

Nanodiamond Promotes Surfactant-Mediated Triglyceride Removal from a Hydrophobic Surface at or below Room Temperature

Supporting Information

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General Experimental

Tristearin lipid and *N,N*-dimethyldodecylamine *N*-oxide were purchased from Tokyo Chemical Industry (TCI) Ltd. Diamond nanopowder 97% (ND97) and other chemicals were purchased from Sigma-Aldrich. All chemicals are used as purchased without purification. Zeta potential and average size of particles in solution were measured by dynamic light scattering (DLS) on a Malvern Zetasizer using a 633 nm laser with a measuring angle of 173°. QCM-D data were recorded on a Q-Sense E4 from *Q-Sense AB*, Sweden at 15° C and 25 °C and processed using QTools software. Powder X-ray Diffraction (PXRD) was carried out on a Bruker D5000 powder X-ray diffractometer with Cu K-alpha radiation. Transmission electronic microscope (TEM) images were obtained on JEOL2100 in the Department of Physics, University of Warwick. Raman spectra were recorded on a Renishaw inVia Raman Microscope with a CCD detector and a laser of 514.5 nm. Infrared (IR) Spectra were recorded on a PerkinElmer Spectrum 100 FT-IR spectrometer, using attenuated total reflectance.

S1. Characterization of diamond nanoparticles by TEM, X-ray powder diffraction and infra-red spectroscopy.

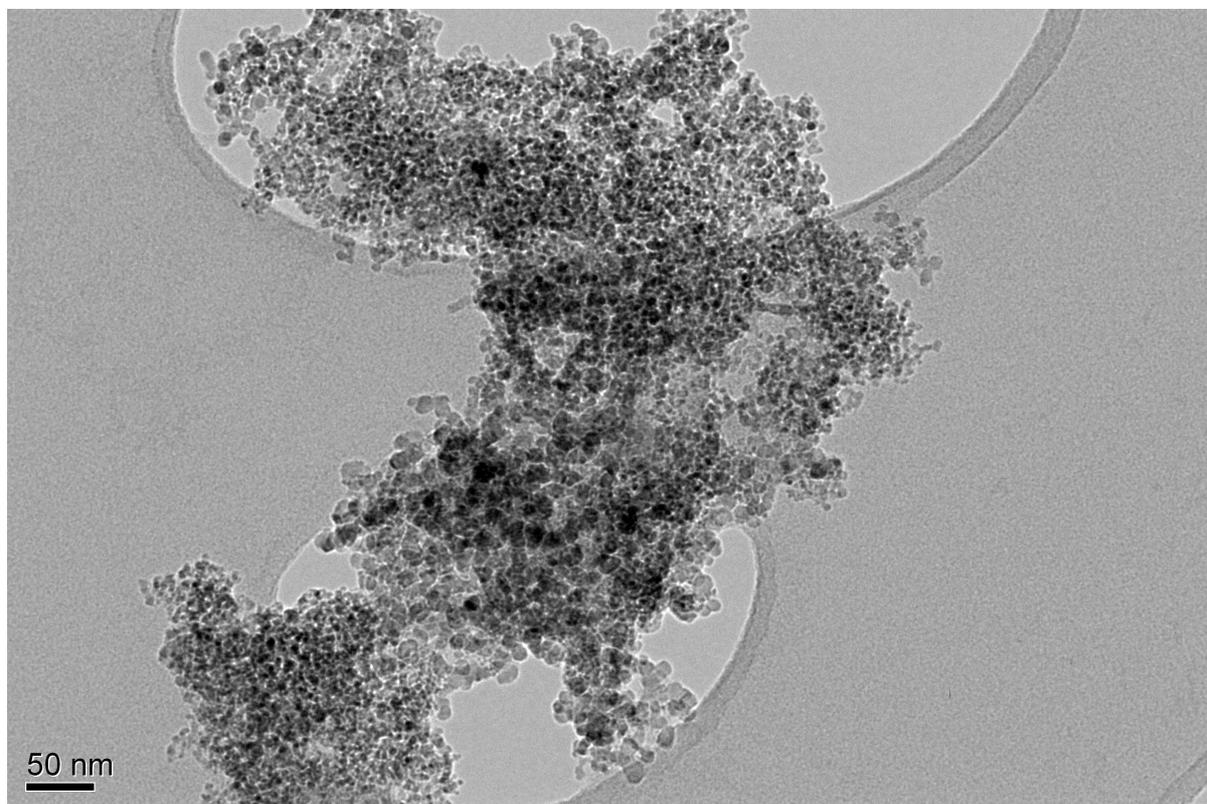


Figure S1. Raw nanodiamond ND97 showing large aggregates of *ca.* 5 nm particles.

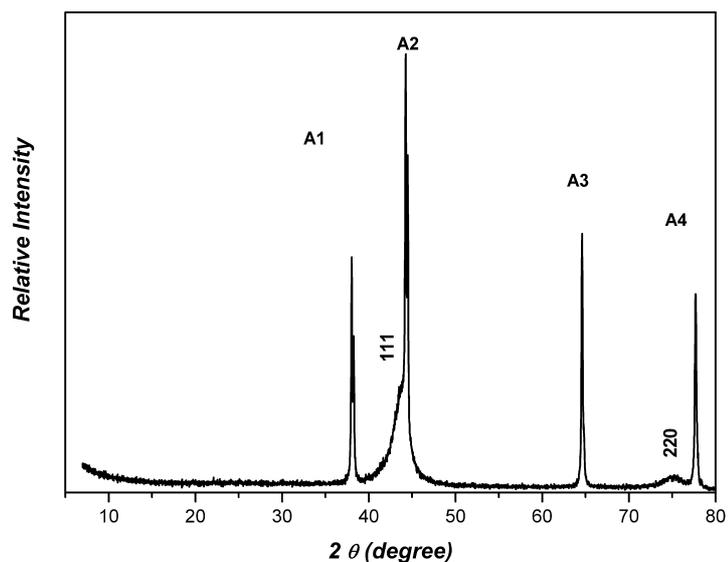


Figure S2: Pattern of X-ray powder diffraction of diamond nanoparticles (ND97). Strong and sharp peaks labelled A1-A4 belong to the aluminium holder. Weak peaks at 44° and 75° were indexed as the diffraction peaks of (111), (220) planes of cubic diamond respectively.

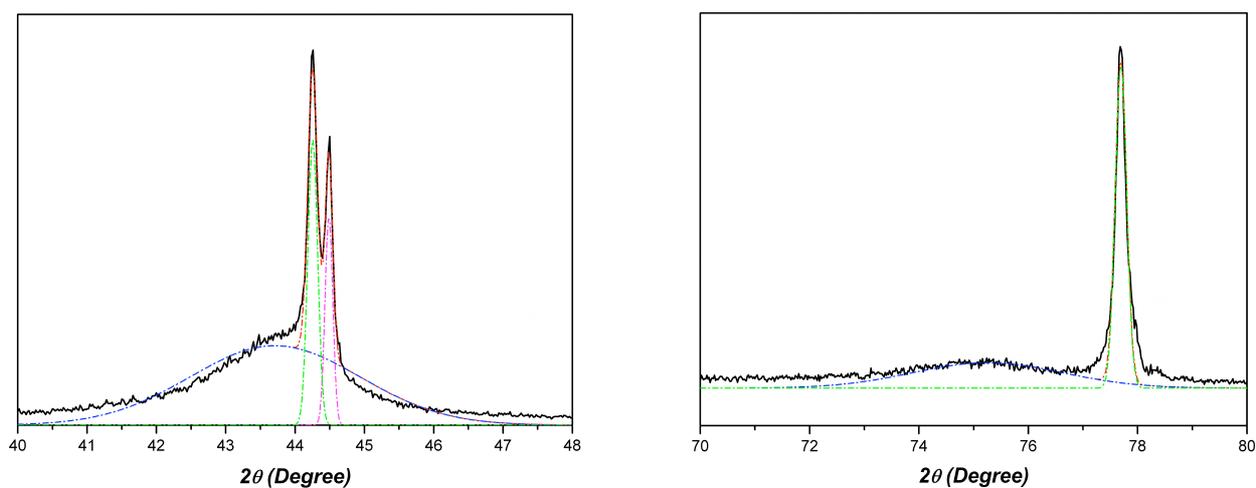


Figure S2A. Gaussian fitting of peaks at around 44° and 75° .

Peak + assignment	<i>Al</i> A1	<i>Al</i> A2	<i>Al</i> A3	<i>Al</i> A4	<i>ND97</i> (111)	<i>ND97</i> (220)
Position (2θ , °)	38.03	44.4	64.6	77.67	43.7	75.2
FWHM (b_0 , °)	0.14	0.14	0.15	0.2	2.4	2.7
FWHM (b_0 , radians)	2.44×10^{-3}	2.44×10^{-3}	2.62×10^{-3}	3.49×10^{-3}	4.19×10^{-2}	4.71×10^{-2}

Table S1. The position and full width at half maximum (FWHM) of peaks. *Al* = peaks assigned to the aluminium sample holder, *ND97* = peaks assigned to diamond nanoparticles. The position and FWHM was measured manually or given by Gaussian fitting using Origin[®] (**Figure S1A**).

The mean size (diameter) τ of nanoparticles can be estimated from X-ray diffraction using the Scherrer equation (Equation S4).

$$\tau = \frac{K\lambda}{\beta \cos \theta} = \frac{K\lambda}{(b - b_0) \cos \theta} \quad \text{Equation S1}$$

Where K represents the shape factor, typically taken to be 0.9, β represents the line broadening at half the maximum intensity (radians), θ represents the Bragg angle, λ the X-ray wavelength (Å), b and b_0 represent the FWHM for the nanoparticles and the standard crystal respectively. The wavelength of $K\alpha_1$ X-ray radiation used is 1.5406 Å. The value of b_0 at 43.7° and 75.2° was calculated as 0.14° and 0.2° respectively, using the highly crystalline aluminium holder as the standard. Hence, the mean size of diamond nanoparticles was calculated to be *ca.* 4 nm as follows:

$$\tau_{(111)} = \frac{0.9\lambda}{(b - b_0) \cos \theta} = \frac{0.9 \times 1.5406 \text{ \AA}}{(4.19 \times 10^{-2} - 2.44 \times 10^{-3}) \cos 21.85^\circ} = 37.8 \text{ \AA} \quad \text{Equation S2}$$

$$\tau_{(220)} = \frac{0.9\lambda}{(b - b_0) \cos \theta} = \frac{0.9 \times 1.5406 \text{ \AA}}{(4.71 \times 10^{-2} - 2.62 \times 10^{-3}) \cos 21.85^\circ} = 33.6 \text{ \AA} \quad \text{Equation S3}$$

Nanoparticles could be larger than predicted by the Scherrer equation because crystal size is not the only factor responsible for broadening of the diffraction peaks and this is usually viewed as an estimate only.

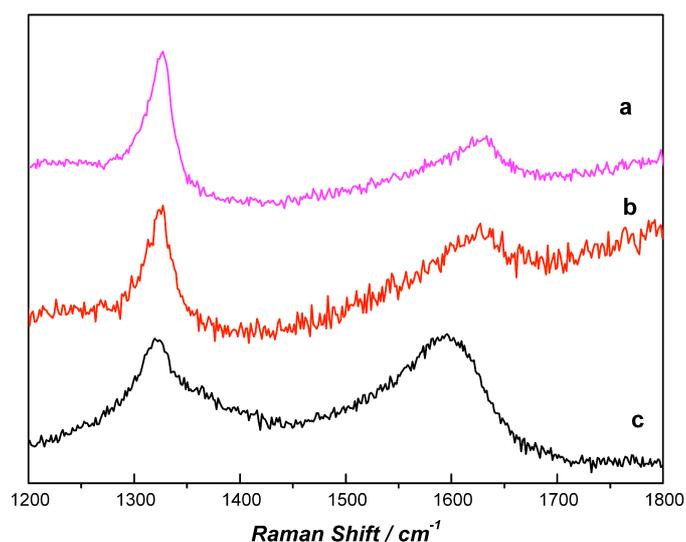


Figure S3. Raman spectra of nanodiamond samples: (a) products obtained by heating ND97 in air at 485 °C for 5 hours; (b) products obtained by heating ND97 in the air at 485 °C for 3 hours; and (c) reference spectrum of untreated ND97. The peaks at 1330 cm^{-1} and 1600 cm^{-1} can be attributed to 'sp³' and 'sp²' carbon respectively. Excitation wavelength is 514.5 nm.

