

## Supplementary Materials

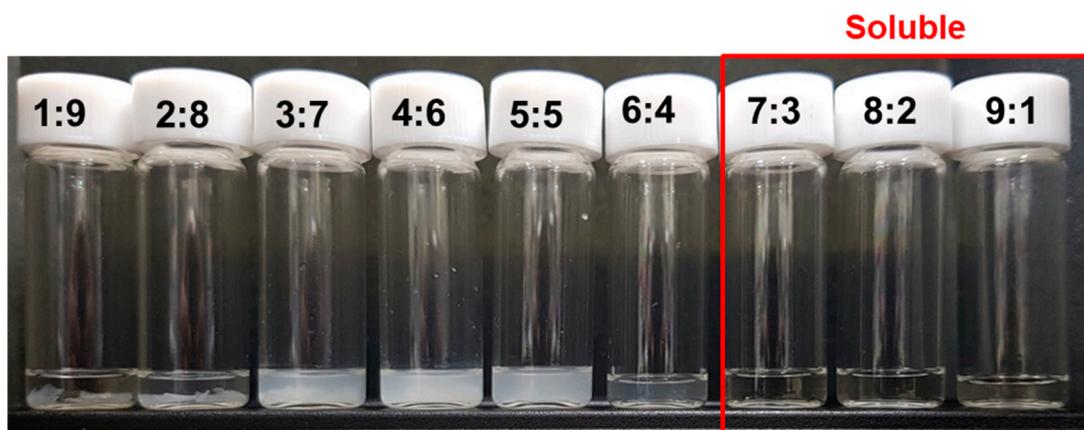
### Different Molecular Interaction between Collagen and $\alpha$ - or $\beta$ -Chitin in Mechanically Improved Electrospun Composite

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**Figure S1.** Representatively conducted solubility test of  $\alpha$ -chitin in various ratio of HFIP:TFA solvent (all the solution concentrations were 1% (w/v)). Pure HFIP and TFA were excluded owing to its extremely poor solubility on chitin.

