

## Morphological and Thermal Analysis of Poly(L-lactic acid)/ Carboxymethyl Cellulose Bio-composites

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### Abstract

Poly (L-lactic acid) (PLLA) and carboxymethyl cellulose (CMC) are derived from renewable resources and both have excellent properties such as biocompatibility and biodegradability. Applications of PLLA are restricted for some of its inadequate physical properties such as low glass transition temperature, relatively low melting point, high crystallization temperature, slow crystallization rate, and poor heat stability. This study aims to prepare and characterize a renewable bio-composite with more adequate properties for applications by solution casting of different PLLA, and CMC ratios. CMC was prepared from microcrystalline cellulose (MCC) of mustard stalks (agricultural wastes) and the yield was about 78%. The samples were analyzed via Fourier transform infrared (FTIR) spectroscopy, differential scanning calorimetric (DSC), wide-angle X-ray diffraction (WAXRD), and scanning electron microscopy (SEM) for spectroscopic and thermal properties characterization. The interaction between carboxylate groups of CMC surfaces and the terminal hydroxyl, terminal carboxyl, and carbonyl groups of PLLA through hydrogen bonds caused the shift and broadening of the band of FTIR spectra. The decomposition temperature of CMC was increased and formed excellent bio composites with PLLA. Composite crystallinity varied with the percentages of the CMC. The fibrous form of CMCs is present in the SEM micrograph indicating cellulose's fibrous structure was not affected by carboxymethylation. In composites, the surfaces of CMC fibers are layered and different from the reported results of SEM micrographs of PLLA/MCC composites. PLLA/CMC performs better in thermal properties but its mechanical characteristics cannot be determined owing to a limitation where the composite was too brittle and not able to undergo tensile testing.

**Keywords:** Poly(L-lactic acid), Carboxymethyl cellulose, Cellulose, Biocomposite, Mustard stalks, Agricultural waste.

### Introduction

Biocomposite materials have attracted significant attention in different fields due to their renewable and sustainable resources and minimal environmental effects. Environmental concerns have led to increasing research to synthesize useful industrial materials from plant biomass. Research extensively focused on developing such composites to apply in ecological and biomedicine fields (Chen et al., 2015; Rahaman et al., 2020). CMC is modified cellulose widely used in different industries, including food, detergents, personal care, pharmaceutical, paints, and textiles (Heinze & Pfeiffers, 1999; Stigsson et al., 2005; Methacanon et al., 2003). It is also used as a preservative for coating fresh fruit and thickener for pharmaceutical products (Olaru et al., 1998; Toğrul & Arslans, 2003; Xue & Ngadis, 2009). It is a high molecular weight, water-soluble, and anionic linear polysaccharide, and can be synthesized by monochloroacetic acid (MCA) reaction with alkali cellulose (Jia et al., 2016). Different varieties of cellulosic materials such as raw cellulose (Mondal et al., 201), wood (Salama et

al., 2018), cotton linters (Jahan et al., 2007), paper sludge (He et al., 2009), fibers (Gu et al., 2012) can be used for CMC preparation and, therefore, considered as renewable, non-toxic, and environmentally friendly material. In addition, the physicochemical properties of CMC can be improved in combination with appropriate fillers.

PLLA has been widely used with polymers such as starch, chitosan, collagen, polysaccharides, and modified cellulose to improve mechanical, thermal properties, crystallinity, degradation rate, and permeability gases or vapors (Oksman et al., 2003; Rahaman et al., 2020; Rahaman et al., 2019; Hosen et al., 2017; Hosen et al., 2017; Arjmandi et al., 2015). PLLA is a semi-biosynthetic biodegradable thermoplastic polymer with excellent mechanical properties, produced by the fermentation of corn starch to lactic acid and subsequent polymerization (Rahaman & Tsuji, 2013). PLLA coupled with other materials

can be tuned for different fields as required, ranging from construction materials to biomedical applications. The Food and Drug Administration (FDA) has approved devices utilizing PLLA for direct contact with biofluids as it is non-toxic and biocompatible (Xiao et al., 2012). It would be possible to develop a biocomposite with better properties from PLLA and CMC derived from muststalkk's cellulose.

