Latex-Based Paper Devices with Super Solvent Resistance for On-the-Spot Detection of Metanil Yellow in Food Samples

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Abstract
The following paper presents a construct for a paper-based device which utilizes latex as the hydrophobic material for the fabrication of its hydrophobic barrier, which was deposited onto the cellulose surface either by free-hand or stenciled drawing. This method demands the least amount of expertise and time from its use, enabling a simple and rapid fabrication experience. Several properties of the hydrophobic material were characterized, such as the hydro head and penetration rate, with the aim of assessing its robustness and stability. The presented hydrophobic barriers fabricated using this approach have a barrier width of 4 mm, a coating thickness of 208 µm, and a hydrophilic resolution of 446.5 µm. This fabrication modality boasts an excellent solvent resistance with regard to the hydrophobic barrier. These devices were employed for on-the-spot detection of Metanil Yellow, a banned food adulterant often used in curcumin and pigeon peas, within successful limits of detection (LOD) of 0.5% (w/w) and 0.25% (w/w), respectively. These results indicate the great potential this fabricated hydrophobic device has in numerous paper-based applications and other closely related domains, such as diagnostics and sensing, signalling its capacity to become commonplace in both industrial and domestic settings.

Keywords
Paper microfluidics · DIY fabrication · Frugal sensors · Adulterants · Point-of-detection

Introduction
Food is fundamental to the survival of an organism and the support of all essential physiological processes. However, this function is satisfied only if the food being consumed retains most of its original quality. Food production and supply-side problems of adulteration by malicious actors done in order to minimize the costs involved in the production, transport, and distribution of food often employ mixing in substitutes of lower quality to the original foodstuffs (Bansal et al. 2017). Such additions result in a reduction of the quality and nutritional value of the product, sometimes leading to the production of toxic substances that may be life-threatening to the consumer (Munikrishnan 2015). A variety of cases of food adulteration have been recorded and remain in practice, such as the addition of cheaper oils and fats to milk and milk products (Khan and Chittora, 2017), toxic ingredients like Sudan dyes or metal salts to spices and herbs (Osman et al. 2019), industrial sugars to honey (Naila et al. 2018), plant and animal proteins to minced beef and pork (Rady and Adedeji 2018), and unwanted preservatives to other foodstuffs (Yim et al. 2014). These practices serve to lower the cost of production, increase profit margins, and make up for the lack of sufficient manpower in the production process (Choudhary et al. 2020). Thus, there is an urgent need to detect these adulterants in a rapid and effective manner to reduce health complications and ensure better supply chain quality integrity.

Multiple methods have been conventionally applied for monitoring food quality. Some of the major techniques include high-performance liquid chromatography (HPLC) (Liu et al. 2018), liquid chromatography-mass spectrometry (LC–MS), gas chromatography-mass spectrometry (GC–MS) (Narang et al. 2017), enzyme-linked immunosorbent assay (ELISA), chemiluminescence (CL), and Fourier...
transform infrared spectroscopy (FTIR) (Muthukumar et al. 2018). These methods, despite being considered industry standards for food adulterant detection owing to their efficiency and accuracy, have certain limitations. These limitations range from them being time-consuming and laborious, resource, and equipment intensive and requiring highly skilled scientists and technicians to conduct the tests (Gao et al. 2018). Therefore, the development of simpler, low cost, and equally efficient methods of testing is urgent. To address the above issues, cellulose-based devices have garnered a significant amount of attention in the past decade (Prabhu et al. 2020b) (Prabhu et al. 2021) (Mani et al. 2020) (Hasandka et al. 2022) (Kelkar et al. 2022). Particularly, paper-based microfluidics (μPADs) is one of the areas that attracts increasing attention (Prabhu et al. 2020a; Ray et al. 2022). The use of paper has made it possible, to a considerable extent, to materialize the concept of the “lab-on-a-chip” (Govindarajalu et al. 2019), enabling experiments that previously required large-scale equipment and machinery to be performed on small pieces of paper when modified in an appropriate manner. This has become possible owing to its intrinsic wicking mechanism (Dossi et al. 2013), biodegradability, and modular nature (Li et al. 2020) (Younas et al. 2019) (Chen et al. 2019) (Li et al. 2015) (Zhang et al. 2014) (Tobjörk and Österbacka, 2011). It also holds the advantages of portability, disposability (Xie et al. 2019), and ease of integration with different detection methods (Kumar and Santhanam 2019), (Trofimchuk et al. 2020), (Katelakah et al. 2021), (Dias et al. 2018). Some of the practical applications involving the usage of paper-based approaches include pH analysis, urinalysis, and fabrication of test strips for less cumbersome experimentation (Jane Maxwell et al., 2013), (Cate et al. 2015).