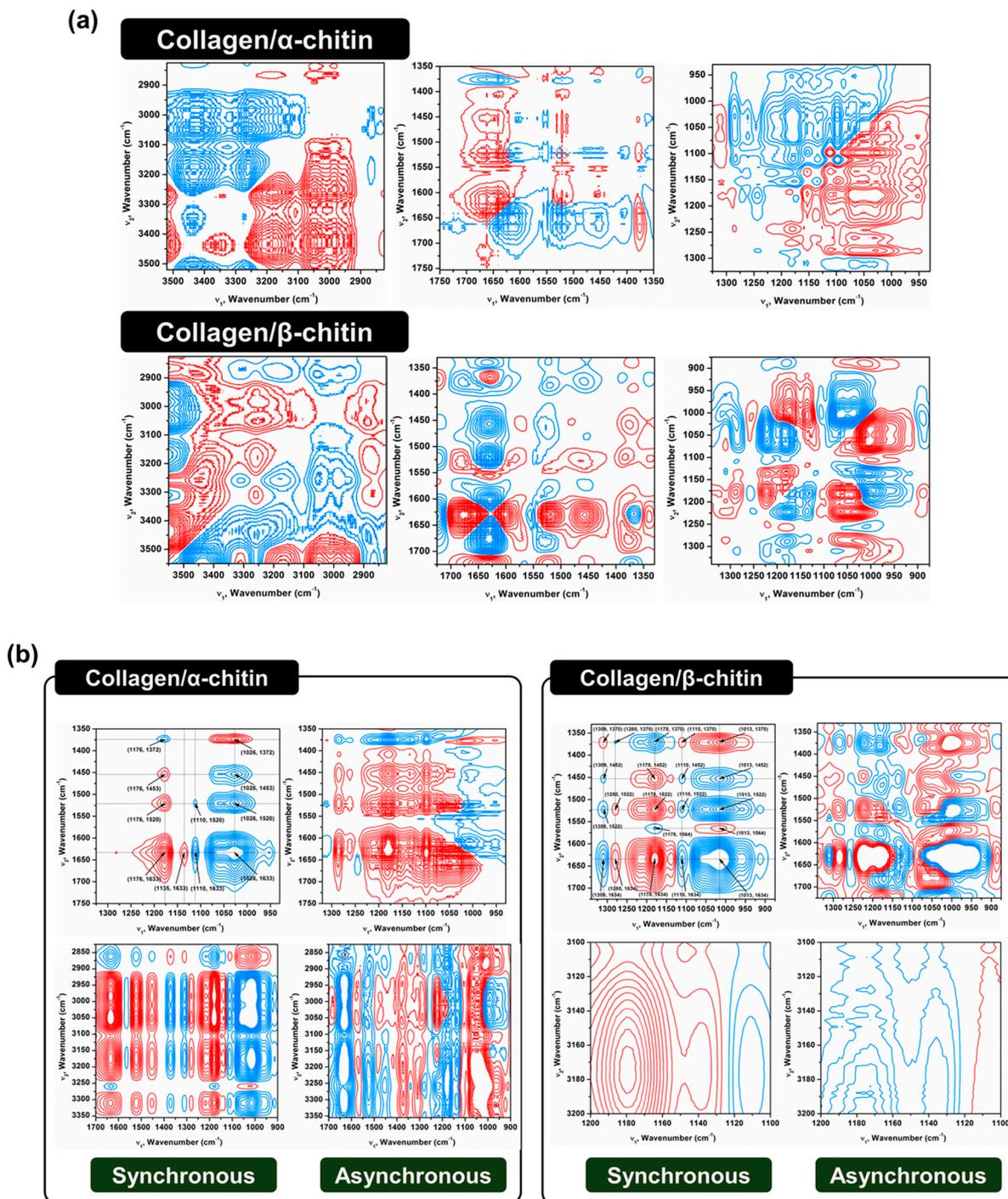


Figure S2. (a) Asynchronous 2DCOS and (b) heterogeneous contour map.

**Table S1.** 2DCOS analysis between collagen and  $\alpha$ -chitin and corresponding sequences and intensity changes.

Sequential order	Wavenumber (cm <sup>-1</sup> )	Intensity change	Assignment	Origin	Reference
1	3436	▼	Intramolecular hydrogen bonding (O–H)	Chitin	[6,9]
2	2866	▼	-CH <sub>3</sub>	Collagen	[2,3]
3	2982	▲	-CH <sub>2</sub>	Chitin	[9]
4	3045	▲	N–H	Chitin	[9]
5	3160	▲	N–H	Chitin	[9]
6	1135	▲	C–O–C	Collagen, chitin	[2,3,9]
7	1176	▲	C–O	Collagen	[5]
8	1633	▲	C=O	Collagen, chitin	[1-3,8,10]
9	1520	▲	Amide N–H	Collagen, chitin	[2,5,9]
10	1372	▼	C–H, CH <sub>3</sub>	Collagen	[1]
11	1453	▲	CH <sub>2</sub>	Chitin	[1,4,5]
12	1026	▼	C–O	Collagen, chitin	[7,9]
13	1110	▼	C=O, C–O	Chitin	[8,9]

**Table S2.** 2DCOS analysis between collagen and  $\beta$ -chitin and corresponding sequences and intensity changes.

Sequential order	Wavenumber (cm <sup>-1</sup> )	Intensity change	Assignment	Origin	Reference
1	1280	▲	C–N, N–H	Collagen, chitin	[1-5,8,9]
2	3310	▲	N–H	Collagen	[2,6]
3	3176	▲	N–H	Chitin	[11,9]
4	1452	▲	Amide N–H	Collagen, chitin	[2,5,8,9]
5	1013	▼	C=O	Chitin	[8]
6	3050	▲	N–H	Chitin	[9,11]
7	3442	▼	OH	Collagen, chitin	[2,6,9]
8	2980	▲	CH <sub>3</sub>	Chitin	[9]
9	1178	▲	C–O	Collagen	[5]
10	1110	▼	C=O, C–O	Chitin	[8,9]
11	1522	▲	N–H	Collagen, chitin	[2,5,9]
12	1309	▼	CH <sub>2</sub>	Collagen	[7]
13	1370	▼	C–H, CH <sub>3</sub>	Collagen	[2]
14	2860	▼	-CH <sub>3</sub>	Collagen	[3]
15	1564	▼	Amide N–H	Collagen, chitin	[5,9]
16	1634	▲	Amide C=O	Collagen, chitin	[2,5,8]

**Table S3.** The results of differential scanning calorimetry (DSC) analysis.

Specimen	Temperature (°C)			$\Delta H$ (J·g <sup>-1</sup> )	State	Reference
	Onset	Peak	End			
Collagen	37	104	179	369.59	Dehydration	
$\alpha$ -Chitin	39	88	184	230.45	Dehydration	
$\beta$ -Chitin	39	91	180	265.97	Dehydration	
Col	37	91	143	184.54	Dehydration	
	205	235	250	20.5	Denaturation	[12,13]
Col/ $\alpha$ -Chi	37	82	139	215.02	Dehydration	
	215	235	257	27.34	Denaturation	
Col/ $\beta$ -Chi	37	85	133	142.03	Dehydration	
	211	237	255	39.77	Denaturation	

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