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Development and evaluation of biodegradable cutlery from sweet lime peel waste: A sustainable and efficient alternative to recycled plastics

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Abstract
With the increase environmental concerns of plastic material, the food industries are adopting the sustainable food holding and packaging solutions. Biodegradable cutleries have arisen as an environmentally friendly solution gaining significant attention due to their degradability, biocompatibility, and biological safety. This study aimed to develop biodegradable cutlery using sweet line peel powder and coconut coir. Steps include ingredient and process optimization followed by evaluation of water holding and absorption capacity, solubility, and biodegradability of developed cutlery. Results revealed that the developed cutlery had significantly increasing (p<0.05) water absorption rate with time, reaching 100% after 45 min. Furthermore, the water holding capacity also exhibited significant (p<0.05) increase with varying water temperature, ranging between 10±0.6 and 25±0.5 to min, within temperature span of 25-100 °C. The solubility was 31±1.23% after 30 min. of water immersion. Notably, biodegradability results demonstrated that the cutlery achieved complete biodegradation in 30 days. The versatility of the developed cutlery extends to serving both hot and cold liquid meals, showcasing its potential sustainable applications in the food industry.

Keywords: Waste valorisation, food waste, biodegradable cutlery, sustainability

Introduction

Cutlery is used for serving and consuming food products worldwide [1]. In ancient times, they were developed with natural components, including seashells, animal bones, or wood. At present, disposable plastic is majorly utilized for developing these products. These cutleries are regularly used in the homes, attributed to their flexibility, economic pricing, and light weight [2].

As per the literature, plastic accumulates in the surroundings and survives for several years and becomes hazardous to both humans and environment [3, 4]. This plastic fragments into microplastic through the effects of extramural forces, and goes into the sea, endangering the life of aquatic species [5]. Geyer *et al*. [6] state that if present manufacturing and waste management practices remained similar by 2050, almost 12,000 MT of plastic waste will be dumped in landfills or the environment. In addition, it might release carcinogenic and toxic substances in food products and affect human health [7].

Significant environmental and health concerns about plastic necessitate the need for potential alternatives [8]. Literature studies have demonstrated the development of biodegradable cutleries using food waste from the fruit, vegetable, and grain sectors [1, 5, 9]. These eco-friendly and organic cutleries aim to reduce plastic usage and protect the ecosystem by providing functional and sustainable alternatives. They offer a new solution to combat plastic pollution and align with the United Nations' Sustainable Development Goals. In addition to this, it will assist in agro-waste utilization, which is another environmental concern [1].

As per Yaradoddi *et al*. [5] the government and private sector are currently taking a significant interest in suitable alternatives to preserve the environment. The Defence Research and Development Organisation (DRDO) Lab has investigated the production of biodegradable cutlery which can be a potential option to replace plastic, as it is degradable, biocompatible, and biological safe in nature. The cutlery is suitable for serving hot and cold meals and can be composted in the natural environment. Countries worldwide have encouraged the use of biodegradable plastic bags through bans or incentives. The Ministry of Environment, Forests and Climate Change has launched the "India Plastic Challenge – Hackathon 2021" to encourage innovation and entrepreneurship in tackling plastic waste pollution.

Both sweet lime and coconut are harvested in abundance in India and have numerous uses in food products [10, 11, 12]. Sweet lime is mainly consumed fresh or in the form of juice. It is an ample source of essential components like vitamin C, pectin, flavonoids, and essential oils. These components impart antimicrobial and antioxidant characteristics [13]. While consuming lime, it is often removed, but it is also an abundant source of bioactive residues, including flavonoids, essential oil, vitamins A and C, and phenols [14, 15]. This lime peel can be used as a base material for cutlery, and its antioxidant and microbial properties can potentially resist microbial growth, hence its shelf life.

The industry dumps coconut shells and husks, which might cause environmental concerns. This warrants the usage of coconut waste [16]. As per the literature studies, the shell of the coconut is an abundant source of cellulose, demonstrating its use in nanofiber production [17]. Hence, its incorporation can provide strength to the cutlery.

Therefore, this study focused on developing biodegradable cutlery using lime peel. Guar gum and xanthan gum were used as binders, while glycerol was used as a plasticizer. The formulated dough was subjected to various processing techniques, including baking over a hot air oven and microwave. Coconut shreds were used for the enhancement in strength and water-holding capacity. The developed cutlery was then tested for water holding capacity, absorption rate, solubility, and soil burial examination, i.e., biodegradability test.