Notably, the use of paper-based devices requires the creation of a hydrophobic barrier on the paper material to confine the sample/solvent. Although paper-based devices offer the above-mentioned advantages, the creation of these barriers is a difficult task, which requires expensive and complex chemicals and materials, as well as expertise in handling the equipment (Singhal et al. 2021). Most importantly, except for scholar glue, no adequate hydrophobic material has been identified to date that has the capability of confining a vast variety of solvents like strong acids and alka lics, organic solvents, and surfactants (Cardoso et al. 2017). Moreover, the existing methods of fabrication suffer from drawbacks, such as low throughput, mild to appreciable toxicity, spillage, and several involved steps, like high heating (Hasandka et al. 2021). Consequently, the search for a “universal and super solvent resistance barrier” continues, with the aim of employing it as an integral component of paper-based studies. To overcome this drawback, synthetic latex has been employed in our construct toward the development of hydrophobic barriers on the paper surface by simple free-hand deposition (Fig. 1). This frugal material was utilized first to test its capacity to confine a variety of solvents, after which we replicated a rapid and sensitive colorimetric detection for Metanil Yellow (MY) or (Acid Yellow 36), an illegally used food dye.

### Materials and Methods

#### Barrier Fabrication

Whatman® filter paper (grade 1) of 180-μm thickness and 11-μm pore size was purchased from Whatman®, India. Berger Advanced Latex Plus was procured from a local hardware store. Firstly, a Pro Circle stencil consisting of circles of radii 5 mm and 9 mm was used to draw a circular pattern of two concentric circles on Whatman® filter paper (grade 1) strips to demarcate the regions for applying the fabrication. Later 60 μL latex was deposited onto the ring-shaped region between the two drawn concentric circles to form a closed, ring-shaped, round barrier to confine the fluids. The coating was allowed to penetrate the paper material and dried for 2 h, followed by flipping the paper and repeating the same patterning process on the other side, to obtain a double-sided coating (Supplementary Fig. 1). Finally, one side of the fabricated latex barrier-based paper device was taped using a 3 M adhesive tape.

#### Solvent Compatibility

Hydrochloric acid (HCl) and sulfuric acid (H2SO4) were obtained from Loba Chemie, India, and Suvidhinath Laboratories, India, respectively. Tween 20 and Triton-X100 were purchased from Calbiochem. Sodium dodecyl sulfate (SDS), dimethyl sulfoxide (DMSO), dimethyl formamide (DMF), and acetonitrile were procured from Merck, India. Acetone, isopropyl alcohol (IPA), and cetyltrimethylammonium bromide (CTAB) were purchased from SRL, India. Ethanol was obtained from Hayman, India. The prepared circular devices were tested for solvent compatibility against 12 solvents. Eight devices were used for each solvent, wherein 30 μL of the solvent was pipetted onto the fabricated circular device and observed for a maximum of 30 min to check for any leakage of the solvent across the barrier.

#### Coating Thickness

The thickness of the uncoated/plain paper and the double-side coated paper was measured using a Mitutoyo Digital Micrometer (293–831), with three readings taken for each. In each reading, the value obtained for the uncoated paper was subtracted from that of the double-side coated paper,
and the average of the three values was taken to determine the coating thickness.

**Hydrophilic Channel Resolution**

Two parallel lines of latex coating, each of 2-mm width, were deposited on the filter paper to form straight channels, considering three different channel widths of 2, 3, and 4 mm. The fabricated straight channels were then observed under a bright field light microscope at fourfold magnification after the addition of water within the channel. Images of each device were captured using Magnúis MLX light microscope, and the lowest width of the channels obtained (channel resolution) was measured using ImageJ software version 1.51 (NIH).

