Supplementary Material

An investigation into the effects of ink formulations of semi-solid syringe extrusion 3D printing on the performance of the printed solid dosage forms

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Formulation of ink base

Figure S1 shows the ink status of polymers dissolved in water after manually stirring for 5 minutes at room temperature (21°C). **Figure S1a** shows 15% w/v HPMC after stirring for 5 minutes - 15HPMC did not mix well in the water and formed large clumps. While increasing the HPMC concentration to 30% w/v and thoroughly stirring for 5 minutes, as shown in **Figure S1b**, the 30HPMC became fully solid due to the high concentration, and there was no free water remaining in the beaker. **Figure S1c** shows 20% w/v PVP in water, where a uniform solution formed quickly upon stirring but it would not be able to proceed for filament printing. **Figure S1d** shows the mixtures of 15% w/v HPMC and 20% w/v PVP. Homogeneous and smooth ink formed after 5 minutes of stirring. Thus, ink composed of HPMC+PVP is a good candidate for SSE 3D printing.

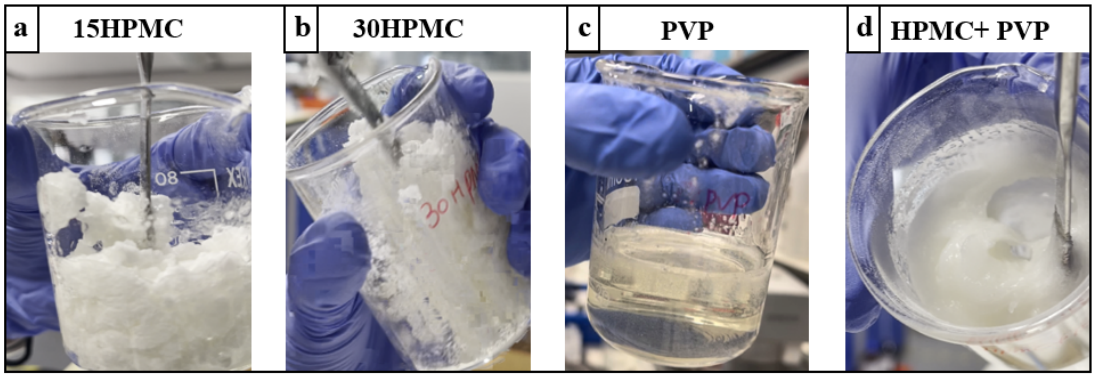

Figure S1. Ink formulation (a) 15% w/v HPMC, (b) 30% w/v HPMC, (c) 20% w/v PVP and (d) 15% w/v HPMC+20% w/v PVP in 20 mL water and stirring for 5 minutes at room temperature (21 °C).

Water content of printed tablet

Water content of printed tablets was identified through thermogravimetric analysis. **Figure S2** shows the TGA profiles of dried HPMC+PVP+15TXA and HPMC+PVP+5PAC samples. A weight percentage reduction (i.e., loss of water content) of HPMC+PVP+15TXA and HPMC+PVP+5PAC up to 100 °C at 1.45±0.10% and 0.94±0.31%, respectively was noted.