Materials and Methods

Raw material procurement

Sweet lime peel (as a fresh waste) and coconut coir were collected from a local market and juice shops in Agra. Guar Gum (Thermo Fisher Scientific India Pvt. Ltd.), Xanthan Gum (Nature), and Glycerol (Thermo Fisher Scientific India Pvt. Ltd.) were obtained from the local manufacturer.

Cutlery preparation

Lime peel powder and coconut shred preparation

Sweet lime peels were washed with water for the elimination of soil and dust particles. Washed peels were divided into pieces of 10 mm and dried at temperature and time combinations of 80 °C for 2.5 h, 80 °C for 5 h, 90 °C for 2.5 h and 90 °C for 5 h in a tray dryer. Dried sweet lime peel was grounded using a grinder followed by sieving using a stainless steel 100 mesh (1 mm pore size) to procure sweet lime peel powder. Powder with the desirable colour was chosen for further studies. Coconut coir drying was conducted at 80°C for 5-6 h, followed by grinding through grinder for particle size reduction. The powder was sieved with a stainless-steel sieve with a mesh size of 60 to separate larger particles or impurities from the desired coconut shreds.

Dough preparation

For dough preparation, dry ingredients (peel powder, guar gum, xanthan gum, and coconut shreds) were mixed thoroughly in a bowl, followed by the gradual addition of water while stirring until the mixture formed a dough. The formulated dough was kneaded on a clean and sanitized surface, followed by adding glycerol. Kneading was continued for 8-10 min until the dough became smooth. After kneading, the dough was portioned, rolled out, and shaped in cutlery.

Ingredient optimization

Different attempts were made to optimize the quantity of guar gum, xanthan gum, glycerol, and coconut shreds to obtain the desirable properties of the dough. Guar gum: xanthan gum ratio @1:0, 0:1 and 1:1 was used (total was @10% of peel powder weight). Glycerol was added @10%, 20%, 30%, 40% and 50% of the peel powder weight while coconut coir was added @5%.

Processing of cutlery

Cutleries from different dough formulations were processed using the baking oven and a combination of a baking oven and a hot air oven. In baking oven, different temperature and time combinations were applied, ranging from 100-120 °C for 35-70 min, respectively. While in combination method of baking oven processing was applied at combinations of 120-140 °C for 20-30 min followed by the hot air oven processing at 120 °C for 10-40 min.

Moisture content

The moisture content of the powders and the dough used was evaluated by taking 10 g of the sample in the petri dish and drying it in the hot air oven at 100 °C for 3 h. The dish was cooled in the desiccator and weighed. It was dried again at 100 °C for 1 h in a hot air oven and cooled in a desiccator and weighed. The process was repeated till three constant readings were obtained. The moisture content was evaluated using equation 1.

$$\text{Moisture (\%)} = \frac{M_1 - M_2}{M_1 - M_3} \times 100 \qquad \text{(Eq. 1)}$$

M_1= Material weight prior to drying (g).
M_2 = Material weight post drying (g).
M_3 = Empty dish weight (g).

Water absorption

Water absorption of the cutlery samples was evaluated as per the ASTM D570 method. Drying of the samples was conducted at 100 °C for 2.5 h. The dried samples were kept in a desiccator at room temperature. The initial dried weight of samples was noted. Samples were immersed in distilled water for certain time. The surplus water was drained, and all the samples were squeezed using scratch and residue-free wipes. The final sample weight was measured. The water absorption percentage was calculated using the equation 2.

$$\text{Water absorption (\%)} = \frac{\text{Initial weight} - \text{Final weight}}{\text{Final weight}} \times 100 \qquad \text{(Eq. 2)}$$

Water holding capacity

A water holding capacity test is conducted for the evaluation of time for which cutlery can hold water without affecting its texture. Water holding capacity was evaluated by filling cutlery samples with 20 mL of water at a temperature range of 25-100 °C, and time was noted till any damp/distortion appeared.