**Penetration Rate**

A straight channel (length of 5 cm, barrier width of 4 mm, and channel width of 4 mm) with a circular portion at one end was patterned on the filter paper using the same procedure as the standard device fabrication. A total of 60 μL of water was added to the circular zone, and the distance traveled by the water in the straight channel was measured at a fixed time interval of 10 s. The same procedure was followed for an uncoated device with the same dimensions of the channel. Three trials of the penetration rate measurement were conducted for the uncoated and coated paper device. Average values of the penetration rate were plotted and compared with the ideal Washburn equation plot.

**Hydro Head Measurement**

A square area of 5-mm diameter of completely uncoated paper, one side coated latex-based paper, and two sides coated latex-based paper were taped onto the lower end of a vertical open-end glass tube, with a ruler set along the tube on a piece of cardboard, aligning the zero mark of the ruler with the end of the tube. Water was pipetted in from the top end of the tube, and the height of water required for penetration through the uncoated paper and coated latex hydrophobic barrier was observed. Three trials were conducted for the measurement, and the hydro head and penetration pressure for the uncoated and coated papers were determined from the average values obtained (Supplementary Fig. 2 and Supplementary Table 1).
**Tensile Strength Measurement**

The experiment for measuring tensile testing was performed on the uncoated and latex-coated paper using a Universal Testing Machine (UTM Instron 3366). The ends of 10 by 1-cm dimensions of uncoated paper and two sides of latex-coated paper were placed inside the grip mechanism of the machine, and the tensile strength of the specimen was tested by applying the load on both ends.

**Detection and Quantification of MY**

Metanil Yellow (MY) or Acid Yellow 36 (Sodium 3-[(4-aminophenyl) diazinyl] benzene sulfonate) was purchased from Loba Chemie, India. For the preparation of the standard curve, 0.001, 0.005, 0.01, 0.03, 0.05, 0.25, 0.5, 1.25, and 2.5 mg/mL of MY were prepared from a stock solution of 5 mg/mL MY. Three circular devices (1-cm diameter) were used for each concentration, wherein 2 µL of concentrated (35%) HCl indicator was drop-casted into the device and dried for 10 min. Subsequently, 18 µL of the samples containing different concentrations of MY was added. Images of the color change were captured for each device after 30 s and analyzed by measuring the RGB intensity using ImageJ software. The results were used to plot a standard graph of intensity versus concentration of MY in mg/mL.

Two food items, *Curcumin longa* (turmeric powder or curcumin) and *Cajanus cajan* (pigeon pea or Arhar Dal), were procured from a local grocery store. Samples of curcumin and pigeon pea were spiked with 0.002, 0.01, 0.06, 0.5, 1, and 2.5% (w/w) concentrations of MY in a total sample weight of 500 mg. The equivalent weight of food samples was taken as the respective control. All samples were mixed with water and vortexed to yield a volume of 10 mL. Three circular devices were utilized for each concentration. A total of 2 µL of concentrated (35%) HCl indicator was added to the device and dried for 10 min. A volume of 18 µL of the spiked samples was added to the devices, and images of the color change were captured after 30 s, which were then analyzed for RGB intensity using ImageJ software.

**Results and Discussions**

**Fabrication of Latex-Based Paper Devices**

The first step in converting a plain paper into a paper-based device was achieved by a free-hand deposition or coating with the commonly available latex on the front and rear side of the paper (Fig. 2a). Then, the microscopic resolution of latex-based paper devices was found, which depicts the hydrophilic channel width between the two hydrophobic barriers. Using image analysis, we determined the resolution of the barrier as $1.34 \pm 0.1622$ mm (Fig. 2b). The average coating thickness of this hydrophobic barrier was measured using a digital micrometer and was noted to be $208.667 \pm 119.3$ µm. This low barrier thickness shows that these devices are significantly less bulky and can be leveraged for fabricating cellulose-based wearable sensors, leading to low discomfort when being worn by the end-user.