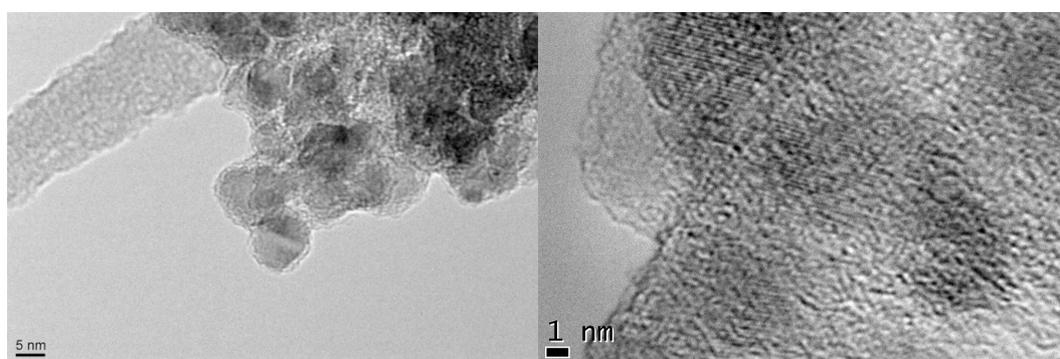


Figure S4. TEM images of diamond nanoparticles before (ND97, left) and after (1, right) thermal oxidation showing removal of paler surface associated amorphous carbon and revealing sp³ diamond-like lattice.

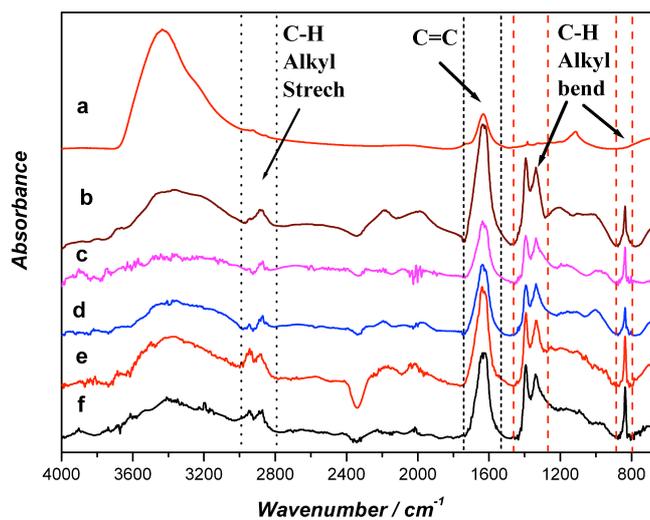
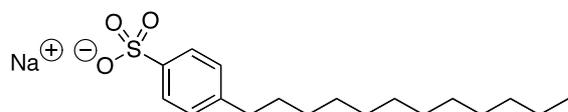
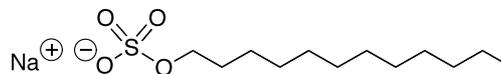


Figure S5. IR spectra of nanodiamond powder exposed to H₂ plasma for 10 minutes at different powers of microwave generator: a) reference spectrum of ND97; b) 1200 W; c) 1100 W; d) 1000 W; e) 900 W; and f) 800 W.

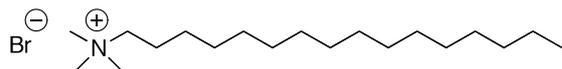
S2. Structures of surfactants used



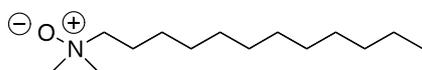
sodium dodecylbenzenesulfonate (SDBS)



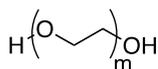
sodium dodecylsulfate (SDS)



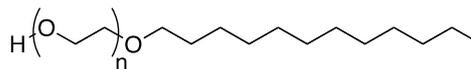
hexadecyltrimethylammonium bromide (cetyl trimethylammonium bromide, CTAB)



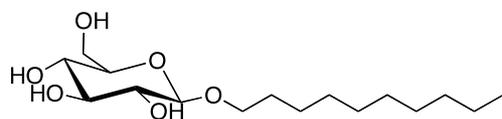
N,N-dimethyldodecylamine *N*-oxide (DDAO) or lauryl *N,N*-dimethylamine *N*-oxide (LDAO)



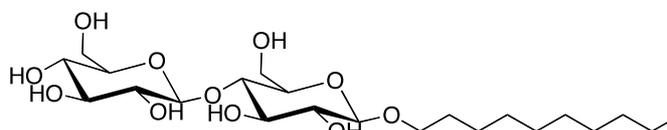
polyethylene glycol, $M_n = 1000$ (PEG-1000)



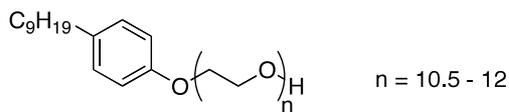
polyethylene glycol dodecyl ethyl (Brij®35)



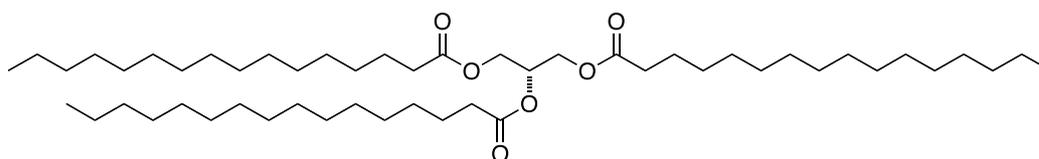
decyl β -D-glucopyranoside (G_1C_{10})



decyl β -D-maltopyranoside (G_2C_{10})



polyoxyethylene(12) nonylphenylether, branched (NFE_{10})



lipid: tristearin

Figure S6. Structures of surfactants and lipid used.

S3. Nanodiamond and colloid characterisation

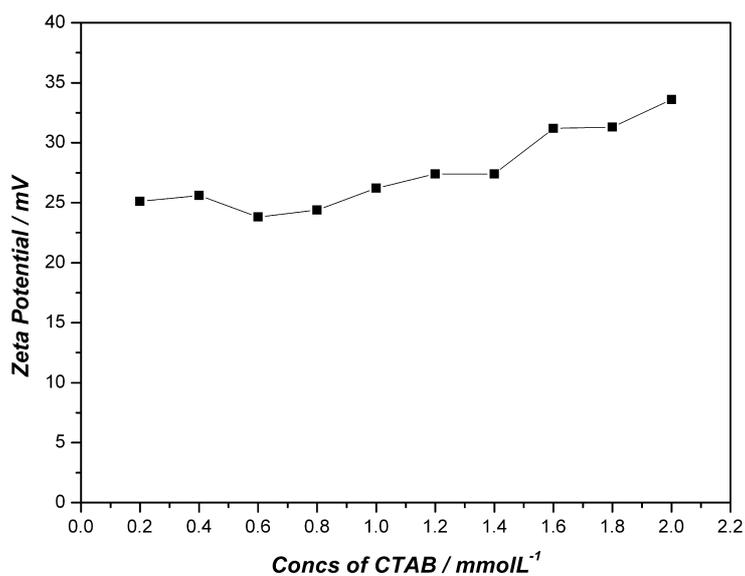


Figure S7. Correlation between the zeta potential of diamond ND97 nanoparticles and concentration of the surfactant CTAB. [ND97] = 0.1 g/L, T = 25 °C.

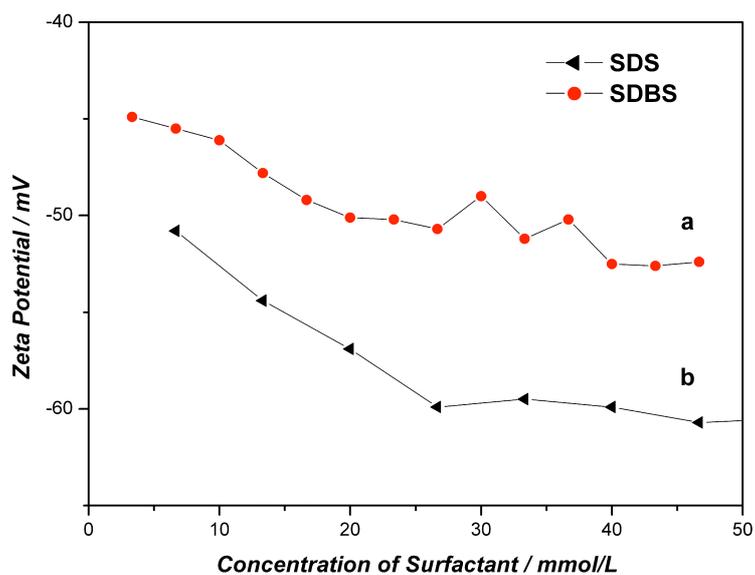


Figure S8. Correlation between the zeta potential of diamond ND97 nanoparticles and concentration of surfactant: (a) SDS, and (b) SDBS. [ND97] = 0.1 g/L, T = 25 °C.

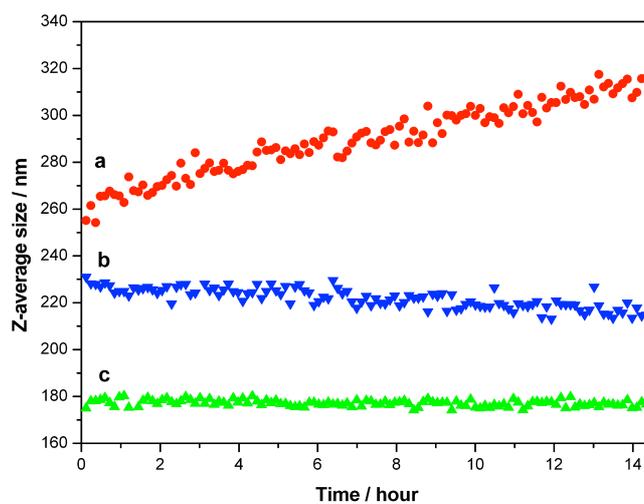


Figure S9. Average size of nanoparticle aggregates in solution over time measured by DLS: (a) ND97 in 40 mmol/L SDBS aqueous solution, (b) ND97 in 10 mmol/L SDBS aqueous solution, and (c) ND97 in 2 mmol/L CTAB aqueous solution. [ND97] = 0.1 g/L, T = 25 °C.