To the best of our knowledge, no report of CMC prepared from mustard stalks (agricultural wastes) was found. In this study, we prepared biocomposites of PLLA and CMC with different ratios. Compositional effects on the structural, morphological, thermal, and crystalline properties have been systematically studied by FTIR spectroscopy, Wide-angle X-ray Diffraction (WAXD), Differential Scanning Calorimetry (DSC), and Scanning Electron Microscopy (SEM) techniques.

## Materials and Methods

### Materials

Mustard stalks were collected from the Kushtia district, Bangladesh. PLLA was collected from Mitsubishi Chemical Corporation, UNITIKA Plastics Division, and Japan. Other chemicals used in the experiments were NaOH, ethanol from Merck, Germany and NaClO<sub>2</sub>, H<sub>2</sub>SO<sub>4</sub>, monochloroacetic acid, acetic acid from BDH, England. All chemicals were laboratory-grade chemicals.

### Methods

#### Preparation of Microcrystalline Cellulose

Mustard stalks were dried in sunlight and then cut into small pieces, approximately 1-1.5 inches scoured with 6.5 g of soap flake and 3.5 g of detergent per liter at 70–75 °C for 30 min (Mondal & Haques, 2007). Cleaned and dried stalks were treated with 17.5% NaOH in a liquor ratio of 1:25 for 4 hours at 45 °C to reduce β- and γ-cellulose (Rahaman et al., 2022). The residue was separated, washed thoroughly with water to neutral pH, and then dried at room temperature. The residue was bleached with 1% sodium chlorite (NaClO<sub>2</sub>) for the removal of lignin. The α-cellulose was washed with water and dried at room temperature. Finally, α-cellulose was treated with H<sub>2</sub>SO<sub>4</sub> solution (60 wt%) to get MCC and dispersed in absolute ethanol for the preparation of CMC (Rahaman et al., 2022; Morán et al., 2008). Figure 1 shows the photograph of α-cellulose, MCC and CMC.



α-cellulose



MCC



CMC

**Figure 1:** Photograph of α-cellulose, MCC and CMC.

#### Synthesis of Carboxymethyl Cellulose

MCC was taken in absolute ethanol with the ratio of solid to liquid was 1:30. Later, 30% NaOH was added with the 2.7 ml NaOH solution ratio for 1 g MCC and conducted alkalization for 1 hour at 30 °C temperature. 120% (w/v) monochloroacetic acid (MCA) was added dropwise under constant stirring to create a cellulose–liquor ratio of 1:1.2 and conducted reaction at 50 °C for 3 hours (Mondal et al., 2015). The residue was separated by centrifuge and neutralized with 90% acetic acid (v/v) (Rahaman et al., 2019; Pushpamalar et al., 2006). The residue was washed with ethanol and dried for constant weight. The yield calculation of CMC is below:

$$\text{Yield (wt\%)} = 100 \times \frac{\text{Weight of dried CMC}}{\text{Weight of dried MCC}}$$

#### Preparation of PLLA/CMC composites by solution casting method

PLLA/CMC biocomposites were prepared by the solution casting method (Hosen et al., 2017; Hosen et al., 2017). Different weights of PLLA dissolved in tetrahydrofuran (THF) (Sato et al., 2013) and different weights of carboxymethyl cellulose (CMC) dispersed in absolute ethanol were mixed and stirred until the formation of a homogeneous mixture. The mixtures were poured into Petri dishes to form liquid composites and dried at room temperature. Finally, the composites were dried

at 60 °C for 1 hour in the oven to get moisture-free composites and preserved for characterization. The biocomposites were designated as PLLA/CMC-X, where X is the percentage of CMC in PLLA/CMC composites.