Water solubility

For the water solubility evaluation, each sample was immersed in distilled water and kept at room temperature (25 °C) for 5-30 min. The insoluble material was filtered through filter paper (Whatman® no. 1) and dried in a hot air oven at 105 °C for 2.5 h. Solubility was evaluated using equation 3 [18].

$$\text{Solubility} = \frac{\text{Initial dried weight} - \text{Final weight}}{\text{Initial dried weight}} \times 100 \qquad \text{(Eq. 3)}$$

Biodegradability

Biodegradability testing is designed to evaluate the ability of a material or product to undergo decomposition and break down into simpler substances by the action of microbes including bacteria, fungi, or other biological agents [19]. It assesses the extent and rate at which these microorganisms can consume and transform a material. This test was conducted using a soil burial method [20]. The developed cutlery was weighed and buried in the 1000 g of soil and weighed regularly at 5 days of interval. Percentage of weight lost after every 5 days was evaluated by using equation 4.

$$\text{Weight loss (\%)} = \frac{\text{Initial cutlery weight} - \text{Cutlery weight after 5 fivedays}}{\text{Initial cutlery weight}} \times 100 \text{ (Eq. 4)}$$

Statistical analysis

All the tests were conducted in replicates where each sample was assigned independently and randomly, and the mean values were reported. To evaluate the effect of different dependent variables (water temperature and time of water immersion) on the independent variables (water holding capacity, water absorption capacity and solubility), one-way ANOVA was carried out and the significant differences among the treatments were determined using Tukey HSD at 5% level of significance ($p<0.05$).

Results and Discussion

Peel powder preparation

The moisture content of the peel powder prepared at 80 °C for 2.5 h, 80 °C for 5 h, 90 °C for 2.5 h, and 90 °C for 5 h in a tray dryer was 12±1.0%, 11.7±0.73%, 11.3±0.96% and 11.0±0.77%, respectively. No significant difference ($p<0.05$) was observed between the moisture content of the powder dried at different temperatures and time combinations. The acceptable colour (visual appeal) was obtained only in case of the powder dried at 80 °C for 2.5 h; therefore, that powder was used in further experiments.

Formulation optimization and processing

The optimization of plasticizer (glycerol) and binders (guar gum and xanthan gum) was conducted during the dough formulation and cutlery processing steps. Dough with no glycerol resulted in cracks at the kneading stage, while dough with 10% and 20% glycerol content was less flexible in moulding than dough with 30% and 40%. According to Aslankoohi *et al.* [21] and Meerts *et al.* [22] a glycerol has a qualitatively similar effect to that of water and it also helps in increasing the extensibility of dough. There was no difference in the flexibility of the dough containing 30% and 40% glycerol. Therefore, a 30% glycerol concentration was chosen for the final dough used in further experiments. Binders' ratio of 1:0 and 0:1 of guar and xanthan gum resulted in cracks in the dough while moulding and 1:1 of both gums resulted in a smooth texture and convenient moulding. The baking and hot air oven dried all dough formulations at different temperatures and time combinations. The observations of the obtained cutlery are summarized in Table 1. Among all the dough and processing combinations, the dough containing a 1:1 combination of guar and xanthan was dried in a baking oven at 140 °C for 30 min and then dried in a hot air oven at 120 °C for 30 min. It had an acceptable texture and appearance. The developed cutlery has been depicted in figure 1.

Table 1: Visual observations of cutleries produced through numerous processing conditions

Processing conditions		Guar gum: Xanthan gum		
Baking oven	**Hot air oven**	**1:0**	**0:1**	**1:1**
110 °C/50 min	-	Contain moisture	Contain moisture	Contain moisture
110 °C/60min	-	Contain moisture	Contain moisture	Contain moisture
-	100 °C/60 min	Contain moisture	Contain moisture	Contain moisture
-	110 °C/70 min	Contain moisture	Contain moisture	Contain moisture
-	120 °C/35 min	Contain moisture	Contain moisture	Contain moisture
-	120 °C/40 min	Not fully dried but texture was appropriate	Moist at base	Moist at base
-	120 °C/50 min	Relatively less moisture but had burnt edges	Fully dried but slightly burnt	Fully dried but burnt from edges
120 °C/30 min	120 °C/10 min	Contain moisture	Contain moisture	Contain moisture
120 °C/30 min	120 °C/20 min	Contain moisture	Fully dried but distorted shape	Contain moisture
120 °C/30 min	120 °C/30 min	Contain moisture	Contain moisture	Relatively less moisture but not fully dried.
120 °C/30 min	120 °C/40 min	Fully dried but burnt	Fully dried but burnt	Fully dried but burnt
140 °C/20 min	120 °C/30 min	Not fully dried but had relatively low moisture	Desirable texture but not fully dried.	Not fully dried.
140 °C/30 min	120 °C/30 min	Not fully dried but had relatively low moisture	Moist at base.	Fully dried, desirable texture and appearance.

