

ORIGINAL ARTICLE

# Colour stabilities of three types of orthodontic clear aligners exposed to staining agents

Chen-Lu Liu, Wen-Tian Sun, Wen Liao, Wen-Xin Lu, Qi-Wen Li, Yunho Jeong, Jun Liu and Zhi-He Zhao

The aim of this study was to evaluate and compare the colour stabilities of three types of orthodontic clear aligners exposed to staining agents *in vitro*. Sixty clear orthodontic aligners produced by three manufacturers (Invisalign, Angelalign, and Smartee) were immersed in three staining solutions (coffee, black tea, and red wine) and one control solution (distilled water). After 12-h and 7-day immersions, the aligners were washed in an ultrasonic cleaner and measured with a colourimeter. The colour changes ( $\Delta E^*$ ) were calculated on the basis of the Commission Internationale de l'Eclairage  $L^*a^*b^*$  colour system (CIE  $L^*a^*b^*$ ), and the results were then converted into National Bureau of Standards (NBS) units. Fourier transformation infrared (FT-IR) spectroscopy and scanning electron microscopy (SEM) were conducted to observe the molecular and morphologic alterations to the aligner surfaces, respectively. The three types of aligners exhibited slight colour changes after 12 h of staining, with the exception of the Invisalign aligners stained with coffee. The Invisalign aligners exhibited significantly higher  $\Delta E^*$  values (ranging from 0.30 to 27.81) than those of the Angelalign and Smartee aligners ( $\Delta E^*$  values ranging from 0.33 to 1.89 and 0.32 to 1.61, respectively,  $P < 0.05$ ). FT-IR analysis confirmed that the polymer-based structure of aligners did not exhibit significant chemical differences before and after the immersions. The SEM results revealed different surface alterations to the three types of aligner materials after the 7-day staining. The three types of aesthetic orthodontic appliances exhibited colour stability after the 12-h immersion, with the exception of the Invisalign aligners stained by coffee. The Invisalign aligners were more prone than the Angelalign and Smartee aligners to pigmentation. Aligner materials may be improved by considering aesthetic colour stability properties.

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**Keywords:** aesthetics; clear aligner; clear aligner material; colour stability; orthodontic appliance

## INTRODUCTION

The aesthetic considerations associated with social perceptions influence orthodontic treatment.<sup>1–2</sup> The increasing demand for more aesthetic orthodontic appliances has elicited an aesthetic revolution marked by the emergence of invisible appliances, such as aesthetic brackets, lingual appliances, and clear aligners.<sup>3–5</sup> Among these appliances, clear aligners are often preferred over brackets by adults, owing to aesthetic and comfort considerations.<sup>6–7</sup>

The clear aligner treatment process is based on the sequential use of aligners made of transparent thermoplastic materials.<sup>8</sup> Various thermoplastic materials are currently used for fabrication,<sup>9</sup> including polyvinyl chloride,<sup>10</sup> polyurethane (PU),<sup>11</sup> polyethylene terephthalate (PET),<sup>12</sup> and polyethylene terephthalate glycol (PETG).<sup>10,12</sup> Many studies have been conducted on clear aligners, particularly focusing on the mechanical properties of the aligners<sup>13–15</sup> or the biomechanics of tooth movement during orthodontic treatment.<sup>12,16</sup> Researchers have also investigated new clear aligner materials to obtain desirable

mechanical properties for orthodontic treatment.<sup>17</sup> However, although clear aligners are promising aesthetic orthodontic appliances, the aesthetic stability of clear aligner materials has scarcely been reported.

From an aesthetic perspective, the colour stability and transparency of orthodontic clear aligners should be stable during the 2-week orthodontic treatment periods.<sup>18–19</sup> However, the colour stability of dental materials is often influenced by various factors, such as ultraviolet irradiation, staining beverages, and mouthwashes.<sup>20</sup> Studies have reported that PU-based elastomeric ligatures are vulnerable to pigment adsorption from food and drink in the oral cavity.<sup>21–23</sup> During usage, it is recommended that aligners be removed before eating and drinking. However, studies have reported that patient compliance regarding removable orthodontic appliances is insufficient,<sup>24</sup> and this often is a matter of concern for orthodontists. The exposure of the aligner to staining agents in the oral cavity is inevitable, especially when users drink without taking the aligners out of their mouths because of time constraints during work. Statistics

State Key Laboratory of Oral Diseases, Department of Orthodontics, West China Hospital of Stomatology, Sichuan University, Chengdu, China

Correspondence: Professor ZH Zhao, State Key Laboratory of Oral Diseases, Department of Orthodontics, West China Hospital of Stomatology, Sichuan University, No.14, Section 3, Renmin South Road, Chengdu 610041, China

