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# Fabrication of electroactive poly(vinylidene fluoride) through non-isothermal crystallization and supercritical $CO_2$ processing

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## I. Peak to valley height ratio method

**Fig. S1** visualizes the definition of $\Delta H_\beta$ and $\Delta H_\gamma$, the height difference between the peaks at 1275 $cm^{-1}$ and 1234 $cm^{-1}$ to their nearest valleys, respectively. The wavenumbers of the valleys differ depending on the FTIR absorbance curve but they are approximately at 1260 and 1225 $cm^{-1}$ for $\Delta H_\beta$ and $\Delta H_\gamma$, respectively.

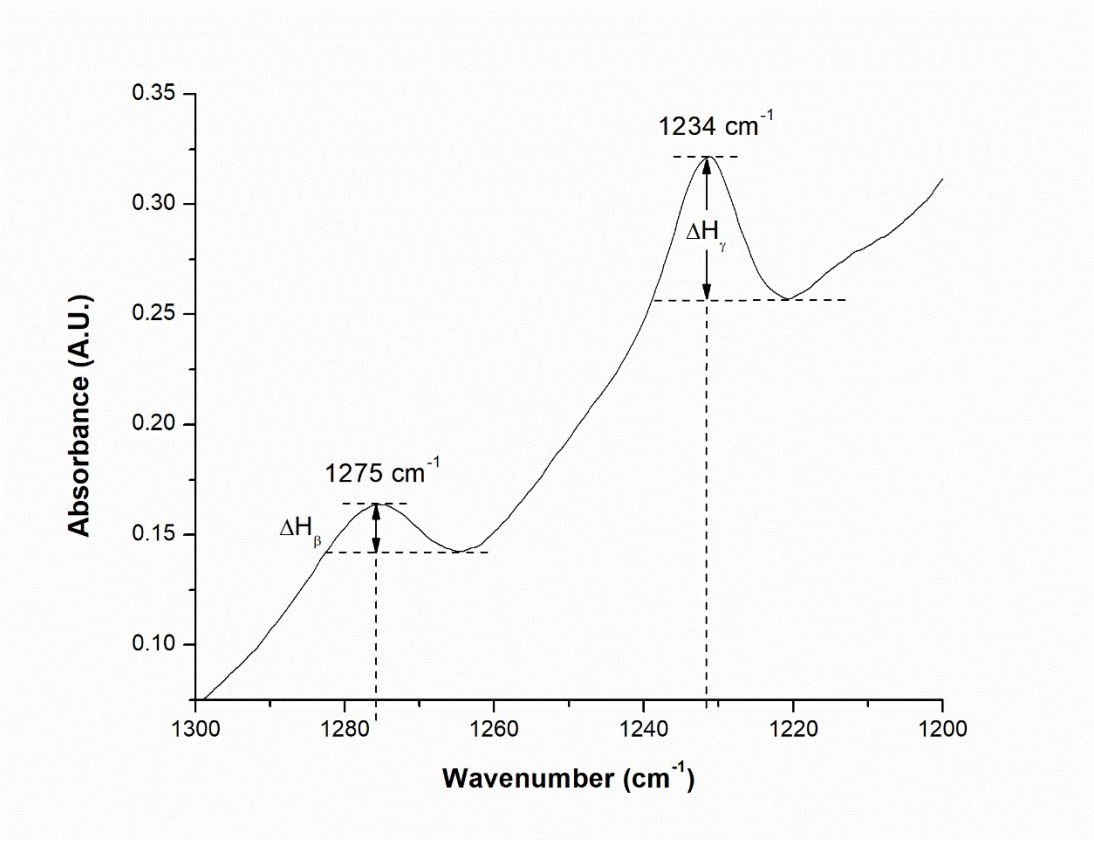


**Figure S1** Visual representation of $\Delta H_\beta$ and $\Delta H_\gamma$ for the peak to valley height ratio method

## II. Volume expansion ratios of samples prepared using temperature profile 1

Equations (1) and (2) were used to determine the volume expansion ratio (*VER)* of $ScCO_2$ processed PVDF samples. Each condition used at least three samples to calculate the average *VER* and the standard deviations. All *VER* had a standard deviation smaller than 0.55 except the samples prepared at saturation pressure and temperature at 2500 psi and 160°C, respectively, with a standard deviation of 2.57. The slightly larger standard deviation did not affect the overall trend of the relationship between the *VER* and the $ScCO_2$ processing conditions.