**Characterization of Latex-Based Paper Devices**

Another test that is gaining an increasing importance is the measurement of the hydro head value of hydrophobic barriers and their comparison with those of plain paper (Fig. 3). The hydro head is roughly understood as the pressure, in the

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**Fig. 2** (a) Circular paper-based device fabricated using latex (scale bar 1 cm). (b) Microscopic image of two parallel hydrophobic barriers and hydrophilic channel (scale bar $1.34 \pm 0.1622$ mm)
form of the height of the water, that the barrier can withstand, before allowing the water to leak through it. Hence, it is a demonstration of its hydrophobic properties. From these results, it was evident that the double-sided coating (123.1096 N/m² or 12.667 ± 1.1547 mm) has a higher hydro head than the single-sided coating (74.945 N/m² or 7.667 ± 0.5773 mm), which in turn has a higher hydro head than the uncoated paper (68.3942 N/m² or 7 ± 1 mm). This agrees with the fact that the hydrophobicity of the barrier allows it to withstand a significantly higher pressure than the uncoated devices and thereby supports the robustness of the employed barrier.

Measurements of penetration rate were performed in the latex-based paper device and plain paper (Fig. 3). Upon analysis, a well-defined reduction in the values of distance traveled with respect to time was observed in the devices coated with the hydrophobic barrier compared to those of the uncoated paper. The readings of the uncoated paper were in close accordance with the predicted values of the Washburn equation, while those of the coated devices yielded lower values. This phenomenon can be attributed to the hydrophobic barriers exerting surface tension in the direction opposite to that of the flowing solvent (here, water) due to its hydrophobic repulsions, and hence, a reduction in its distance traveled and the penetration rate is observed.

The tensile strength is a significant parameter for assessing the robustness of a hydrophobic barrier, whose results can be used for other studies as a reference. In this study, a Universal Testing Machine was used for tensile strength testing and the resulting graphs of the tensile strength (MPa) versus tensile strain (%) were plotted for both uncoated devices and the devices coated with the latex-based barrier. The coated devices showed a significantly higher tensile strain or displacement (6.31%) at the maximum force applied, as compared to the uncoated paper (2.04%), which is in agreement with the fact that the elastic nature of latex allows the device to withstand a larger amount of displacement at a higher maximum force (18.7 N), compared to the uncoated paper (10.07 MPa), thereby aiding us to conclude that the proposed latex-coated paper-based device has a higher tensile strength than ordinary paper (Fig. 4a and b).

**Solvent Resistance of Latex-Based Paper Devices**

To utilize the latex-based device as frugal sensors or point-of-care detection tool, it is of paramount importance to determine their resistance to a wide-range of solvents (polar, non-polar, acids, alkalis, and surfactants). Eight paper devices were used for each solvent. Interestingly, none of the latex-based barriers was disrupted, i.e., no leakage was observed for any of the solvents even 30 min after the addition of the solvents. This study confirms that the latex-based hydrophobic barrier was indeed capable of confining a variety of solvents, including water, surfactants, acids, and alkalis up to certain levels of concentrations, displaying high resistance to all types (Fig. 5). Consequently, the barrier can be potentially applied to various studies involving these solvents, which was previously not possible using other do-it-yourself (DIY)-based and high-throughput fabrication methods. A comparison of different hydrophobic barriers and their solvent resistance is provided in Supplementary Table 2.
Colorimetric Detection of MY Adulterant

To demonstrate their practical significance, the developed latex-based paper devices were utilized for the detection of Metanil Yellow (Acid Yellow 36) which is a water-soluble food dye that has been banned worldwide due to its increasing usage in the adulteration of food products like curcumin (turmeric powder), pulses, and numerous other items. Owing to its rich yellow color, low cost, and easy availability, Metanil yellow is often added to food-stuffs to make the adulterated food items more attractive. Despite being banned, its use in a variety of food products
continues to date (Khan et al. 2020) and contributes to the occurrence of cancer, paralysis, and other diseases (Ashok et al. 2015). The reason behind this potential carcinogenicity is attributed to diphenylamine, one of its metabolites, which is a suspected mutagen and carcinogen (Kourani et al. 2020). Hence, a cheap and rapid detection method that can be applied even by the consumers on their own must be achieved. To this end, paper-based devices have been fabricated to demonstrate the rapid and sensitive colorimetric detection of Metanil Yellow using concentrated hydrochloric acid.