E-mail: zhzhao@scu.edu.cn

Dr J Liu, State Key Laboratory of Oral Diseases, Department of Orthodontics, West China Hospital of Stomatology, Sichuan University, No.14, Section 3, Renmin South Road, Chengdu 610041, China

E-mail: junliu@scu.edu.cn

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have demonstrated that over 50% of American adults drink coffee every day and that their average consumption is 3.2 cups per day.<sup>25</sup> For patients who do not follow the instructions, the pigments in staining agents might accumulate and lead to colour changes in the aligner materials. Thus, clear aligners might become less aesthetically appealing even during the 2-week treatments,<sup>26</sup> and this is a clinical concern. Therefore, there is a need to investigate the colour stabilities of commonly used types of aligners to provide evidence for both patients and orthodontists regarding clinical aesthetic considerations and instructions.

The aim of the present study was to evaluate and compare the colour stabilities of three types of orthodontic clear aligners before and after exposure to staining solutions *in vitro*.

## MATERIALS AND METHODS

### Staining of the aligners

Three sets of polyvinyl siloxane impressions of the upper dentition were taken from a subject. The aligners (and/or retainers) made by thermoplastic materials were manufactured by three companies (Invisalign, Align Technology, Santa Clara, CA, USA; Angelalign, EA Medical Instruments, Shanghai, China; and Smartee, Smartee Denti-Technology, Shanghai, China). According to previous studies and the material safety data sheets provided by the manufacturers, the Invisalign aligner is made of SmartTrack material, which is primarily composed of thermoplastic PU with some modification (the details are not disclosed by the manufacturer).<sup>11,27–29</sup> The Smartee aligner is manufactured with Erkodur foil (Erkodent Erich Kopp GmbH, Pfalzgrafenweiler, Germany), which is composed of PETG.<sup>30</sup>

Because coffee, black tea, and red wine have been reported to cause more severe stains than other beverages,<sup>31</sup> the present study used these liquids as staining agents and distilled water as a control.<sup>32</sup> The coffee (G7 Pure Black Instant Coffee; Trung Nguyen, Bac giang, Vietnam) and tea (Yellow Label Tea; Lipton, Hefei, China) solutions were prepared according to the methods used in previous studies.<sup>23</sup> The concentrations were 3 g of coffee powder or one teabag per 100 mL boiling distilled water.<sup>23</sup> Undiluted red wine (Cabernet Sauvignon red wine; Saflam, Yantai, China) was used. Twenty aligners (and/or retainers) from each manufacturer were randomly divided into four groups according to the solutions. The aligners were immersed in each solution in a water bath at 37 °C for 12 h or 7 days. The solutions were refreshed every day.

### Colour change evaluations

The colour changes were characterised according to the Commission Internationale de l'Éclairage  $L^*a^*b^*$  colour system (CIE  $L^*a^*b^*$ ).<sup>33</sup> The colour parameter  $L^*$  represents lightness (+ bright, – dark),  $a^*$  represents the red (+) to green (–) colour scale, and  $b^*$  indicates the yellow (+) to blue (–) colour scale.<sup>34</sup> The colour parameters  $L^*$ ,  $a^*$ , and  $b^*$  ( $t=0, 1, 2$ ) of aligners were measured with a standard VITA Easyshade Compact<sup>30</sup> colourimeter (Vita Zahnfabrik, Bad Sackingen, Germany) before staining ( $T_0$ ) and after periods of 12 h ( $T_1$ ) and 7 days ( $T_2$ ) of staining. All of the aligners were washed in an ultrasonic cleaner for 5 min and dried with tissue paper before the measurements. Standard measurements were performed by an investigator who was blind to the group division. The flat labial surface of the right upper central incisor of each aligner was measured. A tooth model made using flowable resin (Filtek Universal Restorative; 3M ESPE, Saint Paul, MN, USA) with an A3 Body Shade as was used as the background reference and set behind the labial surface of each aligner.<sup>19</sup> The measurements were performed by firmly contacting the optical sensor tip vertically to the flat surface of the aligner. All measurements were conducted in the same room with standardized

illumination. The total colour change ( $\Delta E^*$ ) value was calculated according to the equation  $\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$ , which represents the colour difference before and after staining.<sup>33</sup>  $\Delta L^*$ ,  $\Delta a^*$  and  $\Delta b^*$  ( $t=1, 2$ ) are the subtractions of the  $L^*$ ,  $a^*$  and  $b^*$  colour parameters, respectively, between  $T_1 - T_0$  and  $T_2 - T_0$ .

