILM

The formed edible paper was cut into lengths of 4x4cm with each of three replicates, and then immersed in water at room temperature for 5 minutes, 10 minutes, 15 minutes, 20 minutes, 25 minutes, and 30 minutes, and the water uptake capacity was measured by weighing them and expressed as a percentage (Setiani et al., 2013).

SCANNING ELECTRON MICROSCOPE (SEM)

Scanning electron microscopy was utilised to investigate the morphology and surface features of the produced paper. Once the produced paper substance is complete, it is connected to a double-sided carbon tape that has a tiny layer of noble metal on one side. SEM with a 5 KV accelerating voltage was used to examine the surface features and morphological parameters of the paper samples (Ibrahim et al., 2019).

X-RAY DIFFRACTION (XRD)

X-ray diffraction is a technique for determining the phase of a crystalline material and obtaining information on the dimensions of the unit cells (Yang et al., 2022). The diffraction consists of three main components: an X-ray tube, an X-

ray detector, and a sample holder (Falsafi et al., 2022). The X-rays are produced in the cathode ray tube by applying heat to produce electrons, which are then accelerated towards the target by applying voltage to the electrons. When enough energy is produced, the inner shell electrons of the material targeted are dislodged, resulting in the characteristic X-ray spectrum (Manner et al., 2014).

PRELIMINARY TRIALS WITH CORN STARCH

The corn starch was taken into consideration for the development of edible paper. The first edible paper was prepared used starch (3%), CMC (0.5%), MC (0.5%), glycerol (5%) and LG(5%). Developed paper is shown in **Fig.1**



Fig.1 Pictorial View of Preliminary Trial

The combination of above-mentioned components was taken for trial and was found that the surface of the film was not uniform. Cluster formation was observed with reduction in the size of the film. Also, cracks were observed at very higher level. The reason for not forming the film was due to less concentrations of starch. Starch provides a base and strength to edible paper. There was no proper cross linking between starch and other components. Similar study was reported by Buonocore *et al*, 2003 where the author had mentioned the cross linking can be increased by increasing the concentration of glyoxal.

EFFECT OF STARCH

Based on the second trial conducted, slight modifications were made in the concentration of starch by increasing it to 5% and 8% respectively from 3% starch. The combination is shown in **Table 1**

Table 1 Combination of Starch for Development of Paper

| Sl No. | Starch (%) | CMC (%) | MCC (%) | Glycerol (%) | LG (%) |
|--------|------------|---------|---------|--------------|--------|
| 1. | 5 | 0.5 | 0.5 | 5 | 5 |
| 2 | 8 | 0.5 | 0.5 | 5 | 5 |

The structure and appearance of the prepared film were not observed to be significantly affected by the variation in starch concentration. The strength of the paper was determined to be good at 8% starch. The paper became more elastic. The paper is shown in **Fig. 2**.



Fig. 2 Pictorial View of Developed Paper with 5 and 8 % Starch

However, the issue with cracking persisted. Poor cross-linking was still discovered. Due to the uneven moisture evaporation, the paper between them began to fracture. The lower glycerol content contributed to this brittleness as well. A similar study that addressed the value of glycerol was published.

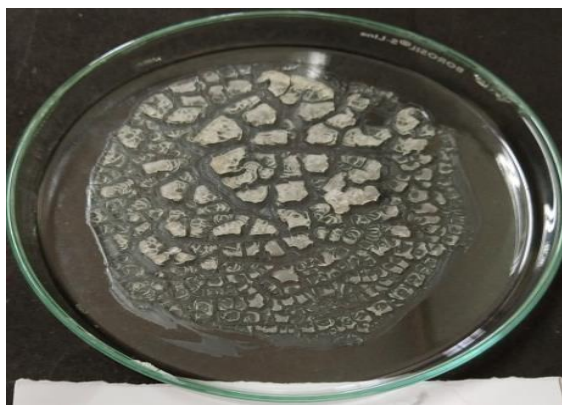
EFFECT OF STARCH AND GLYCEROL

While holding sodium carboxymethyl cellulose, microcrystalline cellulose, and liquid glucose constant, various ratios of starch and glycerin were measured. **Table 2** displays the various combinations. Glycerol concentration ranged from 10% to 15%, whereas starch concentration ranged from 5 to 8%.