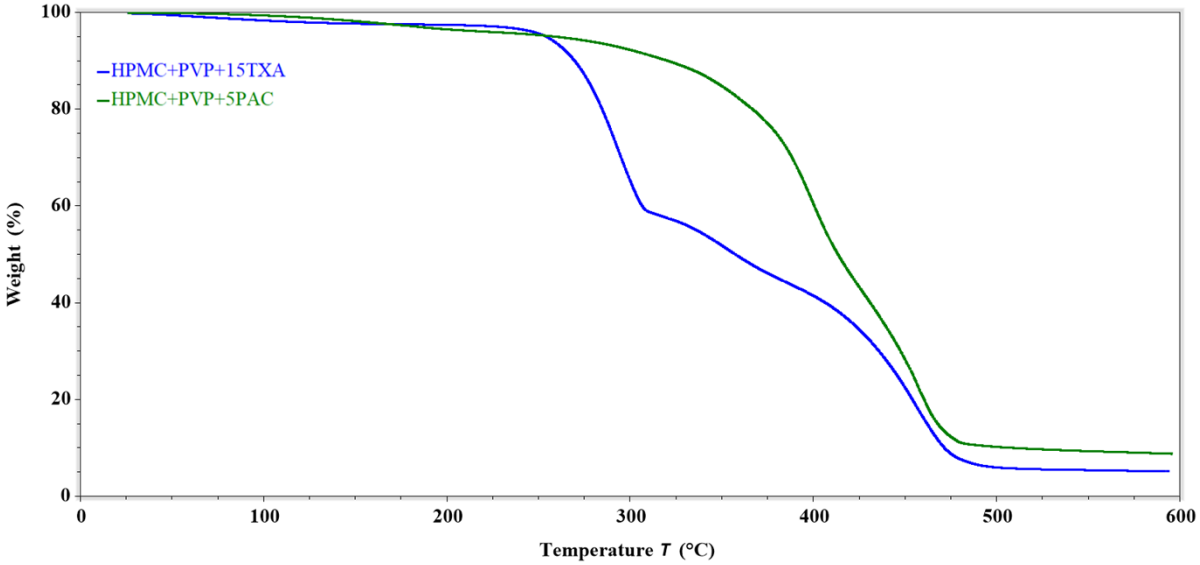

Figure S2. TGA profile of dried HPMC+PVP+15TXA and HPMC+PVP+5PAC samples.

Preliminary thermal analysis

Preliminary thermal analysis was performed using a differential scanning calorimeter (DSC) (TA Instruments, Delaware, United States) to acquire the melting/glass transition temperatures of the raw materials and dried SSE printed samples. Trios software was used for the data analysis on the duplicated samples, and a mean value was used to represent the results. A sample of 3–5 mg was accurately weighed in an aluminium crimped DSC pan and sealed using a lid with a pinhole. All samples were tested from 0°C to 250 °C at a rate of 5 °C/min. Nitrogen purge gas with a flow rate of 50 mL/min was used throughout the experiments. All measurements were performed in triplicate on three different tablets.

Figure S3a shows the thermograms of pure HPMC, PVP and PAC powder. It was clearly noted that the melting point of PAC is 169.49±0.42°C. **Figure S3b** shows the thermograms of the physical mixture HPMC+PVP and dried SSE printed HPMC+PVP. No significant shift ($p>0.05$) of PAC melting peak in the physical mixture HPMC+PAC and dried samples of SSE

printed HPMC+PAC was observed as compared to pure PAC, indicating no interaction between HPMC and PAC as agreed in the FTIR study. **Figure S3c** shows the thermograms of the physical mixture HPMC+PVP+PAC and dried SSE printed HPMC+PVP+PAC. A reduction in enthalpy of the melting peak and melting temperature (159.50±2.15^{o}C) of PAC was observed in the physical mixture HPMC+PVP+PAC could be due to the dissolution of PAC in molten PVP prior to its melting. Interestingly, no PAC melting peak was observed in the dried SSE printed HPMC+PVP+PAC, indicating a reduction of PAC crystallinity within the sample. This could be attributed to the hydrogen bond interaction between PAC and PVP as proved in the FTIR study (Figure 6). Further investigation using a modulated mode would be beneficial in observing a clearer thermal profile transition of samples.