### Colour change rating

The national bureau of standards (NBS) system was used to describe levels of perceptible colour change upon visual inspection (Table 1).<sup>23,35</sup> The  $\Delta E^*$  values were converted into NBS units with the equation  $NBS = \Delta E^* \times 0.92$  to relate the colour changes to a clinical standard.<sup>19,36–37</sup>

### Fourier transformation infrared analysis

Because coffee caused the most marked colour changes, the molecular changes to the aligners' surfaces following coffee staining were examined with Fourier transformation infrared (FT-IR) spectroscopy.<sup>27–28</sup> Before and after 7 days of staining in coffee, part of the material (2 mm × 2 mm) was cut from the aligner ( $n=3$ ). The specimens were washed in ultrasonic cleaners for 5 min and dried with tissue paper. The spectra of the three types of aligners were recorded with an FT-IR spectrometer (Alpha, BRUKER OPTIC GMBH, Ettlingen, Germany) and averaged over 40 scans between 4 000 and 500  $\text{cm}^{-1}$  with a resolution of 4  $\text{cm}^{-1}$ . The FT-IR spectra were compared with data in the ATR-FTIR LIBRARY COMPLETE OF BRUKER (BRUKER OPTIC GMBH, Ettlingen, Germany).

### Scanning electron microscopy examination

Because coffee caused the most marked staining, scanning electron microscopy (SEM) examinations were conducted to further investigate the surface alternations to the materials that were caused by coffee staining. Small specimens (2 mm × 2 mm) were cut from coffee-stained aligners ( $n=3$ ) and then investigated with SEM (INSPECT F; FEL, Brno, Czech) at ×50 000 magnifications.

### Statistics analysis

The  $\Delta E^*$  values of each group at 12 h and 7 days were analysed *via* two-way analyses of variance (ANOVAs), and one-way ANOVAs. Tukey's multiple comparison tests were conducted to investigate the effects of each solution on the materials'  $\Delta E^*$  values.