**Table S1** Raw data for volume expansion ratio for temperature profile 1

| Saturation Pressure (psi) | Saturation Temperature (°C) | Average *VER* | Standard Deviation |
|---|---|---|---|
| 1200 | 100 | 1.02 | 0.01 |
| 1200 | 120 | 1.02 | 0.01 |
| 1200 | 140 | 1.03 | 0.02 |
| 1200 | 160 | 3.78 | 0.41 |
| 1200 | 180 | 1.26 | 0.08 |
| 2000 | 100 | 1.02 | 0.01 |
| 2000 | 120 | 1.03 | 0.01 |
| 2000 | 140 | 1.04 | 0.01 |
| 2000 | 160 | 15.35 | 0.52 |
| 2000 | 180 | 1.22 | 0.08 |
| 2500 | 100 | 1.01 | 0.04 |
| 2500 | 120 | 1.03 | 0.01 |
| 2500 | 140 | 1.08 | 0.01 |
| 2500 | 160 | 12.76 | 2.57 |
| 2500 | 180 | 1.18 | 0.04 |

## III. Degree of crystallinity of samples prepared using temperature profile 1

Equation (3) was used to determine the degree of crystallinity ($X_C$). Each condition used at least three samples to calculate the average $X_C$ and standard deviation. "Compression molded" are samples without undergoing non-isothermal crystallization or $ScCO_2$ processing. The $X_C$ for samples prepared using temperature profile 1 had an average standard deviation of 1.35.

**Table S2** Raw data for degree of crystallinity for temperature profile 1

| Saturation Pressure (psi) | Saturation Temperature (°C) | Average $X_C$ (%) | Standard Deviation (%) |
|---|---|---|---|
| Compression Molded | - | 43.61 | 4.62 |
| No $CO_2$ | 100 | 55.51 | 0.97 |
| No $CO_2$ | 120 | 54.80 | 0.69 |
| No $CO_2$ | 140 | 51.96 | 1.45 |
| No $CO_2$ | 160 | 51.17 | 0.30 |
| No $CO_2$ | 180 | 40.61 | 0.93 |
| 1200 | 100 | 52.67 | 1.41 |
| 1200 | 120 | 54.17 | 1.53 |
| 1200 | 140 | 51.72 | 2.66 |
| 1200 | 160 | 47.85 | 1.75 |
| 1200 | 180 | 41.43 | 1.30 |
| 2000 | 100 | 48.15 | 1.32 |
| 2000 | 120 | 53.20 | 0.80 |
| 2000 | 140 | 53.49 | 1.08 |
| 2000 | 160 | 43.93 | 1.62 |
| 2000 | 180 | 40.34 | 1.73 |
| 2500 | 100 | 51.16 | 0.13 |
| 2500 | 120 | 51.68 | 1.17 |
| 2500 | 140 | 53.40 | 1.23 |
| 2500 | 160 | 46.74 | 0.95 |
| 2500 | 180 | 42.09 | 0.66 |

## IV. Volume expansion ratios of samples prepared using temperature profile 2

This analysis was identical to the *VER* of samples prepared using temperature profile 1, using Equations (1) and (2). Each condition used at least three to calculate for the average *VER* and standard deviations.

**Table S3** Raw data for volume expansion ratio for temperature profile 2

| Saturation Pressure, $P_{sat}$ (psi) | Heating Temperature, $T_H$ (°C) | Saturation Temperature, $T_{sat}$ (°C) | Pressure Drop Rate, $-dP/dt$ (MPa/s) | Average *VER* | Standard Deviation |
|---|---|---|---|---|---|
| 2000 | 180 | 160 | 190.6 | 4.52 | 0.37 |
| 2000 | 180 | 160 | 55.2 | 2.73 | 0.19 |
| 2000 | 200 | 160 | 190.6 | 12.00 | 2.07 |

## V. Degree of crystallinity of samples prepared using temperature profile 2

**Table S4** Raw data for degree of crystallinity for temperature profile 2

| Saturation Pressure, $P_{sat}$ (psi) | Heating Temp., $T_H$ (°C) | Saturation Temp., $T_{sat}$ (°C) | Pressure Drop Rate, $-dP/dt$ (MPa/s) | Average $X_C$ (%) | Standard Deviation (%) |
|---|---|---|---|---|---|
| No $CO_2$ | 180 | 160 | 190.6 | 51.13 | 2.01 |
| 2000 | 180 | 160 | 190.6 | 52.40 | 2.02 |
| 2000 | 180 | 160 | 55.2 | 45.85 | 1.22 |
| 2000 | 200 | 160 | 190.6 | 54.98 | 2.38 |

