Electronic Supplementary Information (ESI)

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To characterize the mica surfaces treated with silanes containing NH_2 or CH_3 end group, contact angle was taken with a commercial goniometer (DIGIDROP by GBX, France). As expected, the water contact angle changed to 108.2 ± 0.5 degree after the mica surface was coated with OTS, comparable to the value reported in the reference.[S1] The water contact angle of APTES modified mica surface became 77.6 ± 0.3 degree.

Mica before and after functionalization with OTS and APTES were also analyzed by using XPS. The XPS spectra were recorded on a Kratos Axis Ultra Xray photoelectron spectrometer (Kratos Analytical, Ltd.) utilizing a monochromated Al Kα X-ray source to determine the C, O, N, K and Si atoms presented on the surfaces. Figure S1 and Table S1 (ESI) show the XPS spectra and the atomic compositions of APTES-mica, OTS-mica and bare mica surface, respectively. We observed presence of nitrogen in all three samples, which is not expected in the composition of bare mica and is likely due to surface contamination. The concentration of nitrogen increased remarkably after the modification with APTES, indicating the presence of NH_2 group and hence APTES on the mica surface, which is in good agreement with the previous report. [S2] Similarly, after modification with OTS, the atomic concentration of carbon significantly increased, suggesting that the surface has been covered by CH_3 group.

Reference:

S1 M. Wang, K. M. Liechti, Q. Wang, and J. M. White, *Langmuir* 2005, **21**, 1848-1857.
S2 N. Crampton, W. A. Bonass, J. Kirkham, and N. H. Thomson, *Langmuir*, 2005, **21**, 7884-7891

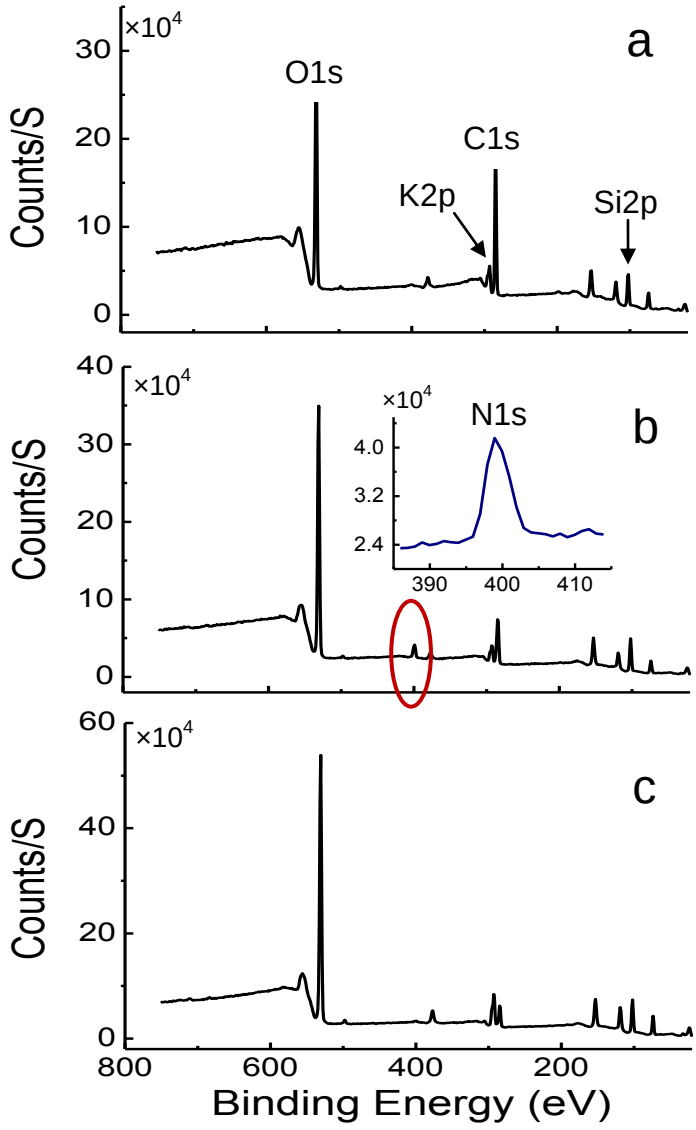

Figure S1. XPS spectra for the detection of the atomic concentration of carbon, oxygen, nitrogen, potassium and silicon of OTS-mica (a), APTES-mica (b) and bare mica (c). The inset spectrum in (b) shows an enlarged part as marked in colored circle.

	Atomic concentration (%)				
	O1s	**N 1s**	**K 2p**	**C 1s**	**Si2p**
OTS-mica	35. 2	0. 73	2. 92	49. 17	11.98
APTES-mica	51. 94	4. 6	2. 69	25. 66	17.43
Bare-mica	65. 88	0. 83	5. 22	10. 64	15.12

Table S1. XPS elemental analysis of OTS-mica, APTES-mica and bare mica.