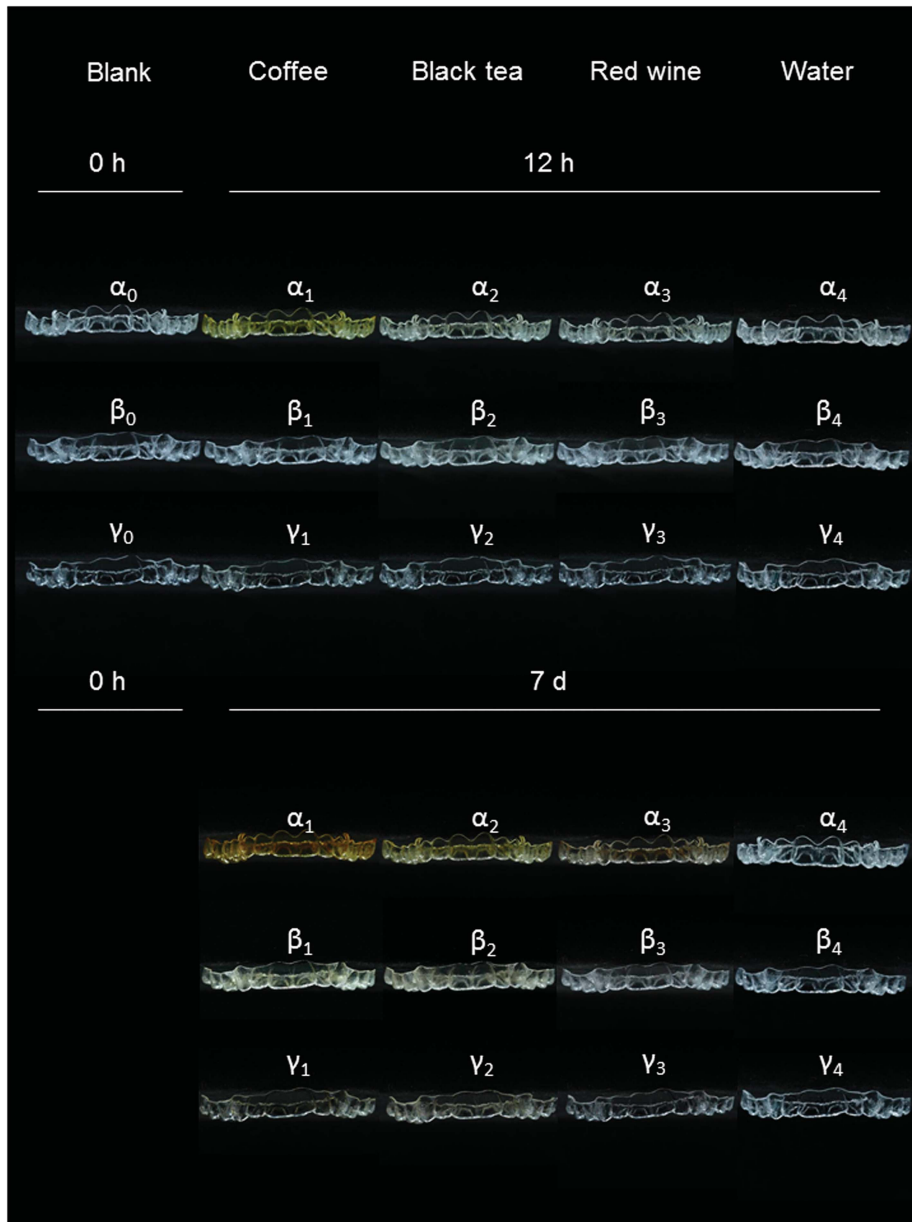
## RESULTS

### Colour change evaluations

Photographs showing the colour change of the aligners before and after immersion in coffee, black tea, red wine, and distilled water are provided in Figure 1. Visual inspection indicated that all three types of aligners exhibited almost no colour change after the 12-h staining, with the exception of the Invisalign aligners in coffee. As the immersion time increased to 7 days, the aligners exhibited enhanced colour changes. The Invisalign aligners exhibited more marked colour changes

**Table 1** National bureau of standards ratings

National bureau of standards units	Descriptions of colour changes
0.0–0.5	Trace: extremely slight change
0.5–1.5	Slight: slight change
1.5–3.0	Noticeable: perceivable
3.0–6.0	Appreciable: marked change
6.0–12.0	Much: extremely marked change
12.0 or more	Very much: change to other colour



**Figure 1** Photographs of the aligners.  $\alpha$ , Invisalign;  $\beta$ , Angelalign;  $\gamma$ , Smartee. Aligners before staining ( $\alpha_0$ ,  $\beta_0$ , and  $\gamma_0$ ) and after staining in coffee ( $\alpha_1$ ,  $\beta_1$ , and  $\gamma_1$ ), black tea ( $\alpha_2$ ,  $\beta_2$ , and  $\gamma_2$ ), red wine ( $\alpha_3$ ,  $\beta_3$ , and  $\gamma_3$ ), and distilled water ( $\alpha_4$ ,  $\beta_4$ , and  $\gamma_4$ ) for 12 h and 7 d.

than the Angelalign and the Smartee aligners in the staining solutions. No perceivable colour changes were detected in any of the three types of aligners after immersion in distilled water even after 7 days.

Quantitative evaluations were further conducted for the colour measurements. The means and standard deviations of the  $\Delta E^*$  (the colour change) values are presented in Table 2 and Figure 2. Analysis of the  $\Delta E^*$  values with two-way ANOVA identified significant differences in the colour changes among the different materials ( $P < 0.05$ ) and the different staining solutions ( $P < 0.05$ ), as well as an interaction effect ( $P < 0.05$ ) at each staining time point. The intergroup comparisons of the aligner types in each solution revealed that the Invisalign aligners exhibited significantly higher  $\Delta E^*$  values than the Angelalign and Smartee aligners ( $P < 0.05$ ), and there was no significant difference between the Angelalign and Smartee aligners (Table 2).

#### NBS rating

According to the NBS ratings (Table 1 and Figure 3), after 12 h of staining, all three types of aligners exhibited only extremely slight or slight colour changes. However, the Invisalign aligners stained with coffee exhibited marked colour change. After 7 days of staining, the Invisalign aligners exhibited extremely marked colour changes, particularly the aligners that were immersed in coffee, whereas the Angelalign and Smartee aligners exhibited only slightly perceivable colour changes. All three types of aligners maintained good colour stability in distilled water even after the 7-day immersion.

#### FT-IR analysis

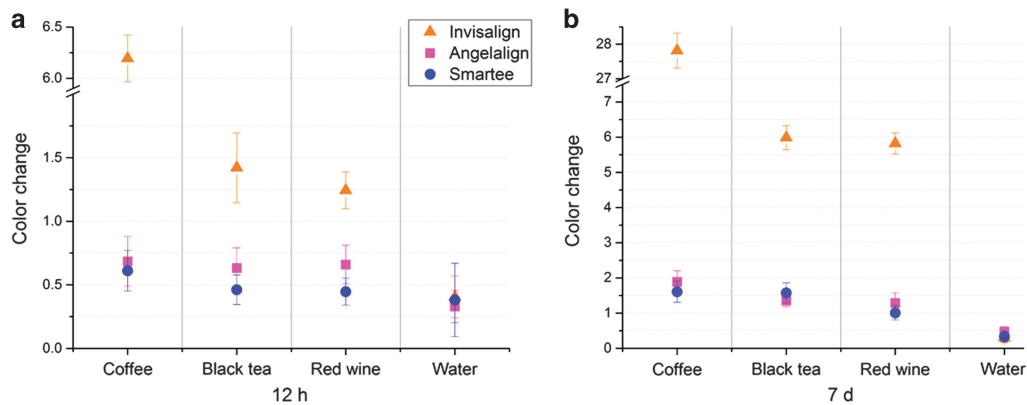
FT-IR spectra of the three types of aligners before and after immersion in coffee were analysed,<sup>38–40</sup> and the IR peaks of the different groups are