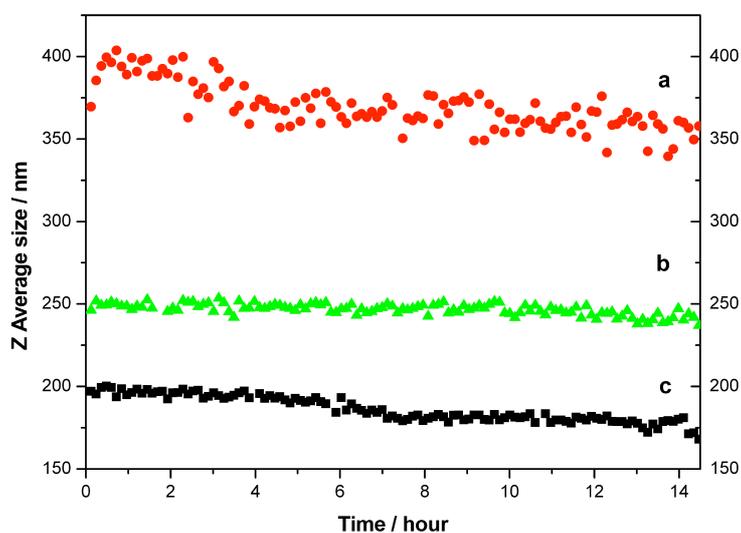


Figure S10. Curves of the average size against time of ND97 in solutions of non-ionic surfactants: (a) Brij[®] 35, (b) PEG-1000, (c) DDAO. [ND97] = 0.1 g/L, [Brij[®] 35] = [PEG-1000] = 8 g/L, [DDAO] = 40 mmol/L; T = 25 °C.

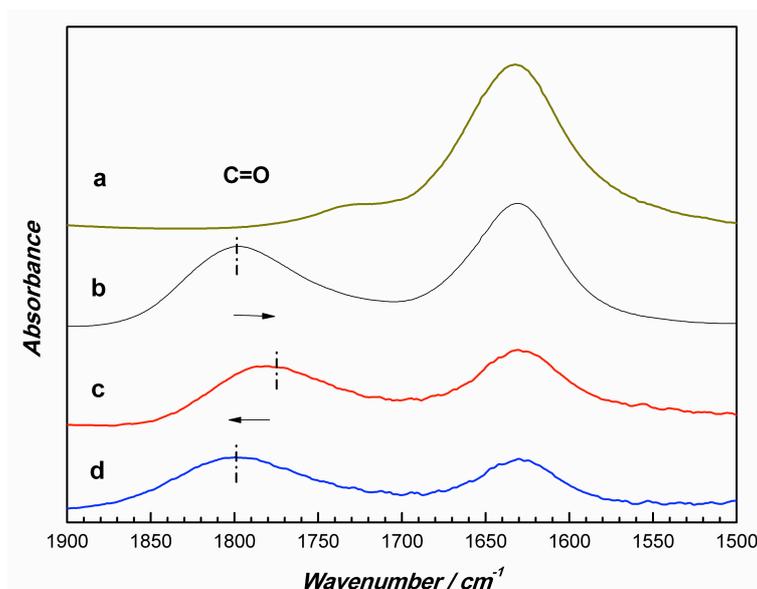


Figure S11. IR spectra of nanodiamond samples: (a) untreated ND97; (b) after heating in air at 485 °C for 3 hours; and (c) after a base treatment with KOH solution, and (d) after an acid treatment with HCl solution. Oxidized ND97 obtained by a thermal treatment was immersed in a 3 M KOH aqueous solution, and then moved into a 3 M HCl aqueous solution.

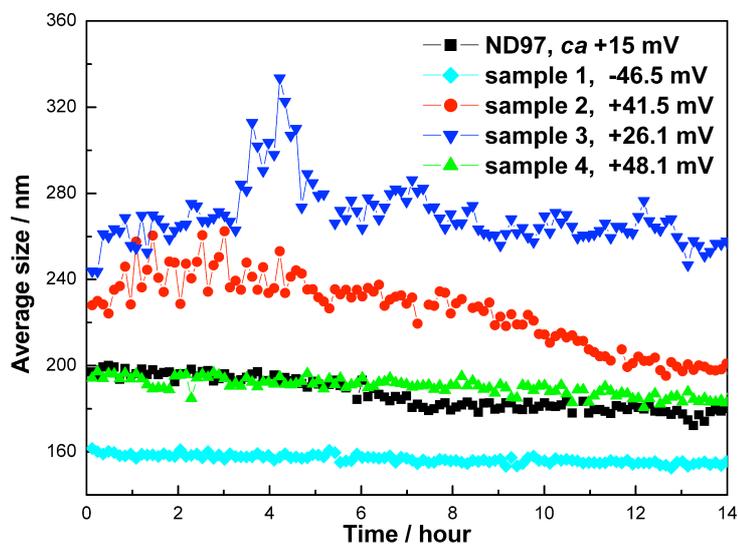


Figure S12. Curves of Z-average size of functionalised diamond nanoparticles in water over time. $[ND97] = [1] = [2] = [3] = [4] = 0.1 \text{ g/L}$.

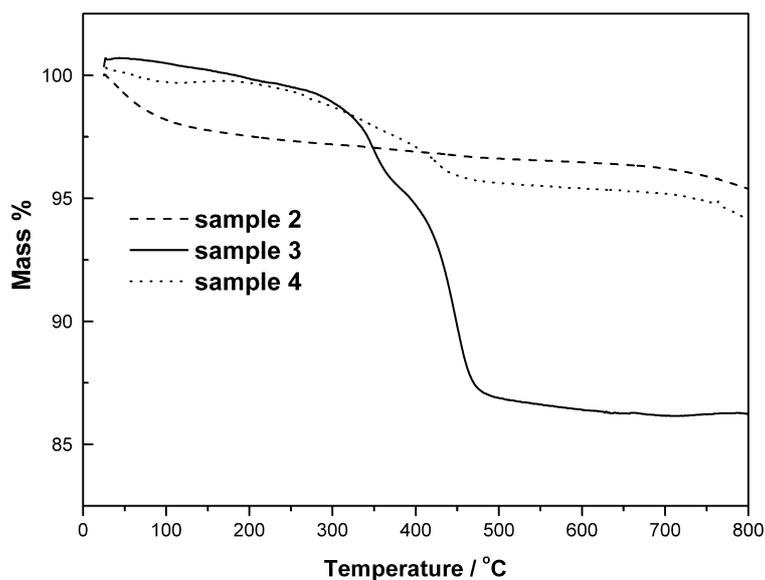


Figure S13. Thermogravimetric curves of starting materials **2**, alkylamine functionalised diamond nanoparticles **4**; and alkylcarboxyl functionalised diamond nanoparticles **3**.

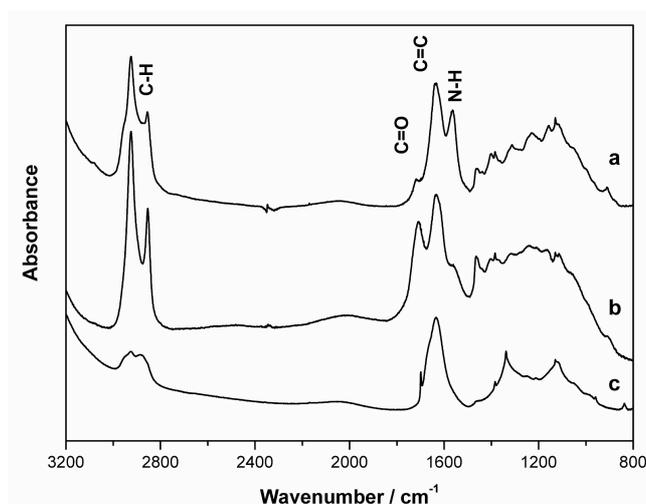


Figure S14. IR spectra of functionalised diamond nanoparticles: a) ω -alkyl-*N*-methylpiperazine amide functionalised diamond nanoparticles **4**; b) ω -alkylcarboxyl functionalised diamond nanoparticles **3**, and c) reference spectrum of starting material **2**.

S4. Methods for Quartz Crystal Microbalance preparation and measurements

Quartz crystal microbalance (QCM) and the proprietary monitoring of signal dissipation (QCM-D) are based on the oscillation of a thin piezoelectric quartz crystal induced by an oscillating electric field. The change in mass of the crystal induces a proportional change in the frequency of the oscillation. If the adsorbed film is thin, rigid and evenly distributed, the additional mass can be calculated from the changes in frequency by the Sauerbrey equation (Equation S4):

$$\Delta m = -k \times \frac{\Delta f}{n} \quad \text{Equation S4}$$

Where Δm is the adsorbed mass, Δf represents the change in frequency, k is a crystal-dependent constant, and n is the overtone number. The oscillation frequency of a crystal in contact with a liquid, as is the case here, may not obey the Sauerbrey equation.^{1,2} In addition to the frequency, QCM-D can also simultaneously measure the dissipation,^{1,3} allowing us to analyse the viscoelastic properties of the film, which is useful for interpretation of measurements of our non-rigid lipid films.³⁻⁵ However, interpretation is sensitive to the modelling parameters, and hence all data presented herein are without modification. Frequency and dissipation signals were monitored for six overtone frequencies (1st, 3rd, 5th, 7th, 9th and 11th) but data are presented only for the 3rd overtone as the other signals exhibited similar behaviour.

QCM-D experimental: To remove air in the reaction chamber inside the cell, pure water was pumped into the cell until no bubbles could be observed in the tubes. In the first few minutes, water was used to obtain a stable baseline, and then swapped to buffers (surfactant solutions or nanodiamond colloids) for lipid removal. After reaching equilibrium, buffers were switched to surfactant solution for 15 minutes and then to water for another 15 minutes for the purpose of cleaning. The rate of flow was maintained at 200 $\mu\text{L}/\text{min}$ and all experiments were carried out at a cell temperature of 25 $^{\circ}\text{C}$ or 15 $^{\circ}\text{C}$.

S5. QCM-D data for lipid removal by different surfactant with or without nanoparticles at 25 °C (frequency and dissipation change to 3rd overtone).

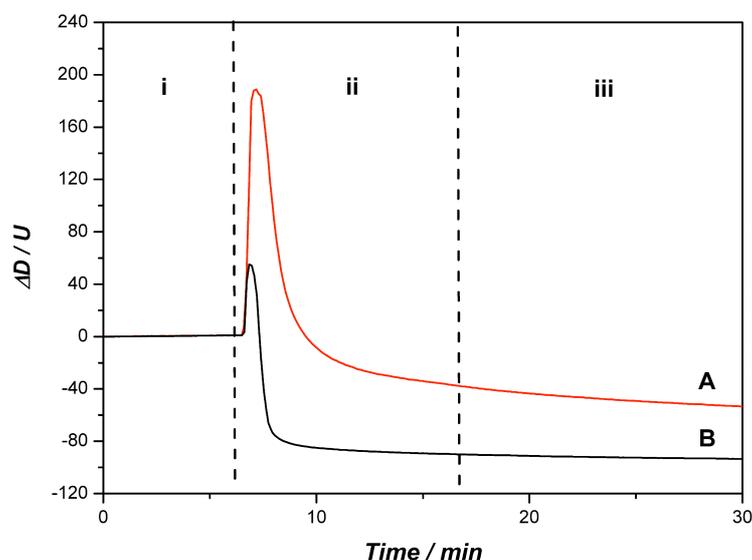


Figure S15. Dissipation changes to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water is used for both sensors to obtain a stable baseline; ii) 7-17 minutes, SDBS/ND97 colloid and SDBS solution were used for sensor A and B respectively, to remove tristearin lipid; iii) 17-30 minutes SDBS solution was used for both sensors, to clean the QCM system. [SDBS] = 40 mmol/L, [ND97] = 0.1 g/L.