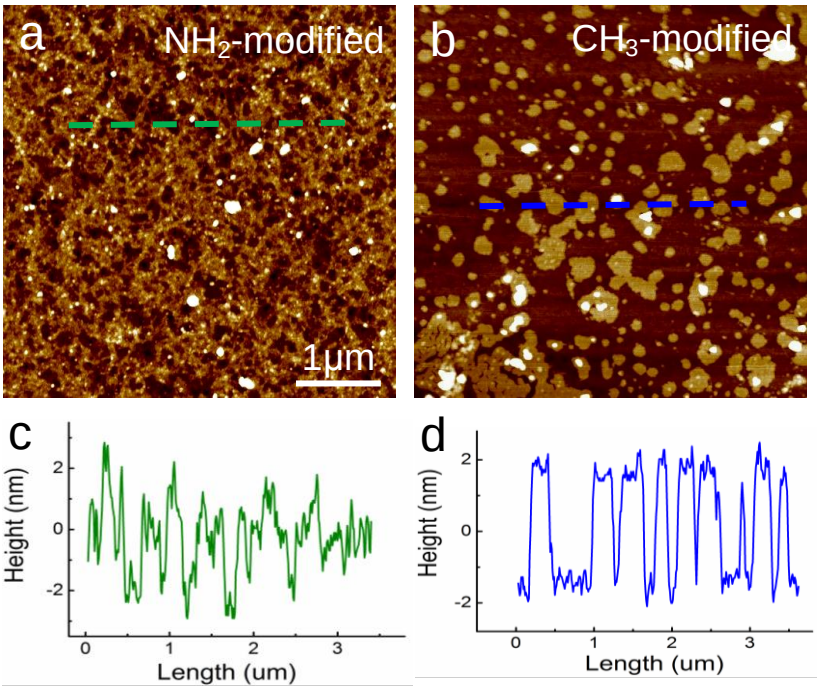

Figure S2. AFM images of insulin aggregates obtained by incubating the peptides on NH_2- and CH_3-modified mica substrates, respectively, under 90 ℃ and ~100% RH. The scale bar in (a) applies to all images. (c) and (d) are the corresponding cross section analysis of the aggregates marked with colored dotted lines in (a) and (b).

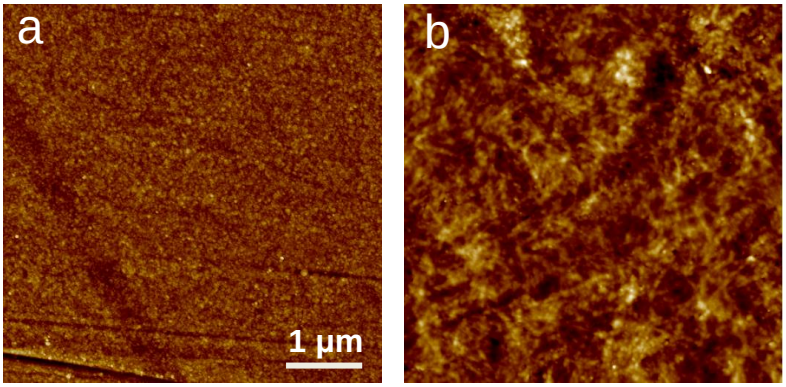

Figure S3. AFM images of glass surface (a) and insulin aggregates obtained by incubating 0.5 mg/mL peptides on glass at 90 °C and ~100% RH for 25 hours (b). No ordered fibrils were observed. The scale bar in (a) applies to both images.

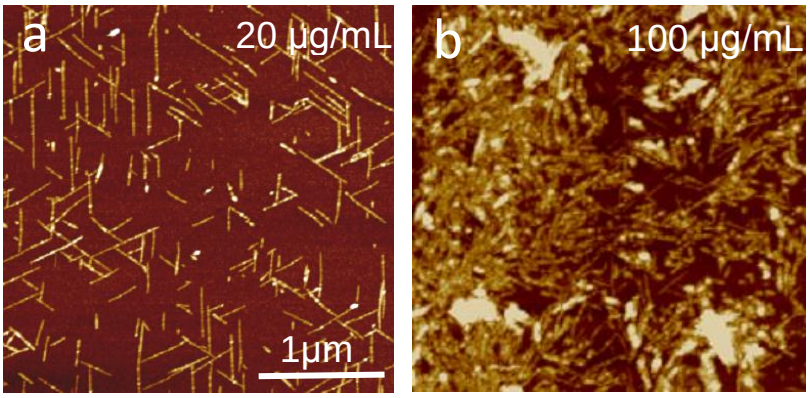

Figure S4. AFM images of insulin fibrils obtained by incubating the peptides at 70 ℃ and ~100% RH with two different concentrations. (a) 20 µg/mL and (b) 100 µg/mL. The scale bar in (a) applies to both images.