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## VI. Scanning electron microscopy of samples prepared using temperature profile 2 for the observation of PVDF's microstructures

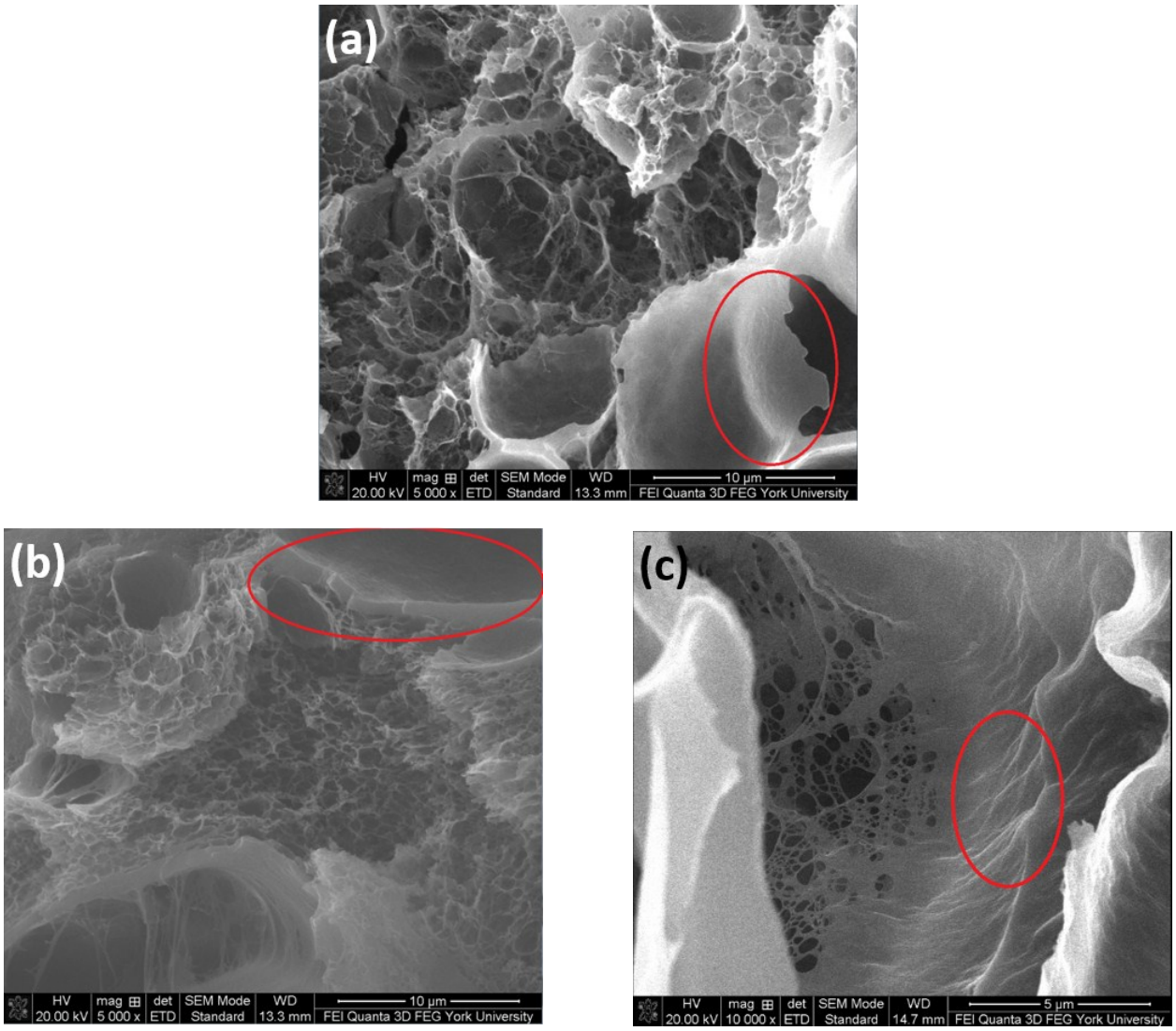


**Figure S2** SEM micrographs of PVDF samples showcasing their microstrucutres after being processed by $ScCO_2$ using temperature profile 2 at 2000 psi, $T_{sat}$ of 160°C and $T_H$ of: (a) 180°C with $-dP/dt$ = 190.6 MPa/s at 5000x; (b) 180°C with $-dP/dt$ = 55.2 MPa/s at 5000x; (c) 200°C with with $-dP/dt$ = 190.6 MPa/s at 10 000x