**Table 2** Comparisons of colour change ( $\Delta E^*$ ) values of three types of aligners in each solution at each time point

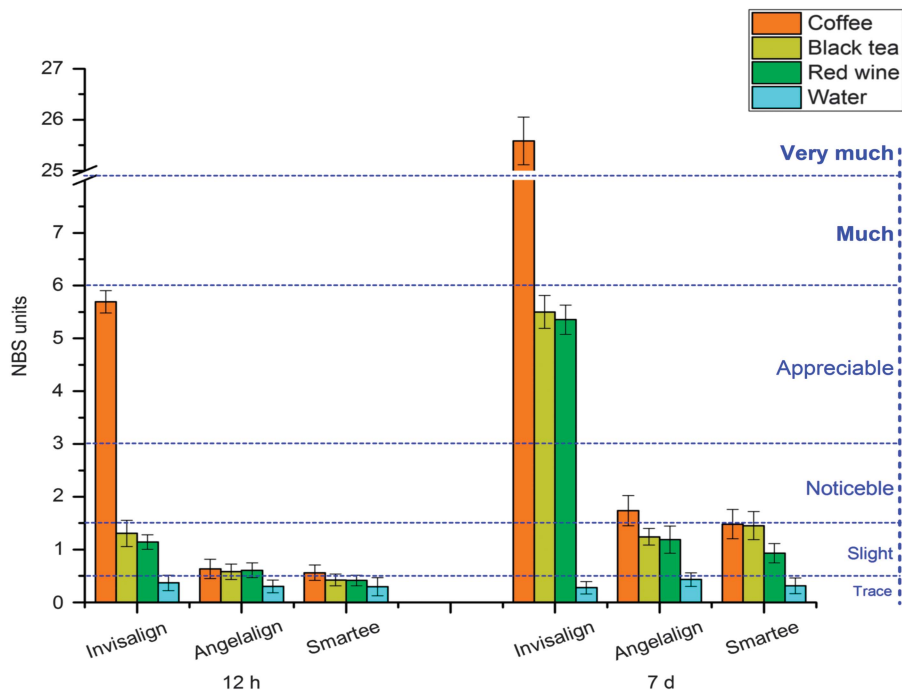
Brands	Coffee			Black tea			Red wine			Water		
	Mean	SD	IC*	Mean	SD	IC*	Mean	SD	IC*	Mean	SD	IC*
12 h												
Invisalign	6.19	0.23	a	1.42	0.27	a	1.24	0.15	a	0.40	0.16	a
Angelalign	0.69	0.20	b	0.63	0.16	b	0.66	0.15	b	0.33	0.13	a
Smartee	0.61	0.16	b	0.46	0.12	b	0.45	0.11	b	0.32	0.19	a
7 day												
Invisalign	27.81	0.51	a	5.98	0.34	a	5.82	0.30	a	0.30	0.13	a
Angelalign	1.89	0.31	b	1.35	0.17	b	1.29	0.28	b	0.47	0.14	a
Smartee	1.61	0.30	b	1.58	0.29	b	1.01	0.20	b	0.34	0.16	a

IC, Intergroup comparison; SD, standard deviation.