MY is an azo dye, which is prepared by the coupling of metanilic acid and diphenylamine. It is an electrochemically active dye and can thus also be detected using electrochemical methods (Shereema et al. 2018). The dye has the characteristics of forming a dark pink/magenta color when exposed to a highly acidic environment (Kourani et al. 2020), and this particular reaction was utilized for its detection in this experiment. A common DIY technique recommended for detecting MY is the use of concentrated HCl as an indicator (Safety and Road 2012), which has been applied to numerous foods. The dye was observed to be sensitive toward 35% concentrated HCl, such that it displayed an instantaneous color change upon contacting the fumes of the indicator. In a previous study, acetic acid had been used for the detection of MY using the same principle (Kourani et al. 2020). Paper devices previously fabricated using wax printing (Chen et al. 2013), contact printing (Dornelas et al. 2015), or a correction pen (Mani et al. 2019)(El-Shaheny et al. 2021) have not reported well-defined resistances toward strong acids. Our solvent resistance studies demonstrate that latex-based paper devices can be leveraged for detecting MY using HCl.

First, different concentrations ranging from 0.001 to 5 mg/mL of MY in water were prepared. These concentrations are commonly used in food samples and were selected based on the existing literature (Dhakal et al. 2016). Prior to the addition of the indicator, the solutions displayed a yellowish tinge that became increasingly darker with the rise in the concentration of MY. The solution containing the lowest amount of MY (0.001 mg/mL) showed a very faint yellowish shade, almost unnoticeable to the naked eye. Upon the addition of 35% concentrated HCl (as an indicator), an instant change from colorless to pink was observed (Fig. 6a). These results indicate that the conventional test for MY showed an increase in the color intensity for increasing concentrations of MY. The same colorimetric assay was performed on latex-based paper devices, where different concentrations of MY were drop-cast on the devices followed by the addition of concentrated HCl. Strikingly, a distinct and visual color change was observed (similar to bulk experiments). Figure 6b depicts the differences in the visual signal of MY. To confirm these qualitative results, an intensity analysis of the images captured was performed by us and a clear rise in the RGB intensity values from concentrations of MY ranging from 0.001 to 5 mg/mL was observed, with lowest intensity corresponding to the control (Supplementary Fig. 3).

**Latex-Based Paper Devices for Colorimetric Detection of MY in Food Samples**

Curcumin samples were spiked with MY at concentrations of 0.002, 0.01, 0.06, 0.5, 1, and 2.5% (w/w), and the images were collected for the same. Visually, a clear rise in the intensity of the magenta color was observed for 0.5% (w/w) and above, indicating positive results for the presence of MY. However, for samples with concentrations from 0.002 to 0.06% (w/w), no well-defined change in color was observed upon addition of the indicator, and only a spot of the undissolved curcumin was observed on the device. The analysis of RGB intensities of all devices using ImageJ software (Fig. 7a) showed that the lowest concentration of MY

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Fig. 6 (a) Colorimetric assay for detecting Metanil Yellow of different concentrations using concentrated hydrochloric acid. (b) Paper-based colorimetric assay for detecting Metanil Yellow of different concentrations (scale bar 1 cm). Average value and ± standard deviation were measured.
that exhibited an intensity greater than the control was 0.5% (w/w). We conclude that the Limit of Detection (LOD) of the paper-based device for the detection of MY in curcumin is 0.5% (w/w). For lower concentrations, the color of curcumin was dominating enough to conceal any possible color change as a result of the reaction between MY and the indicator, such that the intensity analysis had a high probability of yielding false-positive results. Therefore, the intensity analysis for concentrations below 0.5% (w/w) was excluded.

Pigeon pea samples were spiked with MY at concentrations of 0.002, 0.01, 0.06, 0.25, 0.5, 1, and 2.5% (w/w), and the images were collected for the same. A well-defined rise in the magenta color was observed in the concentration range of 0.06 to 2.5% (w/w), indicating that the paper-based device was capable of the desired performance in this range. Furthermore, the limitation of obtaining false positive values, unlike for curcumin, was not observed for pigeon pea, as the latter completely settled at the bottom of the tube and left the supernatant containing the dissolved MY for the actual analysis. Moreover, upon intensity analysis using ImageJ software, the lowest concentration exhibiting a value higher than that of the control was (0.25 (w/w%), indicating the LOD of the paper-based device for the pigeon pea samples (Fig. 7b).