Fig 1: The picture of the developed cutlery used for testing

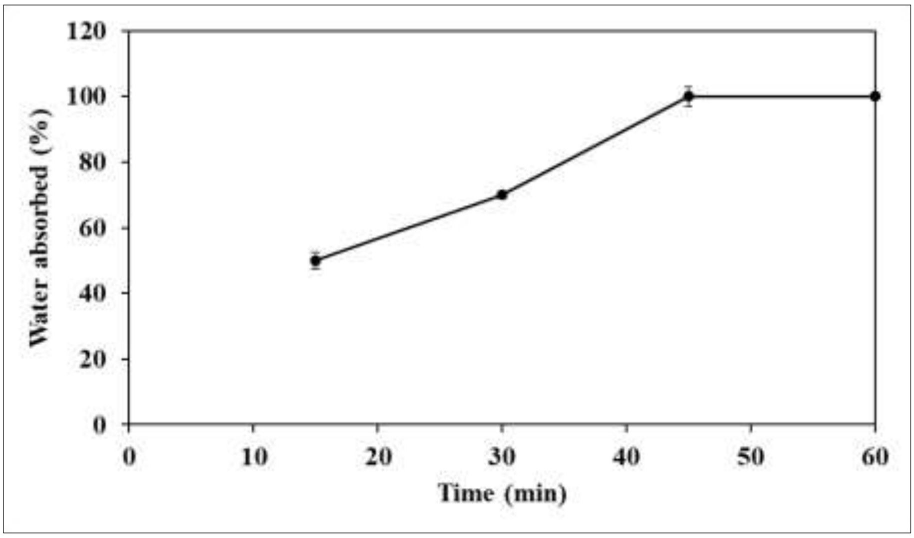

Fig 2: Water absorption (%) of the cutlery at different time intervals

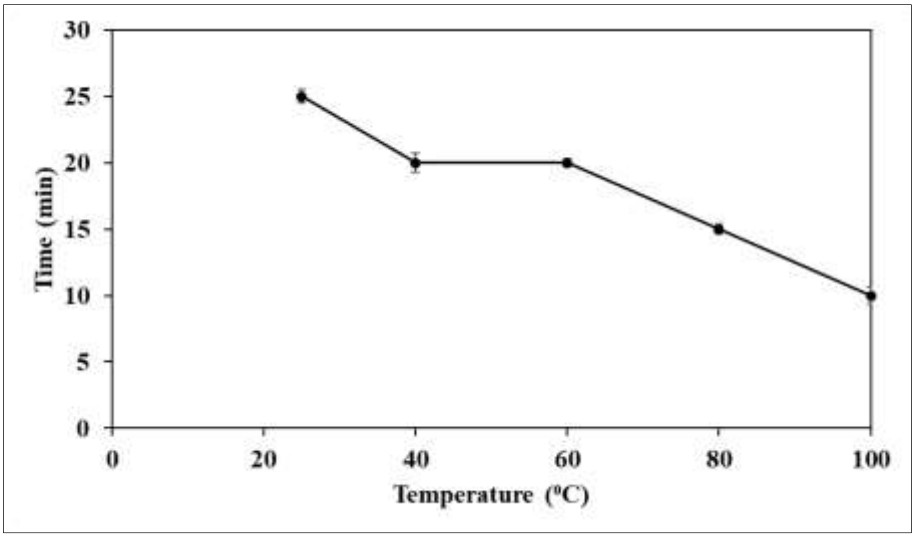

Fig 3: Water holding capacity (min) of the cutlery at different water temperatures