### Characterization

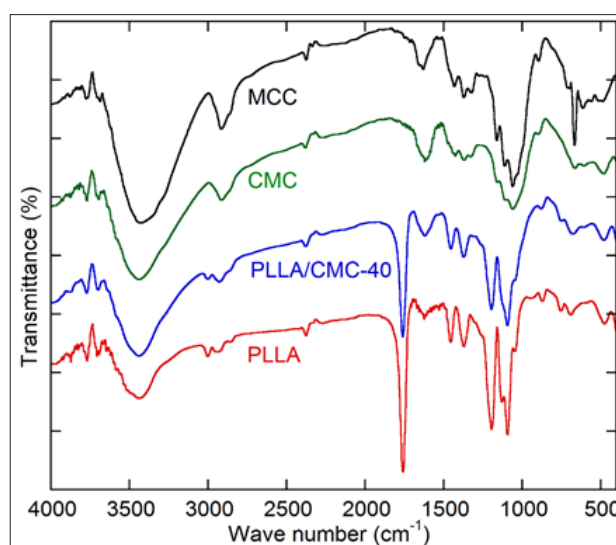
The FTIR (Shimadzu IR Prestige-21, Japan) was used to determine the surface functional groups of MCC, CMC, composite (PLLA/CMC-40), and PLLA. The samples were compressed with dry potassium bromide (KBr) powder for making disks. The spectrum for each sample was recorded in the region of 400–4000  $\text{cm}^{-1}$  at a resolution of 4  $\text{cm}^{-1}$ . The resulting FTIR spectra were studied to confirm CMC formation and composite compatibility with the intensity and shift of vibrational bands. WAXD patterns of composites were measured with a BRUKER D8 Advance wide-angle X-ray diffractometer using Cu K $\alpha$  radiation (0.154 nm) voltage of 50 kV and current of 40 mA with  $2\theta$  ranges from 5 to 50. DSC thermograms were performed with a Shimadzu instrument (DSC-60, Japan). The scanning temperature was from room temperature to 400 °C using a heating rate of 10 °C  $\text{min}^{-1}$  under a nitrogen atmosphere with a gas flow rate of 20  $\text{cm}^3 \text{min}^{-1}$ . The surface morphologies of MCC, CMC, and composites were observed by SEM (SEM Leica AS- 360FE), at various magnification ranges, which was containing Tungsten (W) filament with an accelerating voltage of 20 kV.

## Results and Discussion

### Fourier Transform Infrared Spectroscopy

Figure 2 shows the FTIR spectra of MCC, CMC, composite (PLLA/CMC-40), and PLLA. The characteristic peaks of MCC, CMC, PLLA, and PLLA/CMC in FTIR spectra are given in Table 1. MCC shows characteristics peaks at 3435.22  $\text{cm}^{-1}$  for the stretching vibration of the –OH group and absorption peak at 2916.37  $\text{cm}^{-1}$  corresponds to the –CH stretching vibration

group (Khalil et al., 2001). The vibration peaks at 1371.39  $\text{cm}^{-1}$  and 1433.11  $\text{cm}^{-1}$  are assigned to the bending vibration of the –CH and –C=O bonds in polysaccharide aromatic rings (Nacos et al., 2006). The peaks at 1323.17  $\text{cm}^{-1}$  and 1111.00  $\text{cm}^{-1}$  are due to –OH bending vibration and –C–O–C– stretching, respectively (Adinugraha et al., 2005). A band around 1060.85  $\text{cm}^{-1}$  represents the –C–O and C–H stretching vibration, at wavenumber 896.90  $\text{cm}^{-1}$  is detected for 1, 4- $\beta$  glycoside of cellulose (Alemdar & Sain, 2008; Elanthikkal et al., 2010). The peaks assigned to CMC are almost identical to MCC except for 1622.13  $\text{cm}^{-1}$  and 1440.83  $\text{cm}^{-1}$ . Therefore, the peak formation at 1622.13  $\text{cm}^{-1}$  and 1440.83  $\text{cm}^{-1}$  due to the carboxyl group (–COO–) and carboxyl salts confirm CMC's structure and ensure successful CMC preparation (Rahaman et al., 2020; Rahaman et al., 2019).



**Figure 2:** FTIR spectra of MCC, CMC, composite (PLLA/CMC-40), and PLLA.

**Table 1:** Prominent peaks of MCC, CMC, composite (PLLA/CMC-40), and PLLA in the FTIR spectra.

Accounted bond for the Absorption ( $\text{cm}^{-1}$ )	MCC	CMC	Composites (PLLA/CMC-40)	PLLA
–OH stretching (3600-2500)	3435.22	3442.94	3439.08	3419.79
–CH stretching (Asymmetric) (3100-2900)	2916.37	2914.44	2927.94	3001.24
–C=O stretching (1780-1650)	1631.78	1622.13	1761.01 1620.21	1761.01 1620.21
–CH bending (1390-1365)	1433.11 1371.39	1440.83 1369.46	1454.33 1373.32	1458.18 1371.01
–C–O– stretching (1200-1020)	1163.08 1060.85	1163.08 1060.85	1197.79 1095.57	1195.87 1093.64