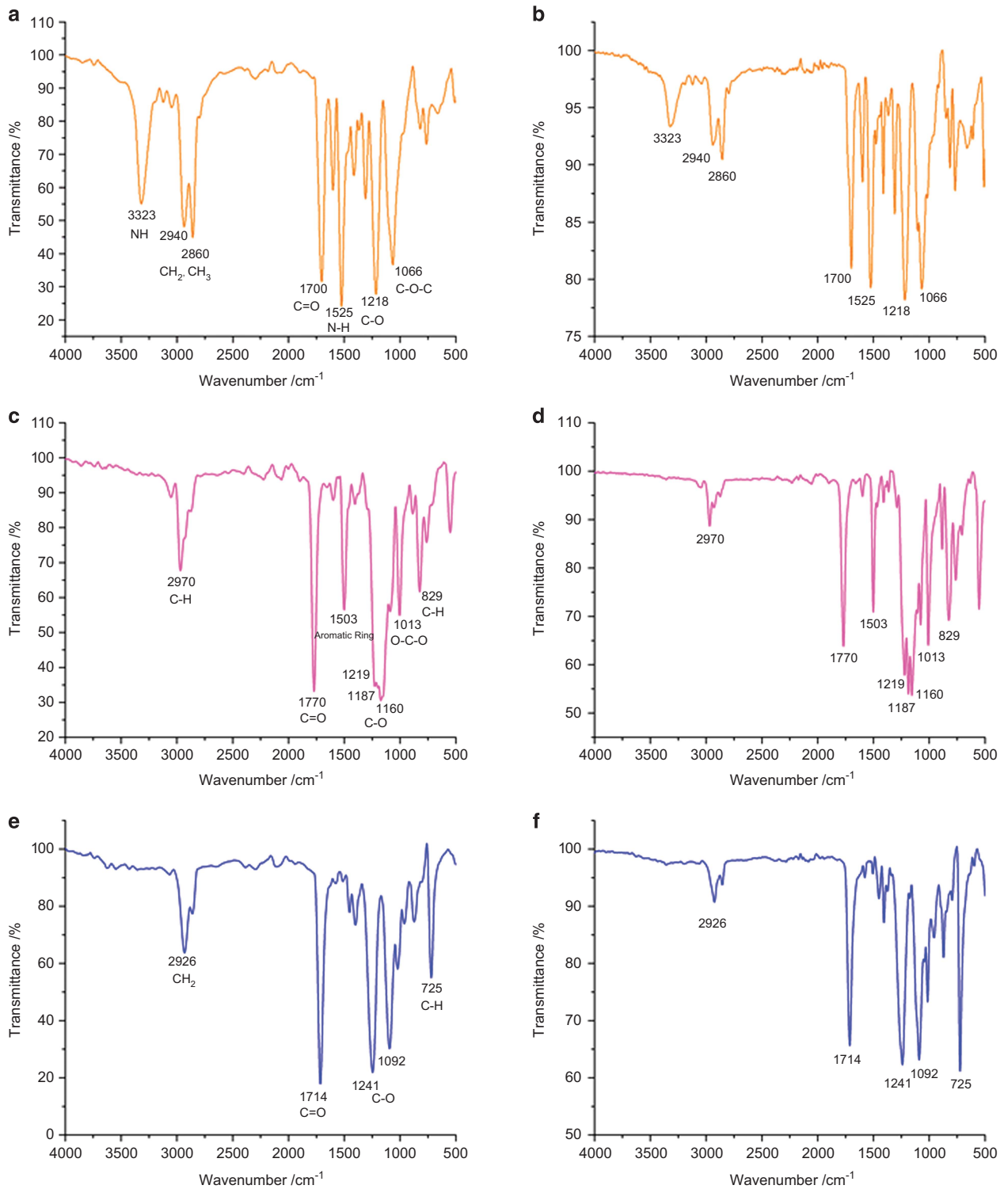
\*Intergroup comparison of colour differences regarding different aligner types according to the Tukey's multiple comparison test. Different letters indicate a statistically significant difference between aligner types in each solution at each time point ( $P < 0.05$ ).