Table 2. Combination of Starch and Glycerol for Development of Paper

| SI No. | Starch (%) | CMC (%) | MCC (%) | Glycerol (%) | LG (%) |
|--------|------------|---------|---------|--------------|--------|
| 1 | 5 | 0.5 | 0.5 | 10 | 5 |
| 2 | 5 | 0.5 | 0.5 | 15 | 5 |
| 3 | 8 | 0.5 | 0.5 | 10 | 5 |
| 4 | 8 | 0.5 | 0.5 | 15 | 5 |

Although the film had surface fractures and lacked the elastic strength of other experiments, it was discovered that the mixture of 0.5 ml and 8% starch produced a transparent film.

**Fig. 3 Pictorial View of Paper with 10% Glycerol and 5% of Starch****Fig.4 Pictorial View of Paper Developed with 10% of Glycerol and 8% of Starch**

Even after raising the concentration from 5 to 10%, cracks could still be seen in the paper made with 5% & 8% of starch and 10% of glycerol. Poor cross bonding, which results from insufficient glycerol, causes the remaining irregular structure. There was a little shift in appearance when the amount increased from 10% to 15%. When compared to the paper with 10% glycerol, the surface was smoother. The paper made with 15% glycerol has excellent translucency. The difficulty in peeled formed paper was the biggest issue. It became overly sticky as a result. More glycerol and starch boosted the sticky qualities.



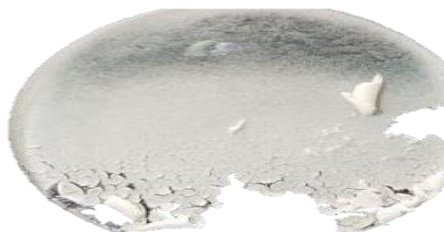
Fig.5 Pictorial View of Paper Developed with 15% of Glycerol and 5% of Starch**Fig.6 Pictorial View of Paper Developed with 10% of Glycerol and 8% of Starch****EFFECT OF STARCH AND RICE FLOUR**

Based on the findings, rice flour was chosen because, despite every combination, the right paper could not be produced. The problems with breaking and stickiness persisted. As shown in **Table 3**, various combinations of starch, rice flour, CMC, MCC, glycerol, and LG were used.

Table 3. Combination of Starch and Rice flour for Development of Paper

| Sl.No. | Starch (%) | RF (%) | CMC (%) | MCC (%) | Glycerol (%) | LG (%) |
|--------|------------|--------|---------|---------|--------------|--------|
| 1 | 3 | 22 | 0.5 | 0.5 | 15 | 5 |

According to **Fig. 7**, paper made its first appearance. The created paper was naturally dried. The cracks did not develop. The paper's tensile strength was subpar. The propensity for paper to roll and crumble was discovered. The plasticizer property of rice flour was observed to minimise the stickiness of created paper. The appropriate uniform structure was found because of crystallinity.

**Fig. 7. Pictorial View of Paper Developed with 15% of Glycerol, 3% of Starch and 22% Rice Flour****EFFECT OF GLYCEROL**

After formation of paper due to rolling and crumbing tendency of developed paper, a decision on increasing the glycerol percent and introducing the xanthan gum was taken.

Table 4. Combination of Xanthan Gum and Glycerol for Development of Paper

| Sl. No. | Starch | RF | CMC | MCC | Glycerol | LG | XG |
|---------|--------|----|-----|-----|----------|----|----|
| 1 | 3 | 22 | 0.5 | 0.5 | 20 | 5 | 1 |

In order to boost the film's flexibility and binding power, the concentration of glycerin was raised. It was discovered that there was a uniformity in the thickness of the material and decreased breakages when xanthan gum was employed to bind the suspension and form the edible paper-like structure. It was discovered that the paper quality and binding capacity were good at a 20% glycerol content. A paper material that is consistent with nature, has a good texture, and binds the particles to form a single paper material was created with the addition of 1% Xanthan gum.

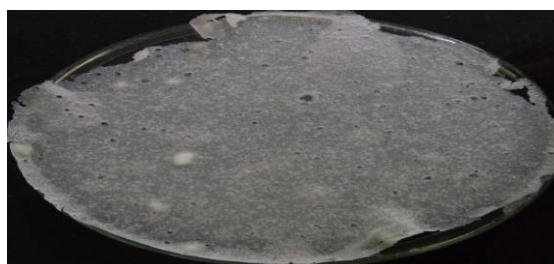


Fig. 8. Pictorial View of Developed Edible Paper**RESULT AND DISCUSSION****Preliminary Trials with Corn Starch**

The invention of edible paper took corn starch into account. Utilizing starch (3%), CMC (0.5%), MC (0.5%), glycerol (5%), and LG (5%), the first edible paper was created. Developed paper is shown in **Fig.1**

It was revealed that the film's surface was not consistent when the components were joined. Cluster formation was observed when the film was shrunk. Additionally, there were found to be much more cracks. Lower starch concentrations were to blame for the lack of film formation. Starch provides stability and strength to edible paper. The necessary cross-linking of starch and other ingredients was lacking. Similar studies were published in 2003 by Buonocore et al., who claimed that increasing the concentration of glycerol could boost cross-linking. [45].