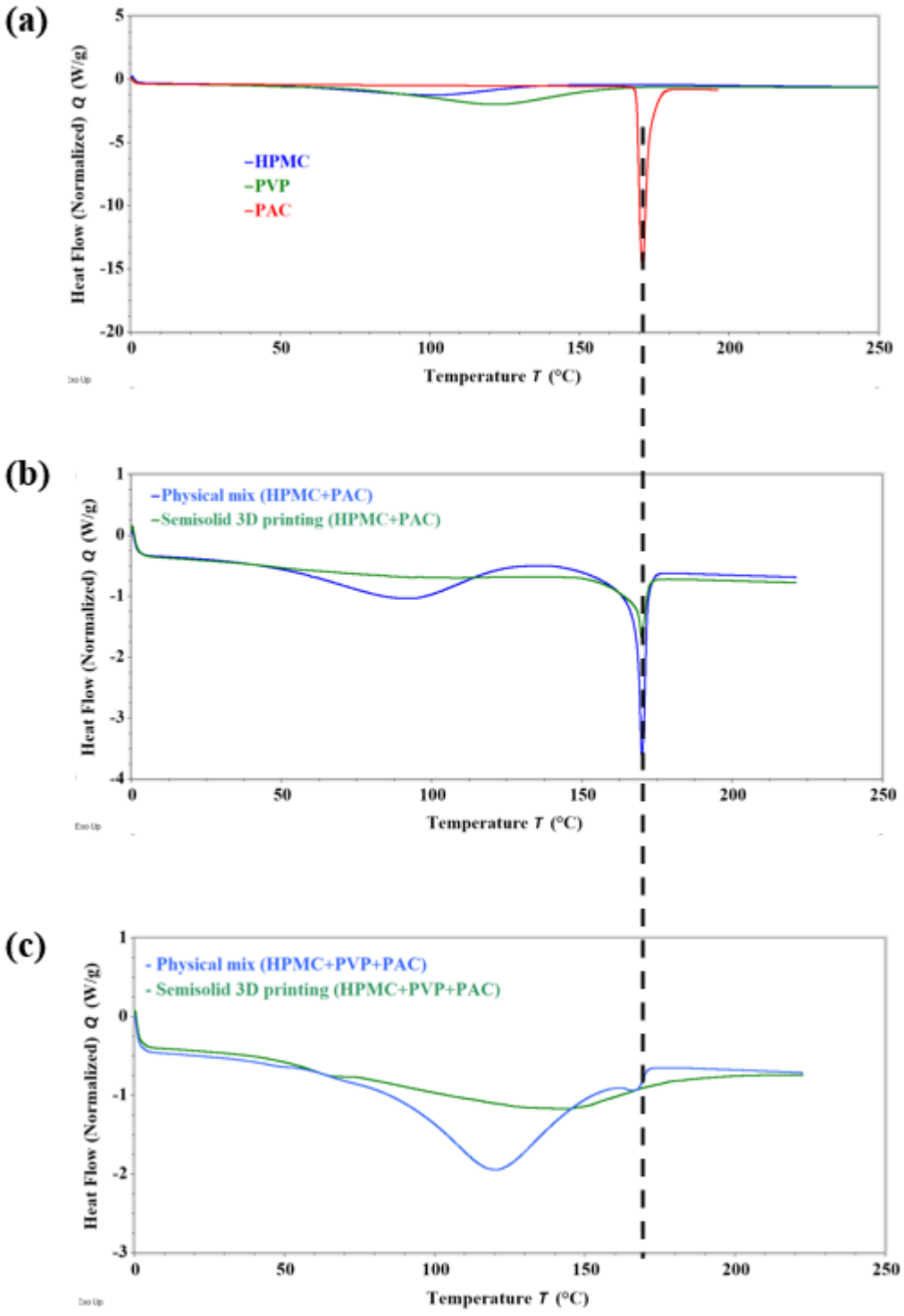

Figure S3. Thermograms of (a) pure HPMC, PVP and PAC powder, (b) physical mixture of HPMC+PAC and dried SSE HPMC+PAC sample, and (c) physical mixture of HPMC+PVP+PAC and dried SSE HPMC+PVP+PAC sample. The black dashed line represents the melting point of PAC powder.