The FTIR spectrum of PLLA shows the absorption band at 3419.79 and 3001.24  $\text{cm}^{-1}$  for –OH and –C–H stretching vibrations, respectively. The peak at 873.75  $\text{cm}^{-1}$  is assigned to the stretching of –C–C– bondage and at 1093.64  $\text{cm}^{-1}$  to stretching of –C–O– bondage. On the other hand, peak at 1371.01  $\text{cm}^{-1}$  and 1458.18  $\text{cm}^{-1}$  represents the –OH and –CH<sub>3</sub> bending, respectively. The peak of –C=O bond stretching has been found at 1761.01  $\text{cm}^{-1}$  (Zhang et al., 2005; Garlotta, 2001; Liuyun et al., 2009). Composite (PLLA/CMC-40) shows slight band-shifts in the FTIR spectra. Symmetry and asymmetry stretching frequency of COO– in CMC are shifted from 1622.13  $\text{cm}^{-1}$  to 1,620.21  $\text{cm}^{-1}$ , and 1440.83  $\text{cm}^{-1}$  to 1454.33  $\text{cm}^{-1}$  in PLLA/CMC-40, respectively. Other stretching absorption bands at 2914.44  $\text{cm}^{-1}$  of –CH and 3442.94  $\text{cm}^{-1}$  of –OH in CMC shift to 2927.94  $\text{cm}^{-1}$  and 3439.08  $\text{cm}^{-1}$  in PLLA/CMC-40 (Xiao et al., 2012; Rahaman & Tsuji, 2013). The interaction between carboxylate groups of the carboxymethyl cellulose (CMC) surfaces and the terminal hydroxyl, terminal carboxyl, and carbonyl groups of PLLA through



hydrogen bonds caused the shift and broadening of the band. These results suggest that carboxymethyl cellulose (CMC) and PLLA formed an excellent composite by physical interactions without forming new functional groups.

### Wide-angle X-ray Diffraction

The diffraction patterns of PLLA/CMC biocomposite are shown in Figure 3. PLLA (PLLA/CMC-0) showing characteristic peaks at approximately 17° and 19° indicate the homo crystallinity present in the structure of PLLA and the area under the baseline shows the amorphous region of PLLA (Pushpamalar et al., 2006; Rahaman & Tsuji, 2012; Saeidlou et al., 2012). CMC (PLLA/CMC-100) shows two characteristic crystalline peaks at about 16.03° and 22.6° and other secondary peaks at higher 2θ values related to cellulose's diffraction (Xiao et al., 2012). The presence of characteristic peaks of cellulose in the XRD patterns of CMC indicates that the carboxymethylation of cellulose does not change the crystallinity and structure of cellulose. PLLA/CMC-80 composite shows two intense peaks at 16.8° and 22.6°, while PLLA/CMC-40 composite shows three intense peaks at about 16.8°, 19.0°, and 22.4°. The higher and sharper peak intensity at 16.8° indicates the presence of the crystal structure of PLLA in the biocomposites. Moreover, the peak intensity at 19.0° increases with an increase of PLLA also supports the presence in the composites. The peak at 22.4° is caused by the crystalline structure of CMC (Mondal & Haques, 2007; Rahaman et al., 2022; Morán et al., 2008; Naduparambath et al., 2018; Ikada et al., 1987; Cartier et al., 2000; Su et al., 2010).

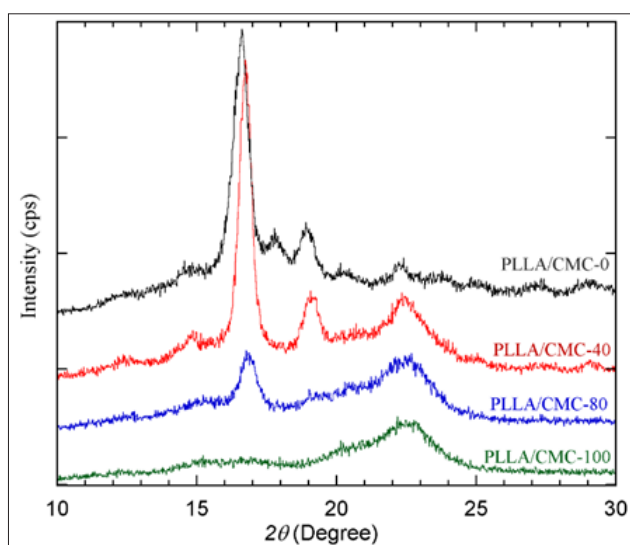


Figure 3: WAXD profiles of the composites.