Effect of Starch

Based on the second trial conducted, slight modifications were made in the concentration of starch by increasing it to 5% and 8% respectively from 3% starch. The combination is shown in **Table 1**

The structure and appearance of the prepared film were not observed to be significantly affected by the variation in starch concentration. The strength of the paper was determined to be good at 8% starch. The paper became more elastic. Fig. 4.2 displays the paper.

The proper gelatinization was what caused the film to form. However, the issue with cracking persisted. Poor cross-linking was still discovered. Because of this, there was uneven moisture evaporation, which caused the paper to break between sheets. The lower glycerol content contributed to this brittleness as well. A similar study that addressed the value of glycerol was published [46].

Effect of Starch and Glycerol

While maintaining sodium carboxymethyl cellulose, microcrystalline cellulose, and liquid glucose constant, various ratios of starch and glycerin were measured. In **Table 4.2**, the various combinations are displayed. Glycerol concentration ranged from 10% to 15%, whereas starch concentration ranged from 5 to 8%.

Although the film had surface fractures and lacked the elastic strength of other experiments, it was discovered that the mixture of 0.5 ml and 8% starch produced a transparent film.

Even after raising the concentration from 5 to 10%, cracks could still be seen in the paper made with 5% & 8% of starch and 10% of glycerol. Poor cross bonding, which results from insufficient glycerol, causes the remaining irregular structure. There was a little shift in appearance when the amount increased from 10% to 15%. When compared to the paper with 10% glycerol, the surface was smoother. The paper made with 15% glycerol has excellent translucency. The difficulty in peeled formed paper was the biggest issue. It became overly sticky as a result. More glycerol and starch boosted the sticky qualities.

Effect of Starch and Rice Flour

Based on the findings, rice flour was chosen because, despite every combination, the right paper could not be produced. The problems with breaking and stickiness persisted. As shown in **Table 3**, various combinations of starch, rice flour, CMC, MCC, glycerol, and LG were used.

According to Fig. 4.7, paper had its first appearance. The created paper was naturally dried. The cracks did not develop. The paper's tensile strength was subpar. The propensity for paper to roll and crumble was discovered. The plasticizer property of rice flour was observed to minimize the stickiness of created paper. The appropriate uniform structure was found because of crystallinity.

Effect of Glycerol

After developed paper began to roll and crumble, leading to the development of paper, it was decided to add xanthan gum and raise the glycerol percentage. In order to boost the film's flexibility and binding power, the concentration of glycerin was raised. It was discovered that there was a uniformity in the thickness of the material and decreased breakages when xanthan gum was employed to bind the suspension and form the edible paper-like structure. It was discovered that the paper quality and binding capacity were good at a 20% glycerol content. A paper substance that is even in nature, has a good texture, and binds the particles together to form a single paper material can be created by adding xanthan gum at a concentration of 1%.

Physical and Hydration Analysis of Developed Paper

The final edible paper was examined for its physical and structural qualities and taken for more research.

Film thickness

The average thickness of the edible paper was found to be $138 \pm 2 \mu\text{m}$. It was observed that varying thickness of the paper is due to un-uniform distribution of the solution during drying. It may be due to addition of different raw material in the paper and variation in the preparation of the paper.

Water absorption capacity

As the paper material met the water, the capacity for water absorption gradually increased. It was discovered that with an increase in the paper material's duration spent in the water being retained, as shown in the table in **Table 7**. After 30 minutes, it was discovered that there had been a greater amount of water absorbed into the paper, indicating that the generated edible substance had good degradability.