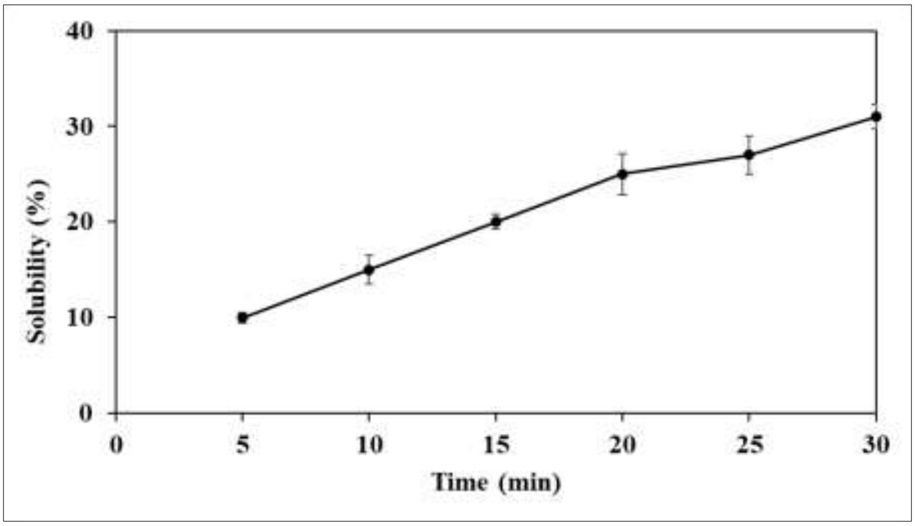

Fig 4: Solubility (%) of the cutlery material at different time intervals

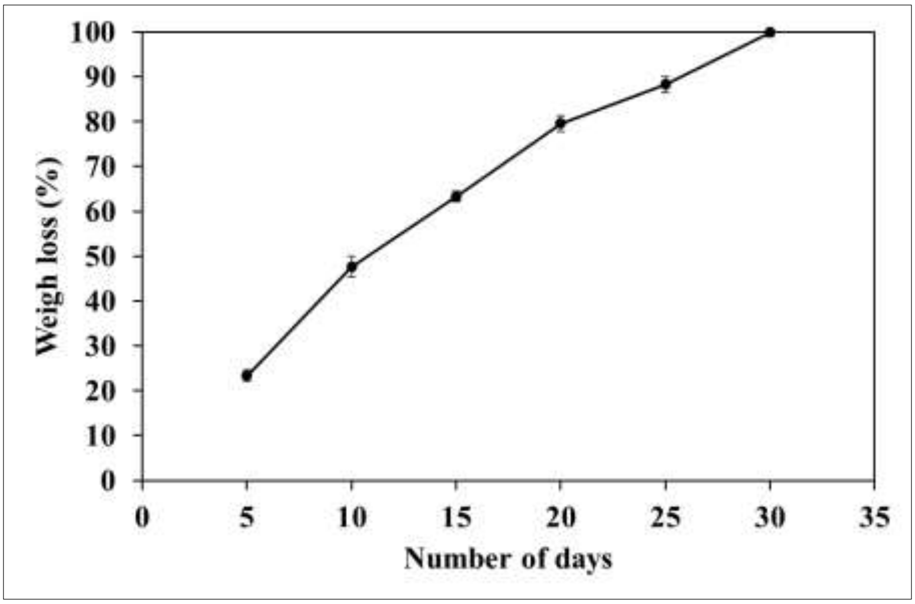

Fig 5: The weight loss (%) of the developed cutlery material (buried in soil) at different days interval

Water absorption and holding capacity.

The water absorption capacity of biodegradable cutlery is determined to evaluate its stability and durability [1]. The results indicate that as the time of immersion in water increased, the water absorption percentage of the cutlery also increased (figure 2). The water absorption of the developed cutlery was mainly due to the lemon peel (pectin), which is hydrophilic in nature [23]. The water absorption of the developed cutlery reached up to 100% after 45 min and stayed the same up to 60 min. Siddiqui *et al.* [1]. observed a >100% absorption rate of the developed cutlery developed with sago powder and *mosambi* peel, where a greater absorption rate was observed in cutleries that had a higher amount of peel (pectin) than powder. These findings emphasize the importance of considering the water absorption properties of the cutlery material and its intended use, as increased absorption can lead to a loss of structural integrity and functionality.

