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Supporting Information

for

Micro-dynamic mechanism of the phase transition behavior of poly(*N*-isopropylacrylamide-*co*-2-hydroxyethyl methacrylate) hydrogel revealed by two-dimensional correlation spectroscopy

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Table S1. The sequential orders of process I (21.8−31.4 °C) obtained from the synchronous and asynchronous 2D correlation FTIR spectra of **Fig. 7**.

Cross peaks (cm^{-1}, cm^{-1})	Synchronous	Asynchronous	Sequential order
(2990, 2947)	+	+	2990→2947
$2990cm^{-1} \rightarrow 2947cm^{-1}$			
(1716, 1706)	+	+	1716→1706
(1716, 1688)	+	-	1688→1716
(1706, 1688)	+	-	1688→1706
$1688cm^{-1} \rightarrow 1716cm^{-1} \rightarrow 1706cm^{-1}$			
(1652, 1648)	+	+	1652→1648
(1652, 1623)	-	+	1623→1652
(1652, 1618)	-	+	1618→1652
(1648, 1623)	-	+	1623→1648
(1648, 1618)	-	+	1618→1648
(1623, 1618)	+	+	1623→1618
$1623cm^{-1} \rightarrow 1618cm^{-1} \rightarrow 1652cm^{-1} \rightarrow 1648cm^{-1}$			
(2990, 1716)	+	-	1716→2990
(2990, 1706)	+	-	1706→2990
(2990, 1688)	+	-	1688→2990
(2947, 1716)	+	-	1716→2947
(2947, 1706)	+	-	1706→2947
(2947, 1688)	+	-	1688→2947
$1688cm^{-1} \rightarrow 1716cm^{-1} \rightarrow 1706cm^{-1} \rightarrow 2990cm^{-1} \rightarrow 2947cm^{-1}$			
(2990, 1652)	-	+	1652→2990
(2990, 1648)	-	+	1648→2990
(2990, 1623)	+	-	1623→2990
(2990, 1618)	+	-	1618→2990
(2947, 1652)	-	+	1652→2947
(2947, 1648)	-	+	1648→2947
(2947, 1623)	+	-	1623→2947
(2947, 1618)	+	-	1618→2947
$1623cm^{-1} \rightarrow 1618cm^{-1} \rightarrow 1652cm^{-1} \rightarrow 1648cm^{-1} \rightarrow 2990cm^{-1} \rightarrow 2947cm^{-1}$			
(1716, 1652)	-	-	1716→1652
(1716, 1648)	-	-	1716→1648
(1716, 1623)	+	+	1716→1623
(1716, 1618)	+	+	1716→1618
(1706, 1652)	-	+	1652→1706
(1706, 1648)	-	+	1648→1706
(1706, 1623)	+	-	1623→1706
(1706, 1618)	+	-	1618→1706
(1688, 1652)	-	-	1688→1652
(1688, 1648)	-	-	1688→1648
(1688, 1623)	+	+	1688→1623
(1688, 1618)	+	+	1688→1618

$1688cm^{-1} \rightarrow 1716cm^{-1} \rightarrow 1623cm^{-1} \rightarrow 1618cm^{-1} \rightarrow 1652cm^{-1} \rightarrow 1648cm^{-1} \rightarrow 1706cm^{-1}$
$1688cm^{-1} \rightarrow 1716cm^{-1} \rightarrow 1623cm^{-1} \rightarrow 1618cm^{-1} \rightarrow 1652cm^{-1} \rightarrow 1648cm^{-1} \rightarrow 1706cm^{-1} \rightarrow 2990cm^{-1} \rightarrow 2947cm^{-1}$

Table S2. The sequential orders of process II (31.4−36.5 °C) obtained from the synchronous and asynchronous 2D correlation FTIR spectra of **Fig. 8**.

Cross peaks (cm^{-1}, cm^{-1})	**Synchronous**	**Asynchronous**	**Sequential order**
(2990, 2947)	+	-	2947→2990
$2947cm^{-1} \rightarrow 2990cm^{-1}$			
(1716, 1706)	+	+	1716→1706
(1716, 1688)	+	+	1716→1688
(1706, 1688)	+	+	1706→1688
$1716cm^{-1} \rightarrow 1706cm^{-1} \rightarrow 1688cm^{-1}$			
(1652, 1648)	+	-	1648→1652
(1652, 1623)	-	+	1623→1652
(1652, 1618)	-	+	1618→1652
(1648, 1623)	-	+	1623→1648
(1648, 1618)	-	+	1618→1648
(1623, 1618)	+	-	1618→1623
$1618cm^{-1} \rightarrow 1623cm^{-1} \rightarrow 1648cm^{-1} \rightarrow 1652cm^{-1}$			
(2990, 1716)	+	-	1716→2990
(2990, 1706)	+	-	1706→2990
(2990, 1688)	+	+	2990→1688
(2947, 1716)	+	+	2947→1716
(2947, 1706)	+	+	2947→1706
(2947, 1688)	+	+	2947→1688
$2947cm^{-1} \rightarrow 1716cm^{-1} \rightarrow 1706cm^{-1} \rightarrow 2990cm^{-1} \rightarrow 1688cm^{-1}$			
(2990, 1652)	-	-	2990→1652
(2990, 1648)	-	-	2990→1648
(2990, 1623)	+	-	1623→2990
(2990, 1618)	+	-	1618→2990
(2947, 1652)	-	-	2947→1652
(2947, 1648)	-	-	2947→1648
(2947, 1623)	+	+	2947→1623
(2947, 1618)	+	+	2947→1618
$2947cm^{-1} \rightarrow 1618cm^{-1} \rightarrow 1623cm^{-1} \rightarrow 2990cm^{-1} \rightarrow 1648cm^{-1} \rightarrow 1652cm^{-1}$			
(1716, 1652)	-	-	1716→1652
(1716, 1648)	-	-	1716→1648
(1716, 1623)	+	-	1623→1716
(1716, 1618)	+	-	1618→1716
(1706, 1652)	-	-	1706→1652
(1706, 1648)	-	-	1706→1648
(1706, 1623)	+	-	1623→1706
(1706, 1618)	+	-	1618→1706

(1688, 1652)	-	+	1652→1688
(1688, 1648)	-	+	1648→1688
(1688, 1623)	+	-	1623→1688
(1688, 1618)	+	-	1618→1688
$1618cm^{-1} \rightarrow 1623cm^{-1} \rightarrow 1716cm^{-1} \rightarrow 1706cm^{-1} \rightarrow 1648cm^{-1} \rightarrow 1652cm^{-1} \rightarrow 1688cm^{-1}$			
$2947cm^{-1} \rightarrow 1618cm^{-1} \rightarrow 1623cm^{-1} \rightarrow 1716cm^{-1} \rightarrow 1706cm^{-1} \rightarrow 2990cm^{-1} \rightarrow 1648cm^{-1} \rightarrow 1652cm^{-1} \rightarrow 1688cm^{-1}$			