Synthetic latex functions as an elastomer and is produced by the polymerization of a variety of monomers. It has several features that make it a suitable candidate for this study compared to natural latex, such as lower hardness, higher elasticity, and higher abrasion resistance (Vijetha et al. 2014). These make it potentially applicable to future experimentation and usage as wearable sensors, which are light-weight and less irritating to the users. Furthermore, natural latex contains phospholipids and proteins (Sansatsadeekul et al. 2011), which may make it unsuitable to perform solvent resistance tests and related studies due to
unknown interactions between the components of the solvent and those of the latex material. Previous research has also utilized sophisticated methods like solid-phase extraction and ultra-performance liquid chromatography electrospray ionization-tandem mass spectrometry (SPE-UPLC-MS/MS) for the simultaneous detection of Metanil Yellow along with three other water-soluble pigments in food products and have achieved high sensitivity in the form of 0.05 mg/L as the LOD (Ntraloulou et al. 2020). Studies have also utilized paper-based devices for the detection of Metanil Yellow such as surface-enhanced Raman spectroscopy (SERS) developed by the use of inkjet-printed thin films comprising of robust nanostructured silver to act as SERS swabs. The study attained a LOD of 1 μM for detecting the dye in organic dals samples using spectral analysis (Kumar and Santhanam 2019). Another study included the use of cyclic voltammetry and differential pulse voltammetry for the detection of this dye in curcumin, where the voltametric behavior of MY and curcumin was studied on bare glassy carbon electrode and carbon quantum dot-modified glassy carbon electrode. MY was detected with high specificity even in the presence of interfering entities at a LOD of 0.03 μM (Shereema et al. 2018). Considering the sensitivity and efficiency of these methods, the technique used by us may not be as sensitive as the ones mentioned. However, for the purpose of obtaining on-the-spot results indicating the presence of MY, this approach offers both simplicity and cost-effectiveness.

**Conclusion and Future Perspectives**

In this work, we focused on a do-it-yourself fabrication of paper-based devices using latex as a hydrophobic barrier, with the primary intention of validating its functionality as a universal barrier with resistance toward a variety of solvents. The solvent confining capability of the proposed hydrophobic barrier was demonstrated through the detection of MY, a commonly used food adulterant in curcumin and pigeon pea. The analysis yielded positive outcomes with significant LODs of 0.5 and 0.25 (w/w%), respectively for Metanil Yellow spiked in curcumin and pigeon pea. Hence, for the first time, latex was successfully proposed for frugal detection systems in the future. Its excellent solvent resistance makes its usage possible in studies involving strong acids and alkalis, thereby eliminating the search for other high-quality materials. Other characteristic studies such as hydro head, penetration rate, microscopic resolution, and tensile strength tests indicated that latex exhibits robustness, desirable hydrophobic properties, excellent elasticity, and tensile strength, all of which make it eligible for future applications such as wearable and textile-based sensors for monitoring important biomarkers like pH, glucose, and sweat quantity.

An important observation made in this study was that this method, used for the construction of the latex-based paper microfluidic device, is a low throughput method. To enable high throughput, sophisticated instruments like pen-plotters, stamps, and inkjet printing with increased printing capacity (Amin et al. 2017) can be used for future applications. The devices can also be potentially applied in the detection of other food adulterants in packaged food products, wherein modified versions of these latex-based devices can be built into the package itself for on-the-spot detection, thereby winning customer confidence regarding the food product. In a broader perspective, such frugal fabrication methods and simple colorimetric assays will boost the spreading of awareness regarding food adulteration among the general population and therefore, refine the supply chain management system as well.

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**Author Contribution** Rohitraj Ray—validation, conceptualization, data curation, writing. Calvin Noronha—validation, writing, resources, software. Anusha Prabhu—supervision, methodology, visualization. Naresh Kumar Mani—conceptualization, project administration, funding acquisition, supervision, visualization.

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**Data Availability** All data generated or analyzed during this study are included in this published article.

**Declarations**

**Ethics Approval** This article does not contain any studies with human participants or animals performed by any of the authors.

**Informed Consent** Informed consent is not applicable.

**Conflict of Interest** Rohitraj Ray declares no competing interests. Calvin Noronha declares no competing interests. Anusha Prabhu declares no competing interests. Naresh Kumar Mani declares no competing interests.
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