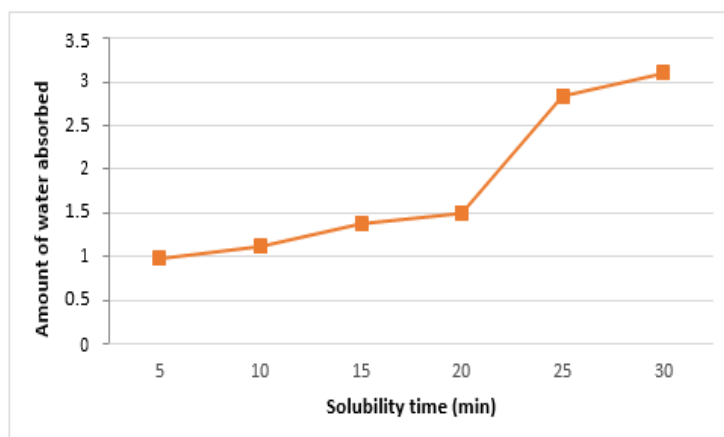


Fig. 9 Graphical Representation of Water Absorption

Solubility

It was discovered that the edible paper has little resistance to the water vapour. Due to the paper's poor water resistance and higher diffusion rate, it dissolved easily when in contact with water. The solubility of the paper substance, which ranged from 89 to 92%, was plainly visible. Thus, it was determined that the edible paper substance had a higher solubility rate.

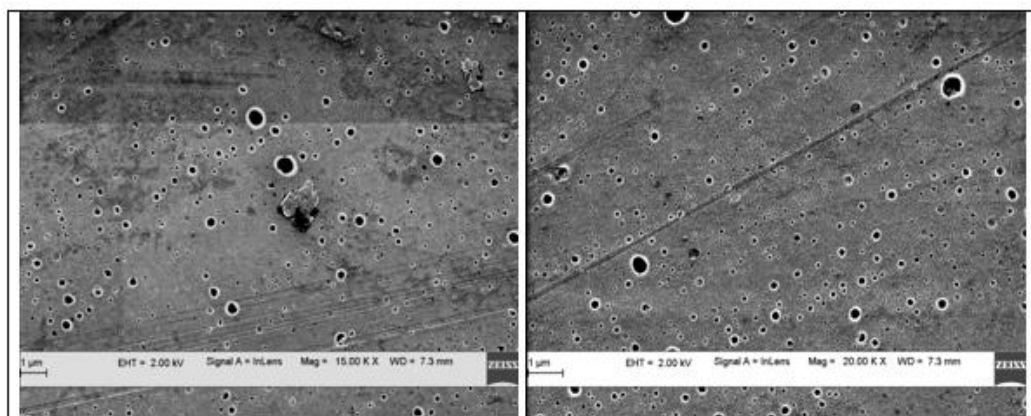
Table 5. Solubility of the Edible Paper

| SI No. | Initial weight of the S | Weight of the un-dissolved sample (g) | % of solubility |
|--------|-------------------------|---------------------------------------|-----------------|
| 1 | 5 | 0.55 | 89.0 |
| 2 | 5 | 0.40 | 92.0 |
| 3 | 5 | 0.48 | 90.4 |

STRUCTURAL PROPERTIES

Scanning Electron Microscopy (SEM)

The morphology of developed edible paper was studied using scanning electron microscopy (SEM) in **Fig.10**. It was discovered that edible paper is inherently rough, uneven, and slightly heterogeneous. Additionally, white circular dots are seen on the paper's surface. This is consistent with the components' weak contact and aggregation during the production of paper. Overall, the addition of various ingredients to the starch and rice flour has a significant impact on the morphology showing internally circular patches fractures, which may be caused by the xanthan gum's dispersion.



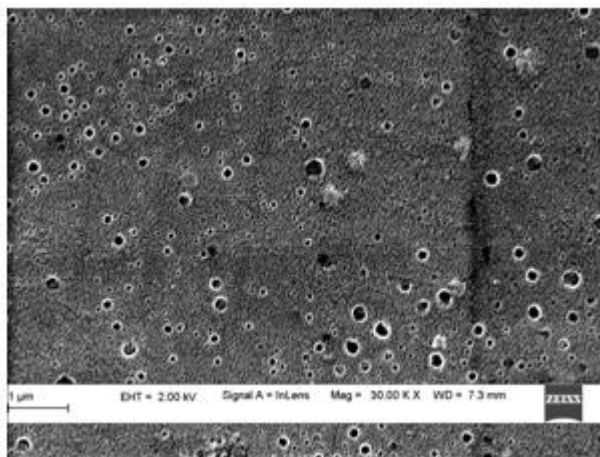


Fig. 10 SEM Micrographs of Edible PaperX- ray diffraction (XRD)

To determine the amorphous and crystallinity of the pure and processed edible paper sample, an X-ray diffraction investigation was carried out. Fig. 11 displays the X-ray diffractogram of the ready samples. The distinctive 2 peaks at 19.4° , 40.6° and 9.8° , 19.5° , 40.8° are consistent with the amorphous nature of starch and rice flour, respectively. The amorphous character of starch and rice flour may result from the hydrogen bonds' ability to provide a free energy balance.