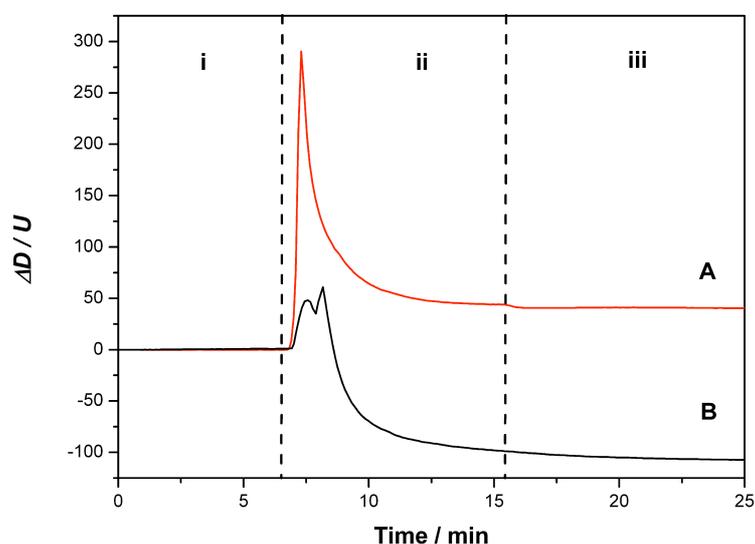


Figure S16. Dissipation change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water is used for both sensors to obtain a stable and flat baseline; ii) 7-16 minutes, G₁C₁₀/ND97 colloid and G₁C₁₀ solution were used for sensor A and B respectively, to remove tristearin lipid; iii) 16-25 minutes G₁C₁₀ solution was used for both sensors, to clean the QCM system. [G₁C₁₀] = 3.1 mmol/L, [ND97] = 0.1 g/L.

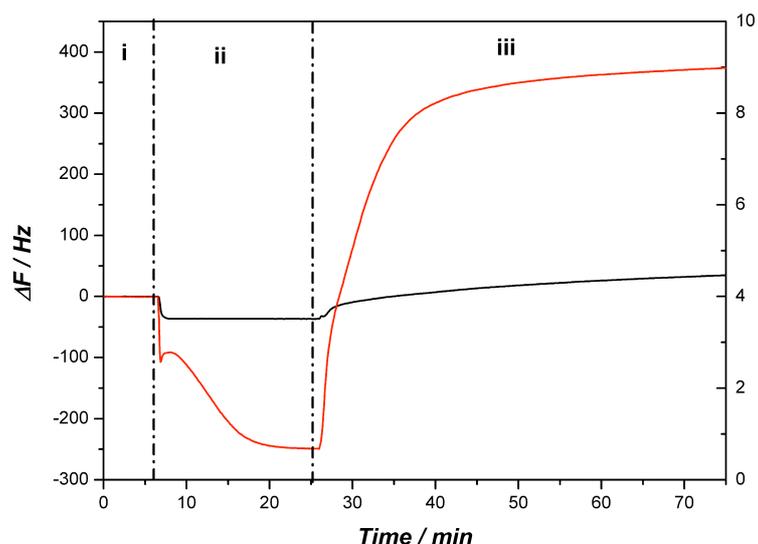


Figure S17. Frequency change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-6 minutes, water is used for both sensors to obtain a stable and flat baseline; ii) 6-25 minutes, $G_2C_{10}/ND97$ colloid and G_2C_{10} solution were used for sensor A and B respectively, to remove tristearin lipid; iii) 25-75 minutes SDBS solution was used for both sensors, to clean the QCM system. $[ND97] = 0.1 \text{ g/L}$, $[G_2C_{10}] = 2.1 \text{ mmol/L}$. $[SDBS] = 40 \text{ mmol/L}$.

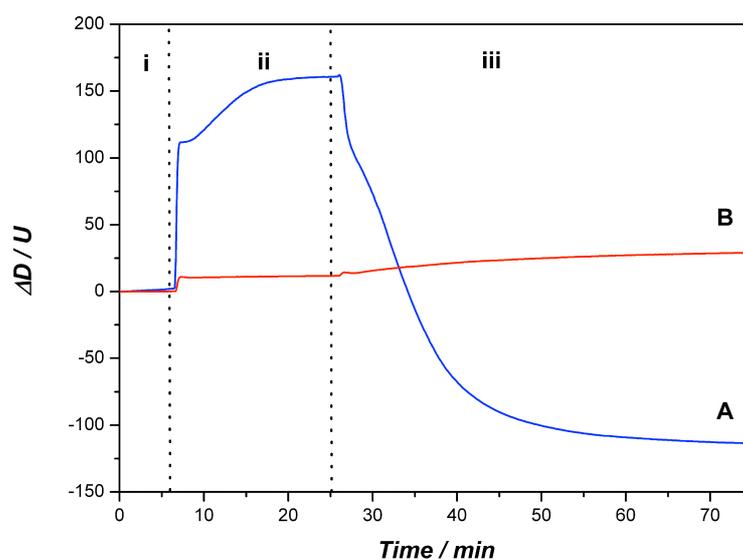


Figure S18. Dissipation change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-6 minutes, water is used for both sensors to obtain a stable baseline; ii) 6-25 minutes, $G_2C_{10}/ND97$ colloid and G_2C_{10} solution were used for sensor A and B respectively, to remove tristearin lipid; iii) 25-75 minutes SDBS solution was used for both sensors, to clean the QCM system. $[ND97] = 0.1 \text{ g/L}$, $[G_2C_{10}] = 2.1 \text{ mmol/L}$. $[SDBS] = 40 \text{ mmol/L}$.

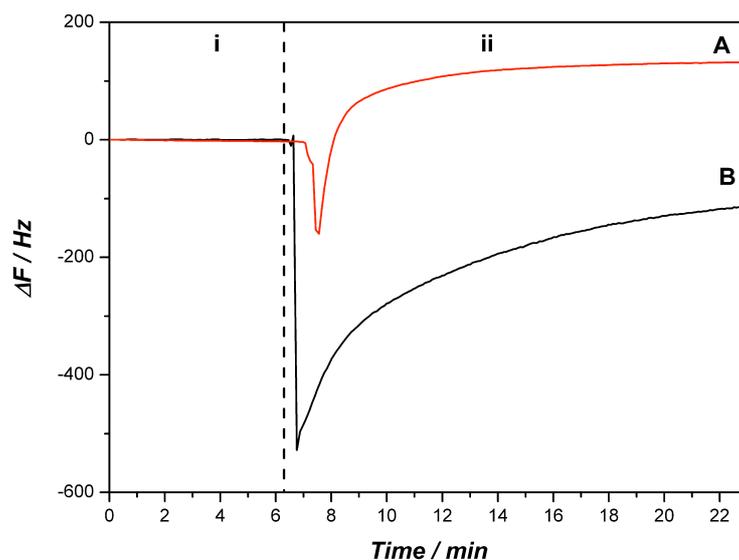


Figure S19. Frequency change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-6 minutes, water is used for both sensors to obtain a stable baseline; ii) 6-23 minutes, DDAO/ND97 colloid and DDAO solution were used for sensor A and B respectively, to remove tristearin lipid. [ND97] = 0.1 g/L, [DDA0] = 40 mmol/L.

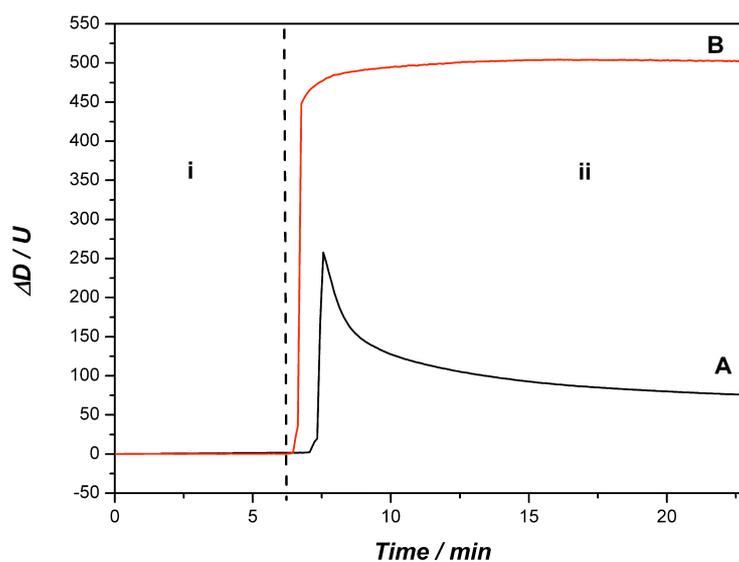


Figure S20. Dissipation change to 3rd overtone when the tristearin coated sensor exposure to buffers: i) 0-6 minutes, water is used for both sensors to obtain a stable baseline; ii) 6-23 minutes, DDAO/ND97 colloid and DDAO solution were used for sensor A and B respectively, to remove tristearin lipid. [ND97] = 0.1 g/L, [DDA0] = 40 mmol/L.

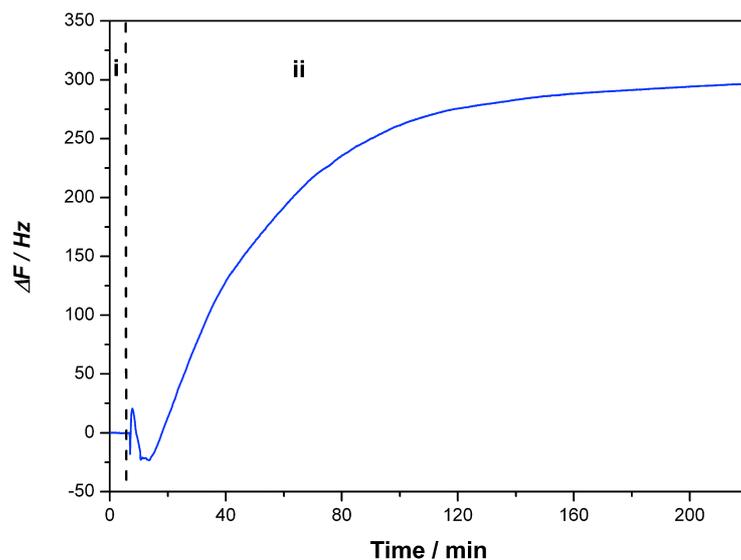


Figure S21. Frequency change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water was used to obtain a stable baseline; ii) 7-220 minutes, NFE₁₀ non-ionic surfactant was used for removal of tristearin lipid. [NFE₁₀] = 0.15 wt.%.

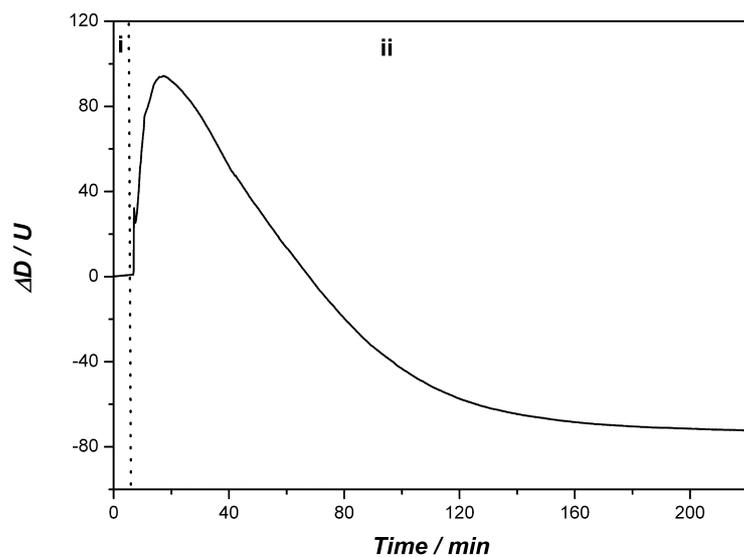


Figure S22. Dissipation change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water was used to obtain a stable baseline; ii) 7-220 minutes, NFE₁₀ non-ionic surfactant was used for removal of tristearin lipid. [NFE₁₀] = 0.15 wt.%.