**Figure 2** Colour changes in the three types of aligners stained in each solution. (a) Stained for 12 h; (b) for 7 d.



**Figure 3** NBS units and colour change ratings of three types of aligners. NBS, National Bureau of Standards.



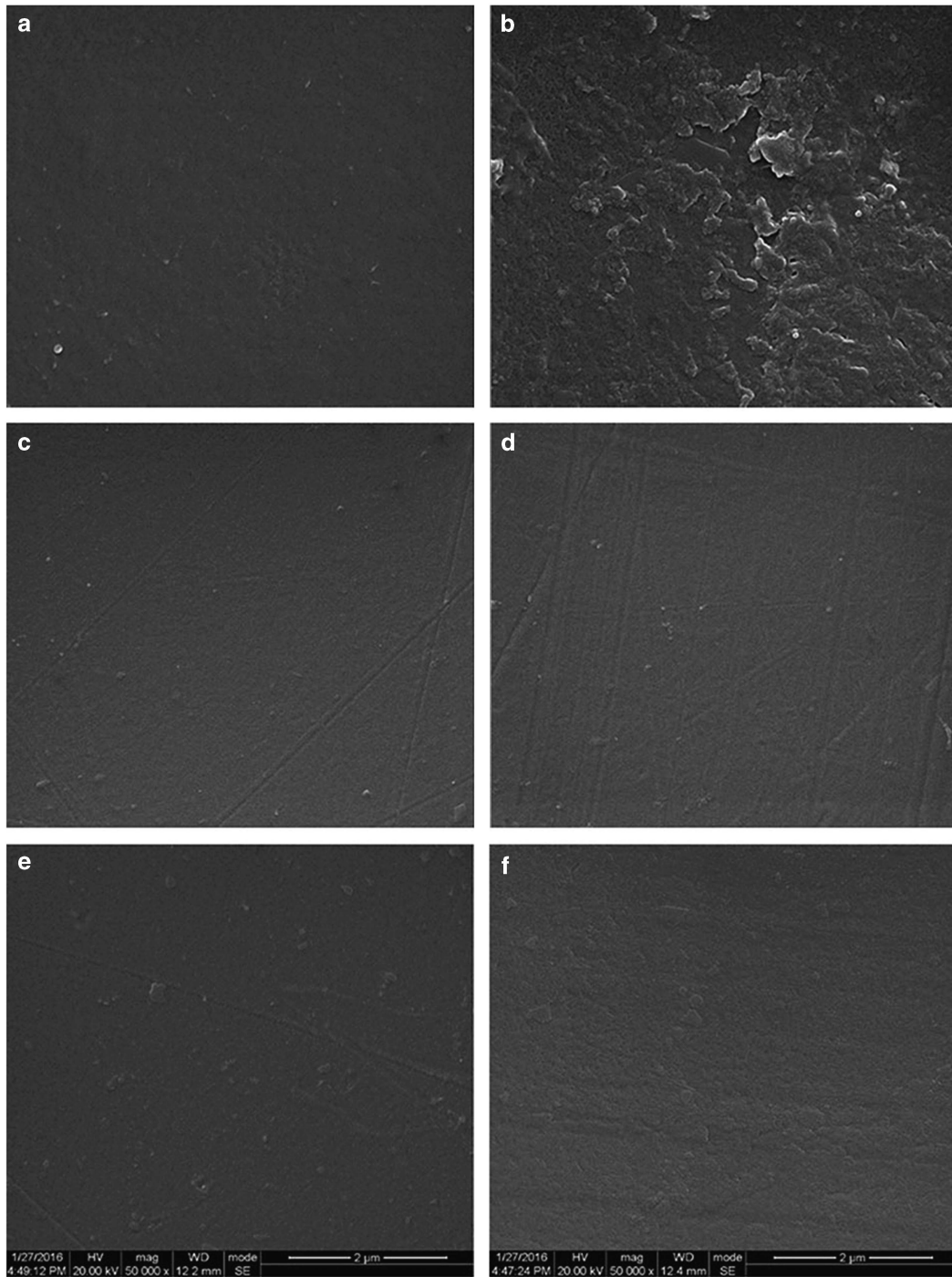
**Figure 4** FT-IR spectra of the aligner materials. (a, b) Invisalign; (c, d) Angelalign; (e, f) Smartee. (a, c, e) Before staining; (b, d, f) after staining in coffee for 7 d. FT-IR, Fourier transformation infrared.

shown in Figure 4. FT-IR analysis confirmed that the Invisalign aligners were a PU-based material,<sup>27–28</sup> the Angelalign aligners and/or retainers were a polycarbonate (PC)-based material, and the Smartee aligners were a PETG-based material. There were no significant chemical differences in any of the three materials after the coffee staining process.

#### SEM examination

The SEM results revealed that the aligner materials exhibited rough surfaces after the 7-day coffee staining (Figure 5). The Invisalign material surface tended to be rougher and to exhibit more severe peeling and pore formation than the other two materials. The Smartee





**Figure 5** SEM examination of the surface alternations of the aligners staining in coffee for 7 d. (a, b) Invisalign; (c, d) Angelalign; (e, f) Smartee. (a, c, e) Before staining; (b, d, f) after staining. SEM, scanning electron microscopy.

material exhibited fewer or less pores and milder peeling, and the Angelalign material did not exhibit obvious pores or peeling.

## DISCUSSION

PU, PC, and PETG are all amorphous polymers with a highly transparent appearance,<sup>40–41</sup> and the applications of such polymers can be found in almost all industrial branches and medical fields.<sup>13,42</sup> PU exhibits favourable features such as elasticity, flexibility, excellent

mechanical properties, chemical resistance, and ease of processing.<sup>40,43</sup> PC is widely used in orthodontic appliances, such as aesthetic brackets and clear aligners and retainers, owing to its good optical, physical, and chemical characteristics.<sup>44–46</sup> PETG is a non-crystallising amorphous co-polymer of PET and is a relatively hard material with good mechanical properties, formability, fatigue resistance, and dimensional stability.<sup>13,17</sup> Thus, all the three types of materials are widely used in the dental field. The FT-IR results suggested that all of the aligner materials were relatively chemically stable after staining in coffee for 7 days.<sup>31</sup>

The colour changes of the clear aligners may have been associated with pigment adsorption or penetration of the material surface when the aligners were immersed in the staining agents. This process might be related to the staining pigments in the agents and characteristics of the aligner materials.<sup>22–23,31,47–48</sup> In a few previous studies, coffee has been identified as the strongest staining agent among common beverages.<sup>31–32,49</sup> The Invisalign instructions for patients suggest that drinking coffee with the aligner in the mouth should be avoided.<sup>50</sup> The staining effect may be due to pigments, such as yellow dye in coffee and tea solutions.<sup>48</sup> The present study revealed that all of the tested aligners exhibited almost no colour change in the distilled water, and the PU-, PC-, and PETG-based aligners exhibited high colour stabilities after 7 days of immersion in a colourless solution. However, after exposure to staining solutions, such as coffee, black tea, and red wine, the aligners exhibited colour changes, although some of these changes were very slight.