However, when starch and rice flour were combined with varying weight percentages of glycerol, liquid glucose, microcrystalline cellulose, and sodium carboxymethyl cellulose, a small phase change occurred and the amorphous nature of the edible paper decreased in comparison to the pure components (9.8 , 19.6 , and 40.7°). A small change in the 2θ value causes the edible paper's crystallinity to decline, resulting in the amorphous form. The increasing interaction of pure and dopants in edible paper is what causes the reduction in amorphous structure. The existence of hydrogen bonding may be the cause of this. The numbers in **Table 2** and the degree of crystallinity.

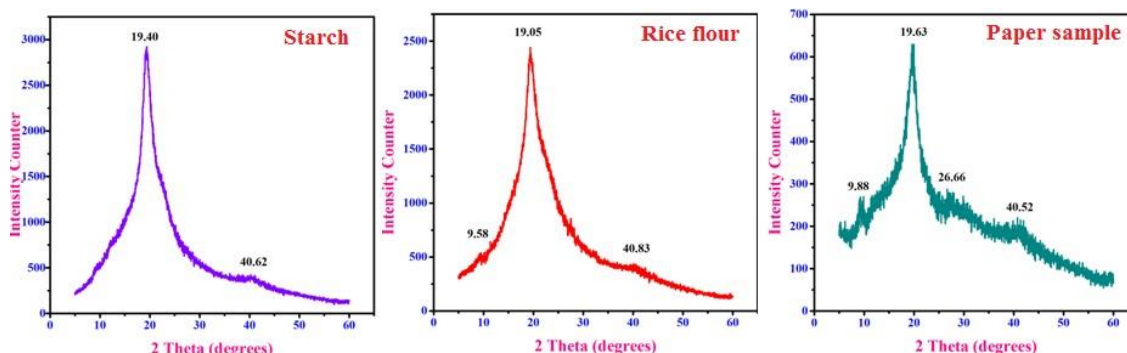


Fig.11 The X-ray Diffraction of Pure Starch, Rice Flour and Edible Paper.

SUMMARY AND CONCLUSION

Summary

Several experiments were conducted in order to choose the best composition for the development of edible paper. The edible paper-based substance was created using a mixture of potato, carrot, and radish starches, where the starch level is crucial to the material's strength. Due to the paper's lack of binding, the created paper was rejected.

The preliminary studies were carried out with the combination of starch (3%), CMC (0.5%), MC (0.5%), glycerol (5%) and LG (5%), the reason for not forming edible paper was due to the less concentration of starch.

Similar 5 combinations were taken with the starch concentration of 5% and 8% by keeping the other components constant it was found that 8% starch the strength of paper was found to be good with increase in the elasticity of the paper.

By keeping the remaining ingredients constant, 5 combinations were studied with starch concentrations of 5% and 8%. It was discovered that 8% starch increased the paper's flexibility while maintaining a good level of strength.

The study further proceeded with combination of starch, rice flour with CMC, MC, glycerol and LG and it was found that the formed paper was completely dried although it was having cracks on the surface. And crystallinity of the paper was found due to less plasticity of the rice flour. It was found that the effect of glycerol (20%) and xanthan gum (1%) on

physical and structural of the paper was found to be successful for preparation of the edible paper, the paper material formed was even in nature, and texture was good which binds the components to form the single edible paper material.

Final developed edible paper was having thickness of $138 \pm 2 \mu\text{m}$, it showed was found to be having more solubility. Hence, found that it was biodegradable in nature. The results of SEM revealed that edible paper is rough and slightly heterogeneous in nature. The X-ray diffraction study confirmed that edible paper is amorphous in nature.

FUTURE SCOPE

Development of paper with edible inks can be further explored by extracting the ink from the colored pigmented fruits and vegetables. Further studies on improved texture of the edible paper, water absorption ratio, anti-microbial and anti-oxidant property, tensile and tearing strength of the edible paper can be subjected for testing. Further studies on the development of the edible paper from waste vegetables could be carried out.

CONCLUSION

Rice flour is an economical alternative ingredient to prepare the paper instead of other commercial ingredients. Broken rice from industries, which generally discarded could be utilized for development of paper.

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