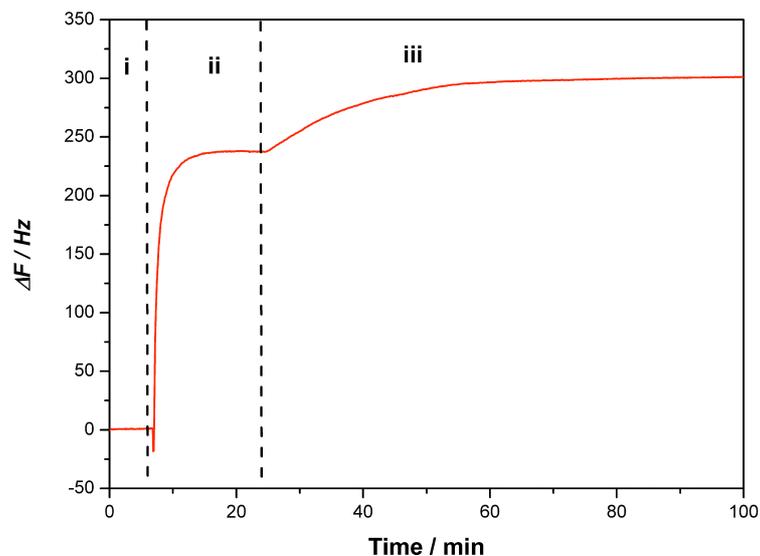


Figure S23. Frequency change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water was used to obtain a stable baseline; ii) 7-25 minutes, ND97/NFE₁₀ colloid was used for removal of tristearin lipid; and iii) 25-100 minutes, NFE₁₀ solution was used for the purpose of cleaning. [NFE₁₀] = 0.15 wt.%, [ND97] = 0.1 g/L.

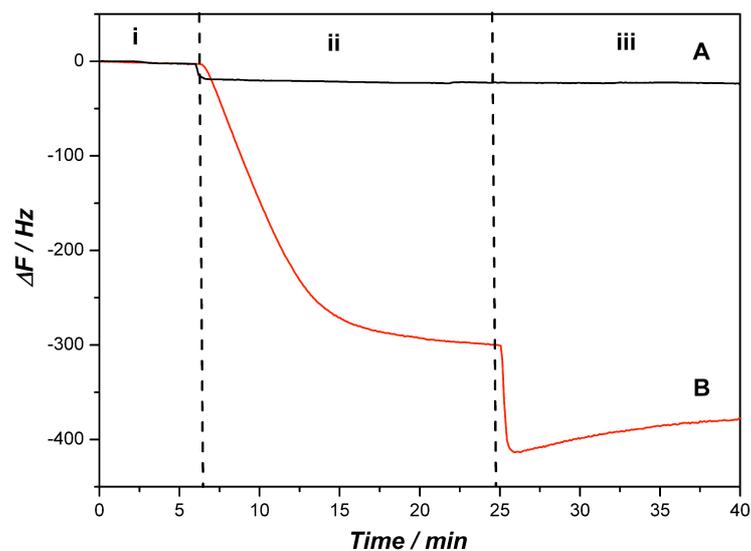


Figure S24. Frequency change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-6 minutes, water was used to obtain a stable baseline; ii) 6-25 minutes, ND97 suspension was used to deposit nanoparticles on the lipid layer for sensor B, and Brij[®]35 solution was used for lipid removal for sensor A; and iii) 25-40 minutes, Brij[®]35 solution was used for both sensors. [Brij[®]35] = 8 g/L, [ND97] = 0.1 g/L.

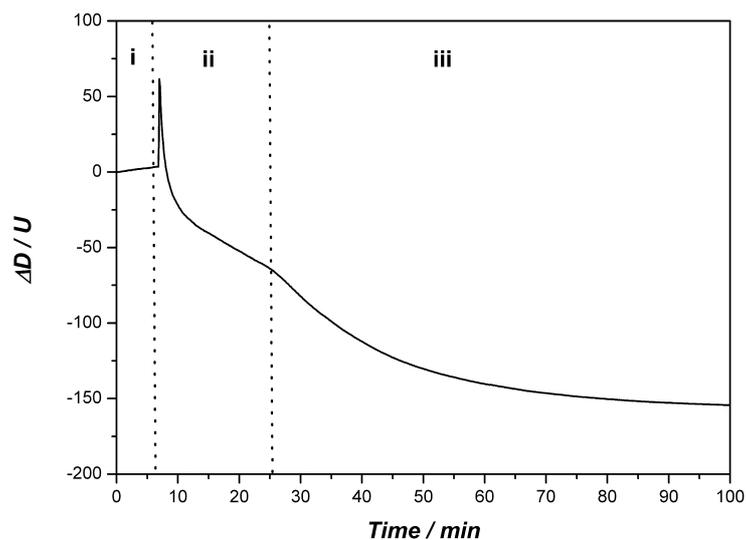


Figure S25. Dissipation change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water was used to obtain a stable baseline; ii) 7-25 minutes, ND97/NFE₁₀ colloid was used for removal of tristearin lipid; and iii) 25-100 minutes, NFE₁₀ solution was used for the purpose of cleaning. [NFE₁₀] = 0.15 wt.%, [ND97] = 0.1 g/L.

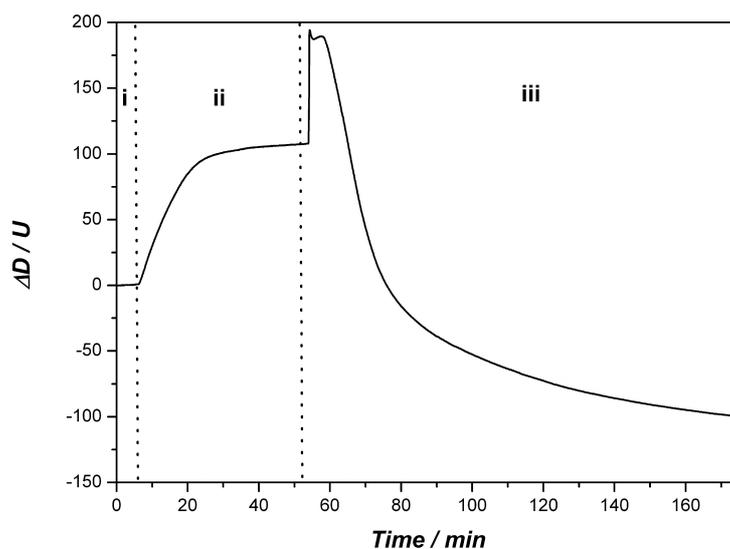


Figure S26. Dissipation change to 3rd overtone when the tristearin coated sensor exposed to: i) 0-7 minutes, water is used to obtain a stable baseline; ii) 7-52 minutes, ND97 suspension is used to deposit particles on lipid; and iii) 52-175 minutes SDBS solution was used for lipid removal. [SDBS] = 40 mmol/L, [ND97] = 0.1 g/L.

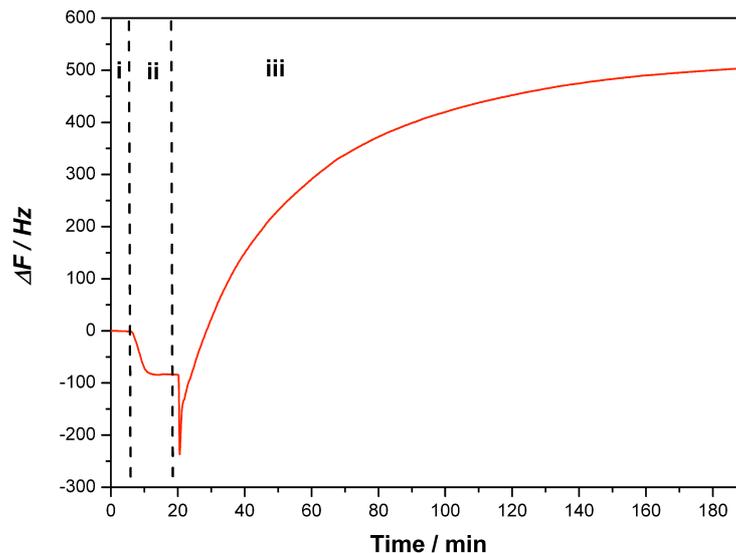


Figure S27. Frequency change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water is used to obtain a stable baseline; ii) 7-18 minutes, suspension of nanoparticle **2** is used to deposit particles on lipid; iii) 18-190 minutes SDBS solution was used for lipid removal. [SDBS] = 40 mmol/L, [**2**] = 0.1 g/L.

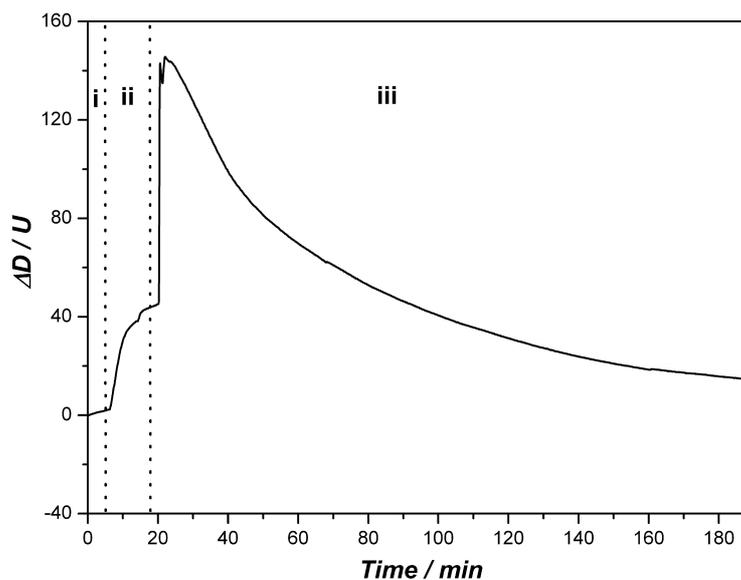


Figure S28. Dissipation change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water is used to obtain a stable baseline; ii) 7-18 minutes, suspension of nanoparticle **2** is used to deposit particles on lipid; iii) 18-190 minutes SDBS solution was used for lipid removal. [SDBS] = 40 mmol/L, [**2**] = 0.1 g/L.

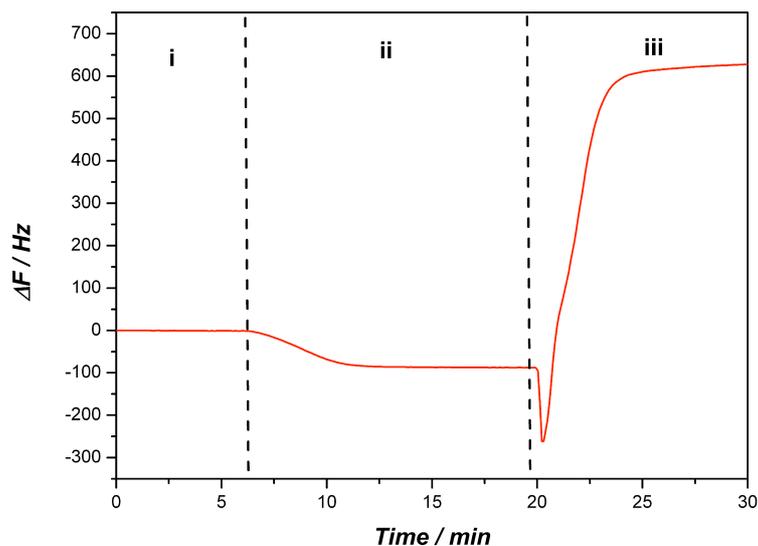


Figure S29. Frequency change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water is used to obtain a stable baseline; ii) 7-20 minutes, suspension of nanoparticle **2** is used to deposit particles on lipid; iii) 20-30 minutes G_1C_{10} solution was used for lipid removal. $[G_1C_{10}] = 3.1 \text{ mmol/L}$, $[2] = 0.1 \text{ g/L}$.

