Supplementary Information

Different types of phase separation in binary monolayers of long chain alkyltrichlorosilanes on silicon oxide

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Fig. S1. AFM image $(10x10\mu m^2)$ of C_{30}/C_{16} , $R_{Sol} = 20$, LTLH at a different area of the same sample as that of Fig. 2a).



Fig. S2. AFM image $(10x10\mu m^2)$ of C_{30}/C_{18} , $R_{Sol} = 1$, HTHH at a different area of the same sample as that of Fig. 2b).



Fig. S3. AFM images $(10x10\mu m^2)$ of C_{30}/C_{16} , $R_{Sol} = 20$, LTLH of two different areas of the same sample as that of Fig. 2c).



Fig. S4. AFM images $(10x10\mu m^2)$ of two different samples exhibiting "filaments", prepared at the same conditions within the same deposition experiment : C_{30}/C_{16} , $R_{Sol} = 1$, LTLH.



Fig. S5. AFM images $(10x10\mu m^2)$ of two different samples prepared at the same conditions within the same deposition experiment : C_{30}/C_{16} , R_{Sol} =0.5, LTLH.



Fig. S6. AFM images $(10x10\mu m^2)$ of two different samples prepared at the same conditions within distinct deposition experiments : C_{30}/C_{18} , $R_{Sol} = 1$, HTLH.



Fig. S7. Phase AFM images $(10x10\mu m^2, z \text{ scale} = 15^\circ)$ corresponding to topography AFM images of Fig. 2.





Fig. S8. Phase AFM images $(10x10\mu m^2, z \text{ scale} = 15^\circ)$ corresponding to topography AFM images of Fig. 5.



Fig. S9. Phase AFM images $(10x10\mu m^2, z \text{ scale} = 15^\circ)$ corresponding to topography AFM images of Fig. 7.



Fig. S10. FTIR spectrum of binary C_{30}/C_{18} SAM, $R_{Sol} = 20$, prepared at LTHH. Intense symmetric and asymmetric stretching bands of methylene moieties are positioned at ~2850 cm⁻¹ and ~2919 cm⁻¹ respectively. Weak broad bands at ~2960 cm⁻¹ and ~2885 cm⁻¹, and the shoulder at ~2935 cm⁻¹ should be attributed to methyl terminal groups.

		C ₃₀ /C ₁₆				C ₃₀ /C ₁₈		C ₃₀ /C ₂₀			
		0.05	1	10	20	1	20	0.1	1	10	20
LTLH	$T_{ell.}$ (Å)	29.2	23.2	27.0	<mark>43.0</mark>	30.4		28.0	37.2	38.6	
	T _{AFM} (Å)	27.4	24.5	27.0	<mark>34.4</mark>	30.0		28.7	35.0	39	
LTHH	T _{ell.} (Å)						36.0		31.2		34.0
	$T_{AFM}(Å)$						36.9		29.1		35.0
HTHH	$T_{ell.}(Å)$					<mark>33.0</mark>			28.5	31.0	
	$T_{AFM}(Å)$					<mark>38.3</mark>			29.7	32.0	
HTLH	$T_{ell.}(Å)$					<mark>29.0</mark>	<mark>38.5</mark>				
	T_{AFM} (Å)					<mark>37.3</mark>	<mark>28.5</mark>				

Table SI. Thickness of the different binary SAMs, prepared at various temperature and humidity conditions: measured by ellipsometry ($T_{ell.}$), and equivalent thickness deduced from island or hole or filament coverage measured by AFM (T_{AFM}) assuming a perfect phase separation between long and short molecules. Values highlighted in yellow exhibit a high discrepancy.