The crystallinity and crystalline species (homo and stereo complex crystalline structure) were analyzed by XRD measurements. Peaks at 16.8° and 19° caused by the homo crystalline structure of pseudo-orthorhombic unit cell of dimension  $a=1.07$  nm,  $b=0.595$  nm,  $c=2.78$  nm containing  $10^3$  helical structures. These two characteristic peaks of pure PLLA (Arjmandi et al., 2015) for homo crystallinity are present in PLLA/CMC composites. The calculated percentage crystallinity of PLLA/CMC-40, PLLA/CMC-80, and PLLA/CMC-100 composites were 39, 48, and 49, respectively.

### Differential Scanning Calorimetry

The differential thermal analysis of PLLA/CMC biocomposites are shown in Figure 4. The PLLA/CMC-100 exhibits two absorption peaks. In the DSC thermogram, the first peak between 50-100 °C is probably due to the absorption of heat by moisture. In contrast, the second peak above 300 °C is probably due to the cleavage of glycosidic linkages heat absorption, which ultimately decomposed the main chain of carboxymethyl cellulose (Biswal & Singh, 2004). Biocomposites show three absorption peaks. In the DSC thermogram, the first peak between 50-100 °C is due to heat absorption by moisture. The second peak at about 166.3 °C is related to the melting temperature of PLLA. In contrast, the third peak above 300 °C is related to the decomposition temperature of carboxymethyl cellulose (CMC) decomposition (Biswal & Singh, 2004). The decomposition peak temperature of CMC was increased from 315 to 326 and 340°C by adding 20% and 60% PLLA, respectively, in the composites. Therefore, PLLA enhanced the thermal properties of CMC in composites.