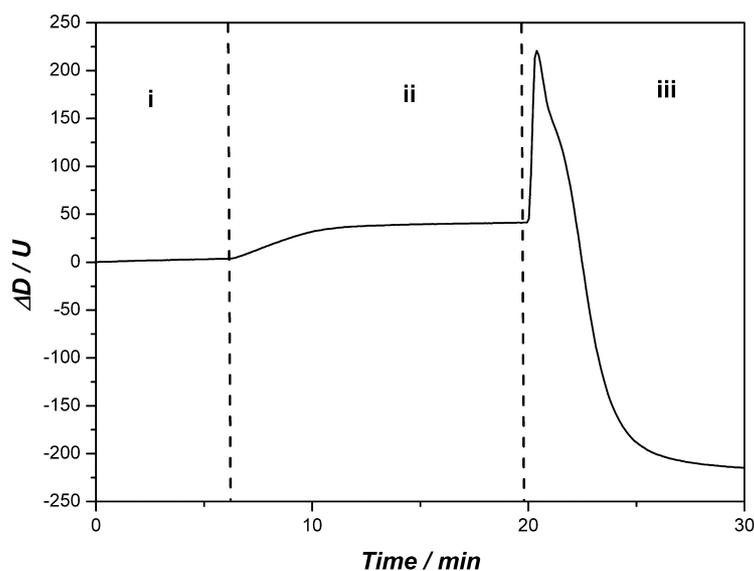


Figure S30. Dissipation change to 3rd overtone when the tristearin coated sensor was exposed to: i) 0-7 minutes, water is used to obtain a stable baseline; ii) 7-20 minutes, suspension of nanoparticle **2** is used to deposit particles on lipid; iii) 20-30 minutes G_1C_{10} solution was used for lipid removal. $[G_1C_{10}] = 3.1 \text{ mmol/L}$, $[2] = 0.1 \text{ g/L}$.

S6. QCM-D data for lipid removal by SDBS at different concentration at 25 °C (frequency and dissipation change to 3rd overtone measured by QCM-D)

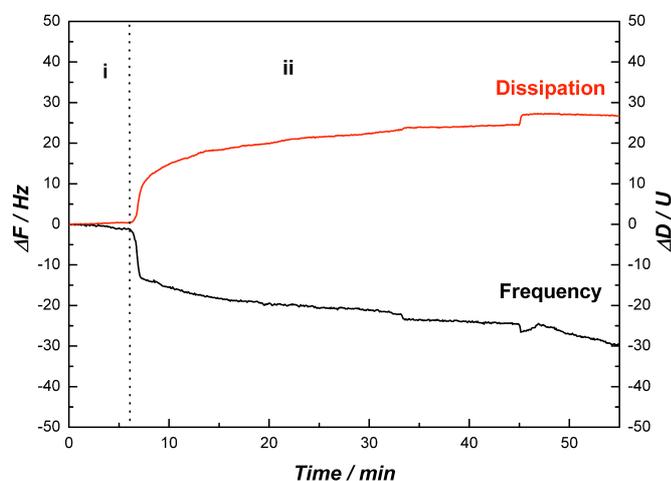


Figure S31. Frequency and dissipation changes on 3rd overtone when tristearin lipid coated sensor exposure to buffers: i) 0-6 minutes, water was used to get a stable and flat baseline; and ii) 6-55 minutes, SDBS solution was used for removing lipid. [SDBS] = 1 mmol/L, T = 25 °C. No desorption was observed at concentration below the critical micelle concentration (CMC, 1.2 mmol/L).

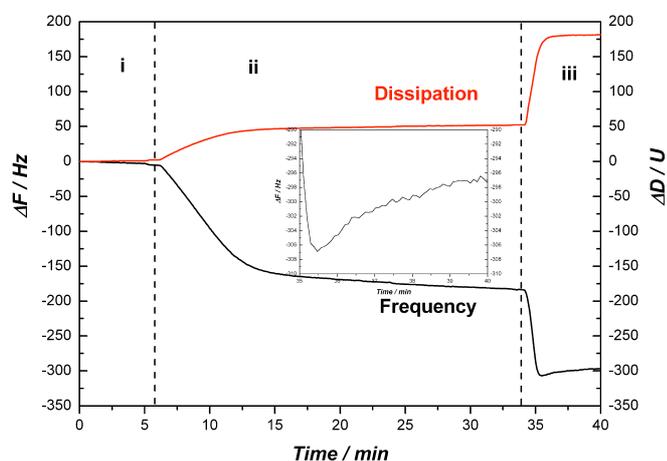


Figure S32. Frequency and dissipation changes to 3rd overtone when tristearin lipid coated sensor is exposed to: i) 0-6 minutes, water was used to get a stable and flat baseline; ii) 6-33 minutes, ND97 was used to deposit nanoparticles on lipid layer, and iii) 33-40 minutes, SDBS solution was used for removing lipid. [ND97] = 0.1 g/L, [SDBS] = 1 mmol/L, T = 25 °C. A very limited desorption was observed with a 10 Hz increase in Frequency, which was achieved after 5 minutes. There is no change observed to dissipation, since the amount of lipid removal is too small to affect the viscoelasticity property of lipid layer.

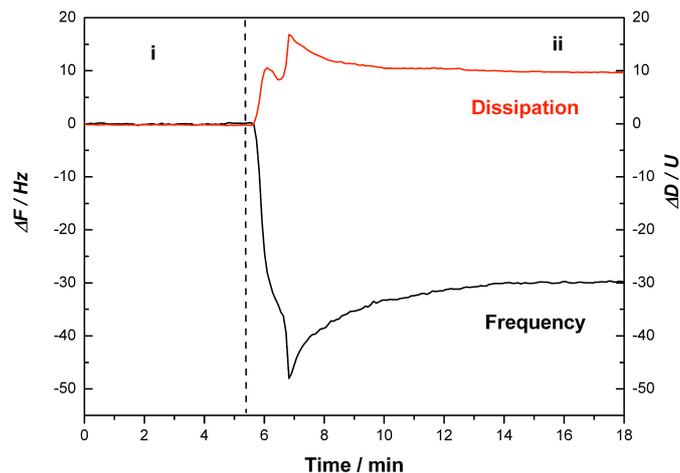


Figure S33. Frequency and dissipation changes to 3rd overtone when tristearin lipid coated sensor is exposed to: i) 0-5 minutes, water is used for a flat baseline; ii) 5-18 minutes, 20 mmol/L SDBS was used. T = 25 °C. Initial adsorption of surfactants caused a decrease in frequency and an increase in dissipation, and the following desorption resulted in an increase in frequency and a decrease in dissipation. In fact, the ‘desorption’ observed on QCM-D is the combine effect of lipid-surfactants aggregate leaving from lipid layer and the adsorption of surfactant from bulk solution. Thus, the frequency increase calculated from the lowest point *R* is attributed to the net removal of lipid. The lipid removal of *ca* 19 Hz was achieved in 7 minutes.

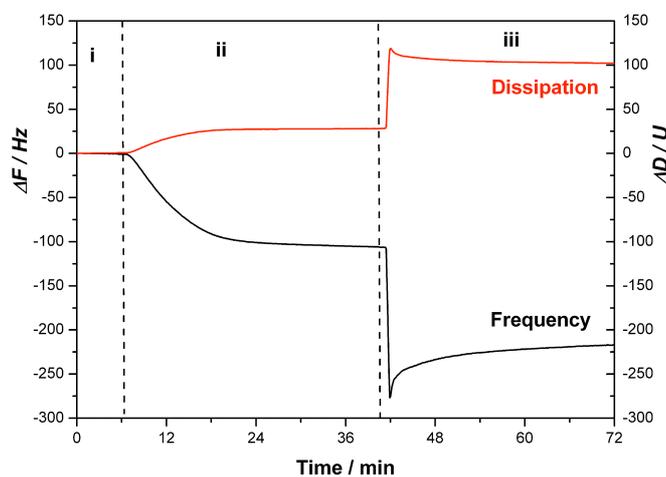


Figure S34. Frequency and dissipation changes to 3rd overtone when tristearin lipid coated sensor exposure to buffers: i) 0-5 minutes, water is used for a flat baseline; ii) 5-19 minutes, 0.1 g/L ND97 was used to deposit nanoparticles on the lipid surface; and iii) 20 mmol/L SDBS was used. T = 25 °C. The maximum desorption was observed to be 60 Hz, and it was achieved after around 20 minutes. However, the decrease in dissipation induced by desorption was relatively small, indicating that nanoparticles remained on the lipid layer at end of lipid removing process. Thus, this maximum desorption calculated from the lowest point was attributed to the total lipid removal.

S7. QCM-D data for lipid removal at 25 °C by 40 mmol/L anionic surfactant SDBS in the presence of nanoparticles with various zeta potential (frequency and dissipation change to 3rd overtone measured by QCM-D).

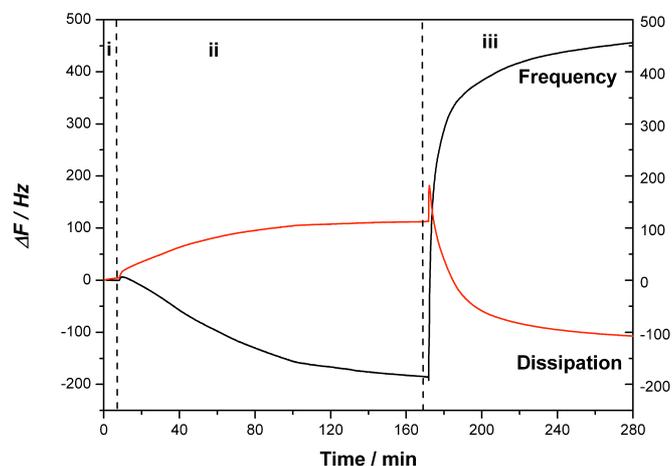


Figure S35. Frequency and dissipation changes of 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water was used for baseline; ii) 7-170 minutes, colloid of nanoparticle **3** was used to deposit nanoparticles on the surface; and iii) 170-280 minutes, SDBS solution was used for lipid removal. $[3] = 0.1 \text{ g/L}$, $[SDBS] = 40 \text{ mmol/L}$, $T = 25 \text{ }^\circ\text{C}$. In the presence of **1** nanoparticles with zeta potential of 26.1 mV, the lipid removal R of 650 Hz was achieved after 110 minutes. Note that the maximum removal possibly is over 650 Hz, as the frequency remains increasing after exposure to SDBS solution for over 110 minutes.

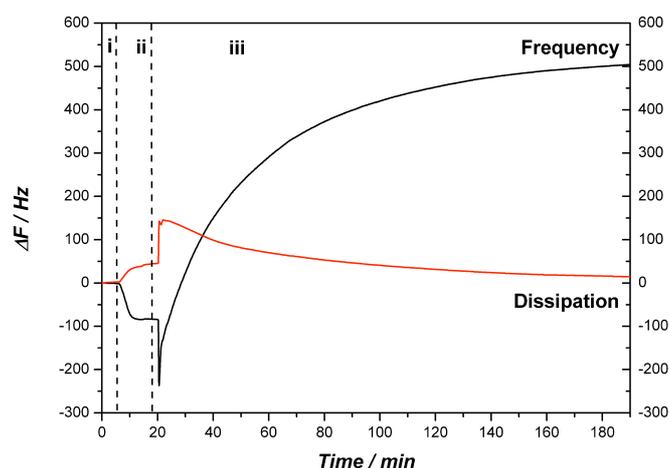


Figure S36. Frequency and dissipation changes of 3rd overtone after exposure of the tristearin lipid coated sensor to: i) 0-7 minutes, water was used for baseline; ii) 7-20 minutes, suspension of nanoparticle **2** was used to deposit nanoparticles on the lipid layer; and iii) 20-190 minutes, SDBS 40 mmol/L. In the presence of **2** nanoparticles with zeta potential of 41.5 mV, the lipid removal R was observed to 740 Hz and achieved after 170 minutes respectively. $[2] = 0.1 \text{ g/L}$, $[SDBS] = 40 \text{ mmol/L}$, $T = 25 \text{ }^\circ\text{C}$.