Water holding capacity refers to the water quantity a substance can handle for a specific time [24]. The developed cutlery had demonstrated a decreased water holding capacity with the elevation in the water temperature (figure 3). The values ranged between 25±0.5 to 10±0.6 min, where highest holding time was observed at 25 ℃ while the lowest was observed at 100 ℃. The results demonstrated that the developed cutlery can efficiently hold the liquid food products for around 25 min, served at room temperature. In contrast, the hot liquid meals (served between 60-80 ℃) can be served in the cutlery for 15-20 min [25]. Interestingly, the holding capacity significantly (p<0.05) declined between water temperatures of 20-40 ℃ and 60-100 ℃ while remained consistent for the 40-60 ℃ which might be attributed to the difference in the water penetration behaviour of pectin with variation in the temperature.

Solubility analysis

When developing biodegradable tableware, water solubility is a crucial parameter since it dictates whether the resulting bio-based tableware is appropriate for holding, serving, and packing liquid food products like milk and oil-based foods, among other things. Additionally, it also plays a significant role in its biodegradability. The solubility analysis in our study indicated that as the immersion time in distilled water increased, the solubility percentage of the cutlery significantly

(p<0.05) increased (figure 4). The maximum solubility was 31±1.23% after 30 min of immersion in water. A similar solubility range (2.07% to 23.88%) was reported in the study by Hanani *et al*. [26]. The low solubility could be attributed to the presence of hydrogen bonds between the ingredients and pectin molecular chains may have decreased the diffusion speed of water molecules into the developed cutlery [27]. These results emphasize the need for careful consideration of the solubility properties of the cutlery material to ensure its performance and suitability for specific applications.

Biodegradability test

Biodegradability testing evaluates the complicated biochemical process that takes place when microbes deplete a specific material. Despite of complexity, the test results evaluate relatively simple markers of the biodegradation process. According to Fritsche and Hofrichter [28], biodegradation occurs through two mechanisms. The first one is growth, in which comprehensive degradation of cutlery components in the soil takes place, transforming it into carbon and energy. The second one is co-metabolism, in which organic components are metabolized by substrates used as an energy and carbon source. The developed cutlery significantly lost weight on being buried into the soil and within 30 days it reached to complete biodegradation (figure 5). This demonstrated that it can be considered an environmentally friendly alternative to conventional plastic cutlery, which can persist in the environment for a much more extended period. Biodegradation of cutleries are dependent upon the material from which it is produced. Kumbhar and Masali [20] reported complete degradation of spoons made from moringa husk powder in 20 days while Hazra and Sontakke [29] reported that their developed cutlery (ragi-sorghum-wheat-based cutlery incorporated with ginseng root powder) took 4-5 days for complete degradation. Iqbal *et al*. [30] also reported the complete degradation of wheat, rice, and sorghum flour-based cutlery in 5 to 7 days. In the developed sweet lime cutlery biodegradation was a bit slower than the cutleries made by other material in different literature studies, which could be attributed to the lower water solubility. However, our developed cutlery, similar to others, also achieved 100% biodegradation, which signifies the environmentally friendly nature of our product.

Conclusion

The dough formulation comprising a 1:1 ratio of guar and xanthan gum, combined with 30% glycerol, and subjected to specific processing conditions (baking (in oven) at 140 °C for 30 min, followed by hot air drying at 120 °C for an additional 30 min), yielded cutlery with an appealing texture and appearance.

The developed cutlery exhibited notable characteristics, including a significant increase in water absorption rate (p<0.05), reaching 100% after 45 min. Solubility of the cutlery material was measured at 31±1.23% after 30 min. Water holding capacity fell within the range of 10±0.6 to 25±0.5 min, across a temperature span of 25-100 °C. This underscores its suitability for serving both cold and hot liquid meals, with the former having a recommended serving time of 25 min and the latter ranging between 15-20 min Furthermore, biodegradability tests indicated that the cutlery achieved complete degradation within a span of 30 days which demonstrates the environmentally friendly nature of the cutlery.

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Conflict of interest

There is no conflict of interest from any author.

Availability of data and material

All the available data has been included in this manuscript.

Authors Contribution

Divya Singh Chauhan: Conceptualization, Methodology, Investigation, Visualization, Writing-original draft, review, and editing.

Apoorva Behari Lal: Writing-original draft, review, and editing.

Dhruv Thakur: Conceptualization, Methodology, Validation, Resources, Supervision, Writing-Review & Editing.

Khushi Singh: Writing-original draft**.**

Pranav Vashisht: Conceptualization, Methodology, Validation, Resources, Supervision, Writing-original draft, review, and editing.

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