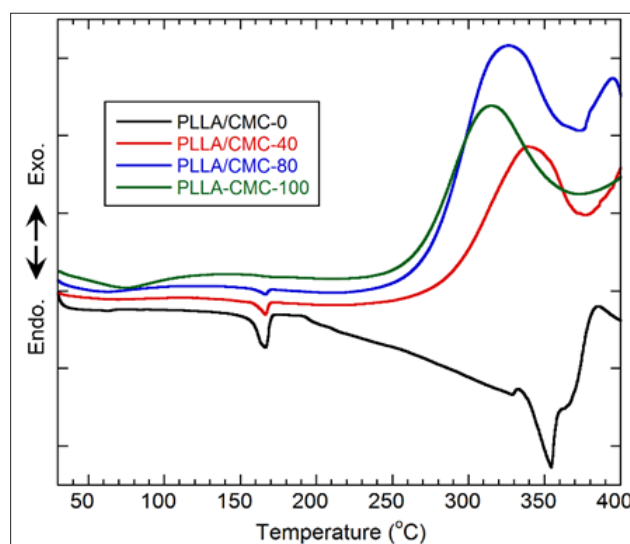
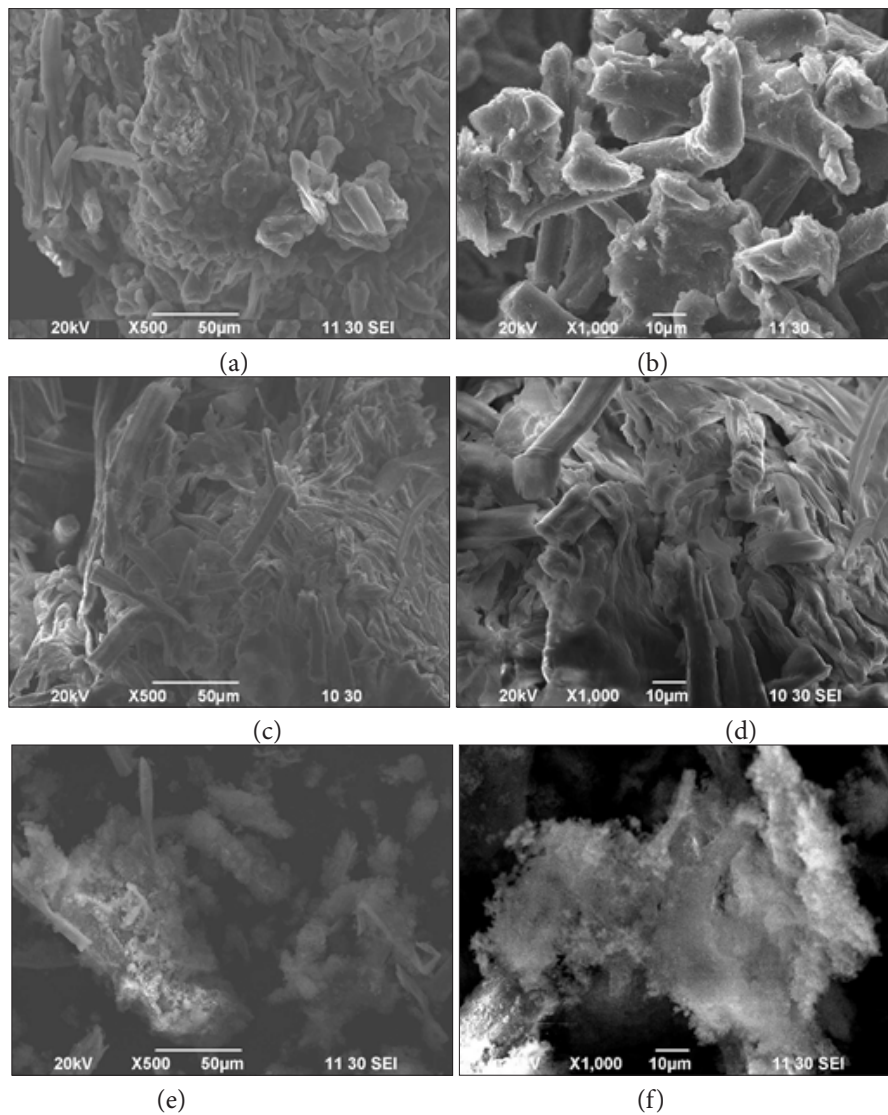


Figure 4: DSC profiles of the composites.

### Scanning Electron Microscopy

Figure 5 represents the lower (500×) and higher (1000×) magnification of the scanning electron micrographs of MCC, CMC, PLLA/CMC-40. Micro-sizes are defined as between 1 and 1000 μm particles in size. MCC micrographs (Figure 5a, 5b) show different sizes (5-50 μm) but most of them are 10-20 μm which conforms micro in size. The fibrous form of CMCs is present in the micrograph (Figure 5c, 5d), which indicates that cellulose's fibrous structure was not affected by carboxymethylation. This observation also indicated that carboxymethylation has occurred only on the surfaces of cellulose. In addition, the micrograph of PLLA/CMC-40 (Figure 5e, 5f) shows that the CMC fibers are coated with a layer of PLLA which indicates CMC and PLLA have better compatibility which was absent in reported results of SEM micrographs of PLLA/MCC composites (Rahaman et al., 2020; Rahaman et al., 2020; Hosen et al., 2017).



**Figure 5:** SEM micrographs of MCC (a, b), CMC (c, d), and PLLA/CMC-40 (e, f) at 500 and 1000 $\times$ , respectively.

### Conclusions

CMC was synthesized successfully from mustard stalks with a yield of 78%, and the composites with different ratios of PLLA and CMC were prepared by the solution casting method. The shifting peak in the FTIR spectrum of PLLA/CMC biocomposites indicates the interaction between PLLA and CMC through hydrogen bonds and forms excellent composites. The highest crystallinity was found for 80% CMC in the composites. PLLA increased the decomposition temperature of CMC from 10 to 25  $^{\circ}\text{C}$  depending on its amount in the composites. Therefore, CMC and PLLA have good compatibility, and these composites can be applied for biomedical and industrial purposes.

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### Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data Availability Statement

Data supporting this study are included within the article and/or supporting materials.

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