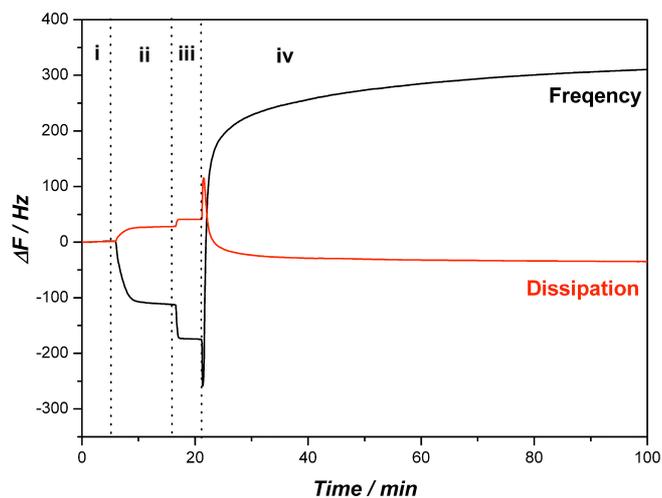


Figure S37. Frequency changes of 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water was used for baseline; ii) 7-16 minutes, suspension of nanoparticles **2** was used to deposit nanoparticles on the lipid layer; iii) 16-21 minutes, colloid of nanoparticle **1** was used to neutralize the positive charged surface of nanoparticles, and iv) 21-100 minutes, SDBS was used for cleaning. In the presence of both positively charged hydrogenated **2** and negatively charged oxidised nanoparticle **1**, the lipid removal of 570 Hz was achieved after exposure to SDBS solution for around 65 minutes. [**1**] = [**2**] = 0.1 g/L, [SDBS] = 40 mmol/L, T = 25 °C.

S8. QCM-D data for lipid removal at 25 °C by by non-ionic surfactant decyl β -D-glucopyranoside (G_1C_{10}) in the presence of nanoparticles with various zeta potential (frequency and dissipation change to 3rd overtone).

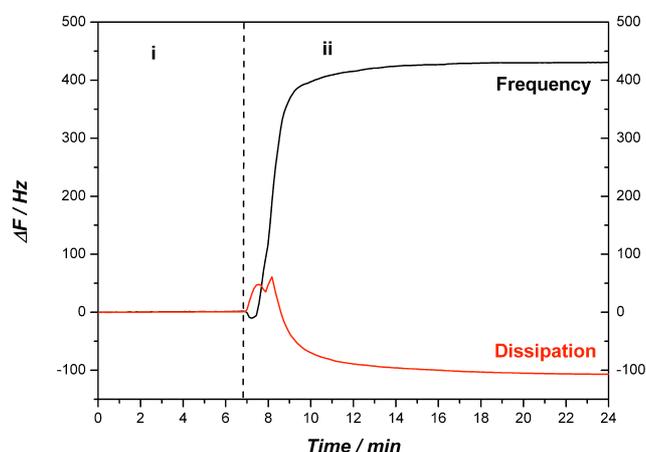


Figure S38. Frequency changes to 3rd overtone after exposure the tristearin coated sensor to: i) 0-7 minutes, water; ii) 7-24 minutes, G_1C_{10} solution 3.1 mmol/L. Without any nanoparticles, the lipid removal of 440 Hz was achieved after exposure G_1C_{10} solution for 8 minutes. $T = 25\text{ }^\circ\text{C}$.

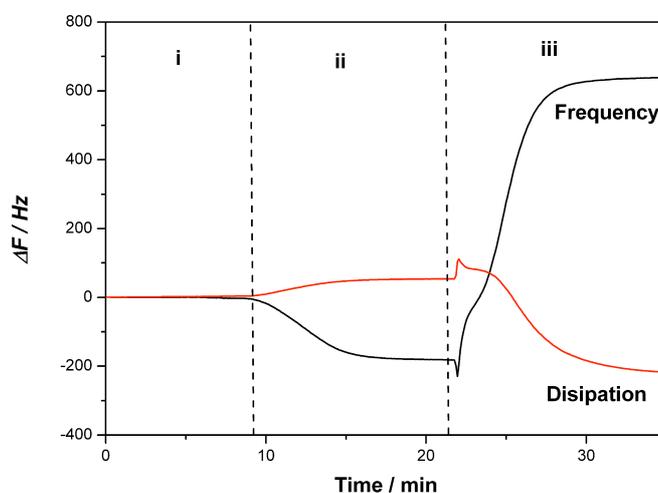


Figure S39. Frequency changes to 3rd overtone after exposure of the tristearin coated sensor to: i) 0-8 minutes, water; ii) 8-22 minutes, ND97 suspension was used to deposit nanoparticles on lipid layer; iii) 22-35 minutes, G_1C_{10} solution was used for lipid removing. $[ND97] = 0.1\text{ g/L}$, $[G_1C_{10}] = 3.1\text{ mmol/L}$, $T = 25\text{ }^\circ\text{C}$. The maximum lipid removal of 865 Hz was achieved within 8 minutes after exposure to G_1C_{10} solution, in the presence of ND97 nanoparticles with a zeta potential of *ca* 15 mV.

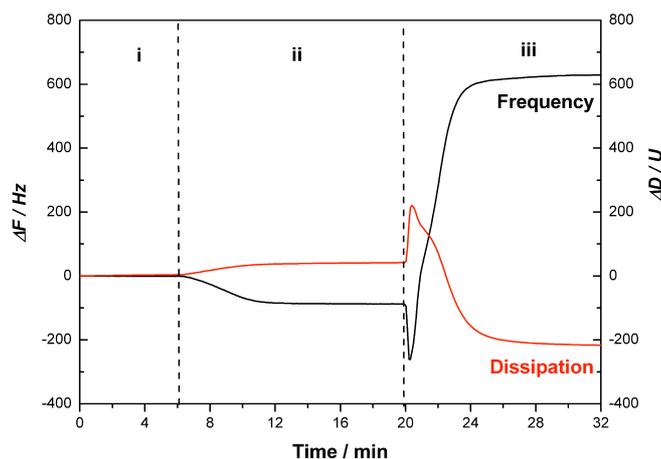


Figure S40. Frequency changes to 3rd overtone after exposure the tristearin coated sensor to buffers: i) 0-7 minutes, water was used for baseline; ii) 7-20 minutes, suspension of nanoparticle **2** was used to deposit nanoparticles on the lipid, and iii) 20-32, G₁C₁₀ solution was used for lipid removing. In the presence of nanoparticles **2** with zeta potential of 41.5 mV, the lipid removal of 900 Hz was achieved for exposure to G₁C₁₀ for 8 minutes. [2] = 0.1 g /L, [G₁C₁₀] = 3.1 mmol/L, T = 25 °C.

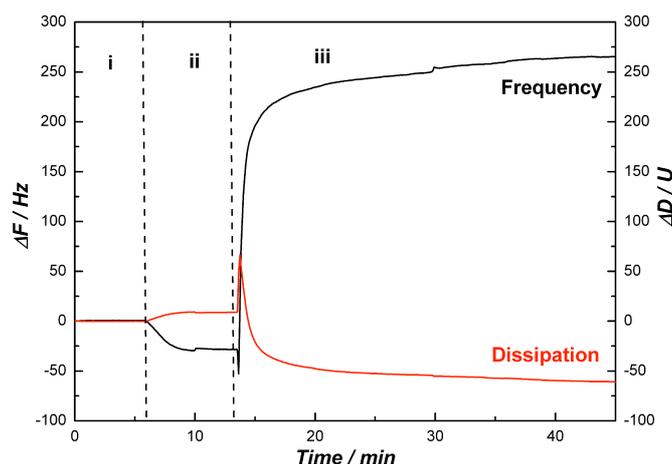


Figure S41. Frequency changes to 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water; ii) 7-13 minutes, suspension of nanoparticle **4** was used to deposit nanoparticles on lipid layer, and iii) 13-45, G₁C₁₀ solution was used for lipid removing. In the presence of nanoparticle **4** with a zeta potential of 48.1 mV, the lipid removal of 320 Hz was achieved for exposure to G₁C₁₀ for 27 minutes. [4] = 0.1 g /L, [G₁C₁₀] = 3.1 mmol/L, T = 25 °C.

S9. QCM-D data for lipid removal at 25 °C by cationic surfactant CTAB in the presence of nanoparticles exhibiting various zeta potentials (frequency and dissipation change to 3rd overtone).

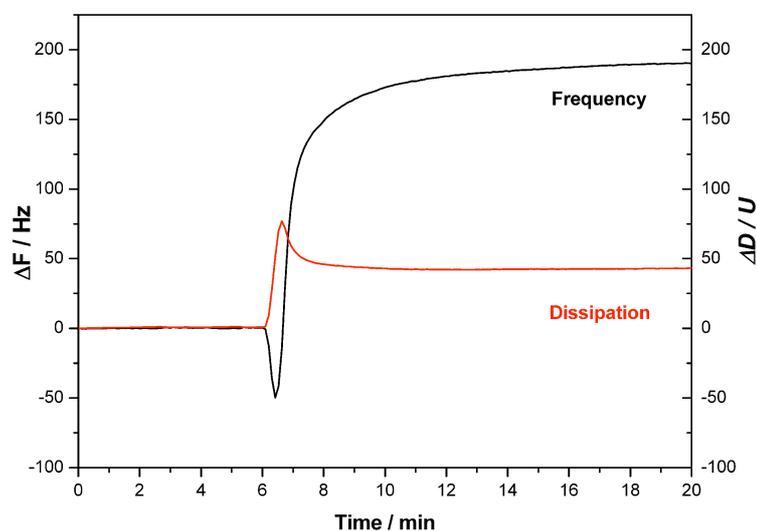


Figure S42. Frequency changes to 3rd overtone after exposure of the tristearin coated sensor to: i) 0-6 minutes, water; ii) 6-20 minutes, CTAB solution. [CTAB] = 2 mmol/L, T = 25 °C. The maximum lipid removal of 240 Hz was achieved after exposure to CTAB solution for *ca.* 12 minutes.

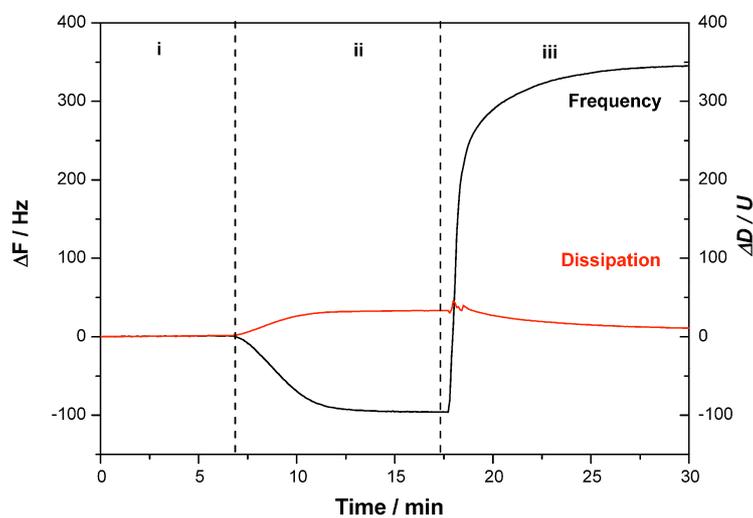


Figure S43. Frequency and dissipation changes to 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water; ii) 6-17 minutes, ND97 was used to deposit nanoparticles; and iii) 17-30 minutes, 2 mmol/L CTAB solution was used for cleaning. The maximum lipid removal of 392 Hz was achieved after exposure to CTAB solution for *ca.* 50 minutes. [ND97] = 0.1 g/L, [CTAB] = 2 mmol/L, T = 25 °C.