The differences in the colour changes of the three types of aligners may have been due to their different polymer-based materials, which might have different susceptibilities to staining agents. The PU-based Invisalign aligners exhibited significantly greater colour change values than those of the PC and PETG-based aligners. Similarly, previous studies have found that PU is vulnerable to pigment adsorption and generally does not provide adequate colour stability.<sup>22–23,51</sup> PU-based orthodontic retainers and ligatures exhibit visible colour changes after immersion in beverages.<sup>23,31</sup> In contrast, PC-based aesthetic wires have been reported to maintain high levels of colour stability.<sup>19</sup> PC is generally recognised as a low-adsorption material<sup>43</sup> that can provide good colour stability. One study has also indicated that PU materials exhibit greater water absorption rates than PC and PETG in a simulated intraoral environment. Water absorption results from the attachment of the water molecules to the material surface and subsequent absorption or penetration into the material,<sup>52</sup> which might facilitate the adsorption and attachment of the pigments in the solutions to the material surfaces. Moreover, PU polymers contain the surface polar group ‘-NHCOO-’, and this structural unit is prone to the formation of hydrogen bonds that interact with hydrophilic pigments in solutions,<sup>53</sup> thus facilitating pigment adsorption onto the material. In contrast, PC and PETG polymers contain surface groups, such as ‘-COO-’ and ‘-C-O-C’, that are less polar than ‘-NHCOO-’.<sup>53</sup>

Furthermore, the variations in the colour changes among the three types of aligners may be explained by the materials’ surface characteristics, such as roughness, which might also accelerate pigment accumulation.<sup>23</sup> The SEM results in the present study suggested that the PU-based material surface tends to be rougher with more severe peeling. The porosities of the aligner surfaces may be the result of the degradation of the materials.<sup>22</sup> When aligners are worn in the oral cavity, their surface roughness would be more prominent because of microcracks and abrasions from speech, swallowing, and bruxism, which could enhance pigmentation and lead to the loss of the transparency of the thermoplastic aligner.<sup>23,54</sup>

The present results suggested that all the three types of aligners exhibit minimal colour changes after 12-h staining, with the exception of the Invisalign aligners stained by coffee. It has been reported that 50% of Americans adults drink coffee every day and consume an average of 3.2 cups per day, drinking each cup over the course of 15 min.<sup>25,32</sup> According to a previous calculation,<sup>32</sup> 12 h of *in vitro* staining simulates 2 weeks’ coffee exposure time in clinical aligner usage. This result illustrates the aesthetic stabilities of the three types of clear aligners, particularly the PC and PETG-based aligners. However, to maintain the colour stabilities of the aligners for clinical aesthetics and to protect the mechanical properties of aligners, patients should follow the instructions to remove the aligners before eating and

drinking. Orthodontists are supposed to inform patients of the instructions, especially patients who are coffee drinkers.

Orthodontic aligner appearance may influence social perceptions and thus is a factor in the selection of aligners by patients.<sup>1</sup> In contrast, the functional performance of clear aligners in terms of mechanical properties and force delivery features are also vital for orthodontic treatment. Although the colour of the PU-based aligners is relatively less stable, particularly after coffee exposure, the soft PU elastomer aids in improving the elasticity and is able to provide continuous light force to the teeth, which is beneficial for orthodontic tooth movement.<sup>14,30,40</sup> Considering the aesthetic and functional improvements of aligner materials, further studies should be conducted to improve the colour stability and mechanical properties of aligner material. Because the pigment adsorption is related to polymer surface polar groups and roughness, increases in the surface hydrophobicity and integrity might reduce pigment adsorption and penetration. Studies have been conducted regarding the modification of material surfaces with low surface energy by using fluorinated or silicon compounds to fabricate superhydrophobic surfaces.<sup>55–56</sup> Thus, fluorinated or silicon compound-modified PU might have the potential to achieve better colour stability.<sup>57–58</sup> Researchers have also attempted to improve the functional properties of the relatively colour-stable PETG material. Using the blending modification method, PETG/PC/thermoplastic PU polymer has been produced and used as an aligner material with optimal mechanical properties.<sup>17</sup>

## CONCLUSIONS

As aesthetic orthodontic appliances, the three types of clear aligners (Invisalign, Angelalign and Smartee) exhibited colour stability after 12-h immersions, with the exception of the Invisalign aligners stained with coffee. The Invisalign aligners were more prone to pigmentation than the Angelalign and Smartee aligners. Aligner materials may be improved in terms of their aesthetic stability properties.

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