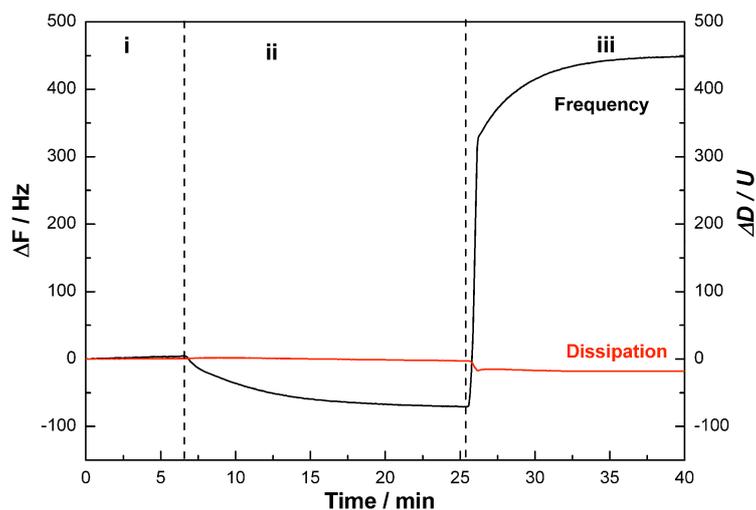


Figure S44. Frequency and dissipation changes to 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water; ii) 6-25 minutes, suspension of **2** was used to deposit nanoparticles; and iii) 25-40 minutes, 2 mmol/L CTAB solution was used for cleaning. The maximum lipid removal of 392 Hz was achieved after exposure to CTAB solution for *ca* 50 minutes. [**2**] = 0.1 g/L, [CTAB] = 2 mmol/L, T = 25 °C.

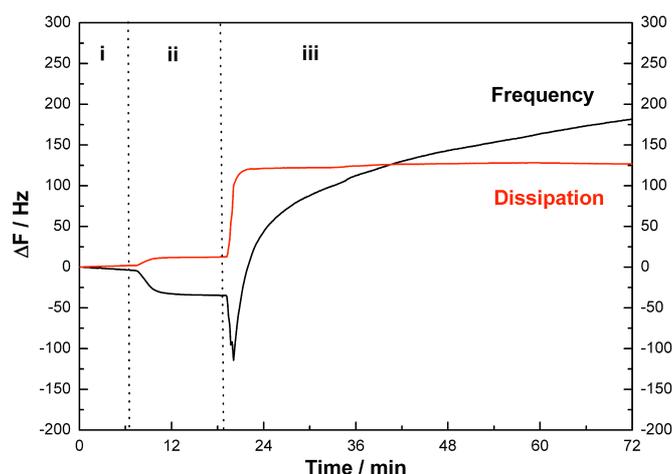


Figure S45. Frequency and dissipation changes on 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water; ii) 6-19 minutes, suspension of **4** was used to deposit nanoparticles; and iii) 19-72 minutes, 2 mmol/L CTAB solution was used for cleaning. The maximum lipid removal of 300 Hz was achieved after exposure to CTAB solution for *ca* 53 minutes. [**4**] = 0.1 g/L, [CTAB] = 2 mmol/L, T = 25 °C.

S10. QCM-D data for lipid removal at 15 °C by anionic surfactant SDBS and non-ionic surfactant G₁C₁₀ (frequency and dissipation change to 3rd overtone).

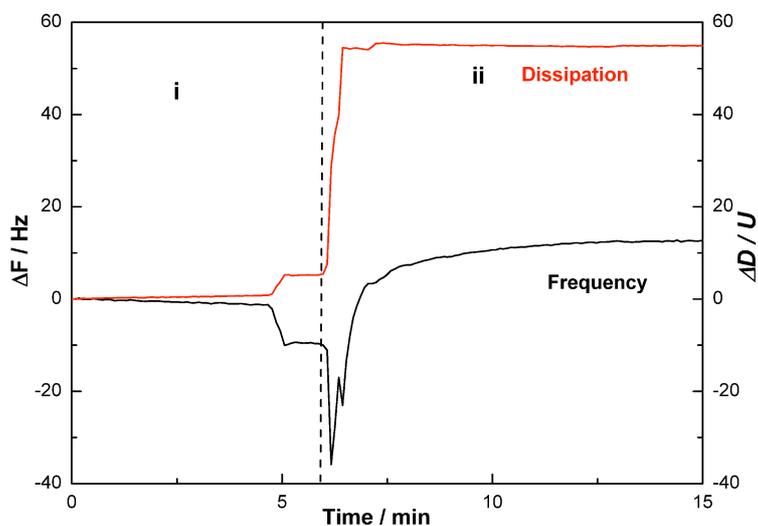


Figure S46. Frequency and dissipation changes on 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water; and ii) SDBS solution. [SDBS] = 40 mmol/L, T = 15 °C.

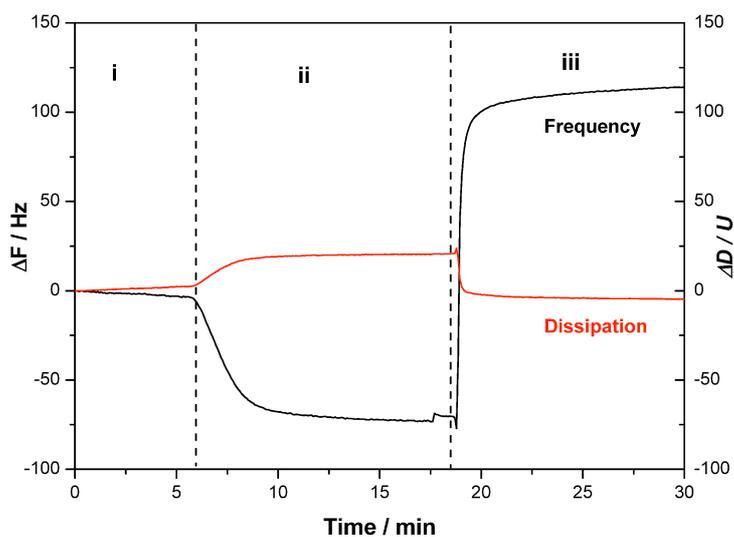


Figure S47. Frequency and dissipation changes to 3rd overtone after exposure of the tristearin coated sensor to: i) 0-7 minutes, water; ii) 6-18 minutes, ND97 suspension was used to deposit nanoparticles; and iii) 18-30 minutes, 40 mmol/L SDBS solution was used for cleaning. [ND97] = 0.1 g/L, [SDBS] = 40 mmol/L, T = 15 °C.

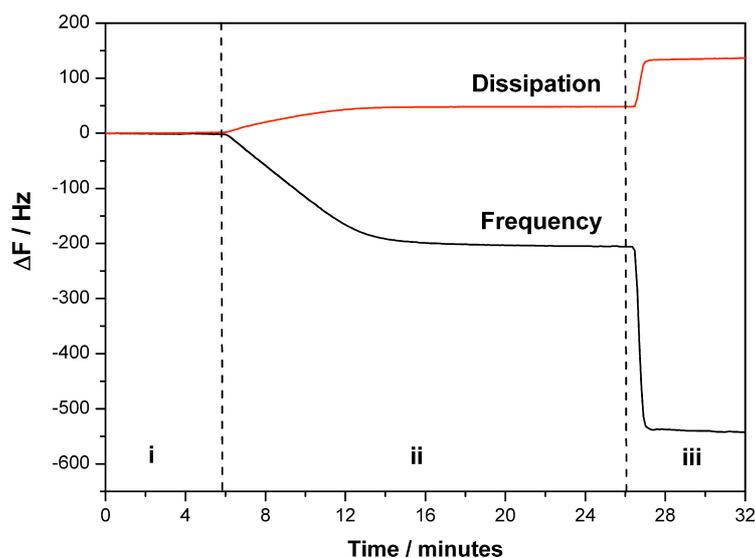


Figure S48. Frequency and dissipation changes to 3rd overtone when the tristearin coated sensor was exposed to the following: i) 0-6 minutes, water is used to obtain a stable baseline; ii) 6-26 minutes, ND97 colloid was used; iii) 26-32 minutes G₁C₁₀ solution was used. [G₁C₁₀] = 3.1 mmol/L, [ND97] = 0.1 g/L, T = 15 °C.

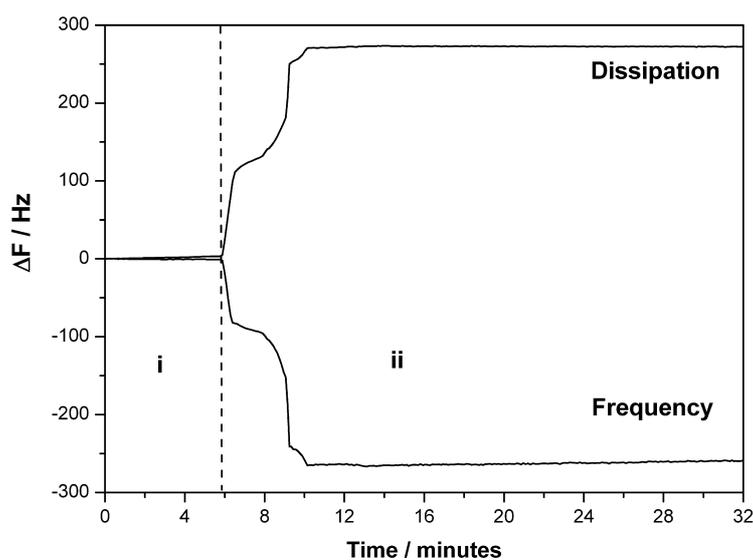


Figure S49. Frequency and dissipation changes to 3rd overtone when the tristearin coated sensor was exposed to the following: i) 0-6 minutes, water is used to obtain a stable baseline; ii) 6-26 minutes, G₁C₁₀ solution were used, to remove tristearin lipid; iii) 26-32 minutes G₁C₁₀ solution was used for two sensors, to clean the QCM system. [G₁C₁₀] = 3.1 mmol/L, T = 15 °C.

Supporting Information References

- (1) Höök, F.; Rodahl, M.; Kasemo, B.; Brzezinski, P. *Proc. Natl. Acad. Sci. U. S. A.* **1998**, *95*, 12271.
- (2) Voinova, M. V.; Rodahl, M.; Jonson, M.; Kasemo, B. *Phys. Scr.* **1999**, *59*, 391.
- (3) Rodahl, M.; Hook, F.; Fredriksson, C.; A. Keller, C.; Krozer, A.; Brzezinski, P.; Voinova, M.; Kasemo, B. *Faraday Discuss.* **1997**, *107*, 229.
- (4) Ohlsson, G.; Tigerstrom, A.; Hook, F.; Kasemo, B. *Soft Matter* **2011**, *7*, 10749.
- (5) Christov, N. C.; Denkov, N. D.; Kralchevsky, P. A.; Broze, G.; Mehreteab, A. *Langmuir* **2002**, *18*, 7880.