# SURFACE MODIFICATION OF COLLOIDAL SILICA SOL WITH LONG-CHAIN FATTY ACIDS: INTERFACIAL INTERACTIONS, COLLOIDAL STABILITY, AND HYDROPHOBIC PROPERTIES

**Baxranova Dilnoza Axmedovna**
**Ramazonova Sitora Amrillo qizi**
Zarmed University.

***https://doi.org/10.5281/zenodo.24079278***

***Abstract.*** *Long-chain fatty acids may alter the wettability and interfacial behavior of colloidal silica, but adsorption, free-acid deposition, and covalent grafting are often conflated.*

*This article critically reviews evidence concerning palmitic (C16:0), stearic (C18:0), and oleic (C18:1) acids and proposes a reproducible framework for evaluating modification while retaining dispersion stability. Published findings support hydrogen bonding and physical adsorption as plausible interactions on unactivated silica. Oleic-acid-treated silica has stabilized approximately 200 nm Pickering emulsion droplets, while a silica–stearic-acid coating system showed a water contact angle of 142 ± 6°; alumina activation has separately increased stearic-acid attachment. These results refer to different material configurations and are not directly comparable. A controlled study combining FTIR, dynamic light scattering, electrophoretic mobility, thermogravimetry, adsorption mass balance, and contact-angle measurements is recommended. pH, modifier loading, solvent composition, ionic strength, and acid unsaturation should be controlled. Durable modification should be claimed only when retained surface species and application-level effects are demonstrated after washing and storage.*

***Keywords:*** *colloidal silica; silica sol; fatty acids; oleic acid; stearic acid; adsorption; hydrophobicity; colloidal stability.*

## 1. Introduction

Colloidal silica sol is a dispersion of amorphous silicon dioxide particles, commonly in water. Its high specific surface area and surface silanol groups (≡Si–OH) support applications in coatings, composites, and interfacial stabilization. However, these hydrophilic groups can hinder compatibility with nonpolar materials and contribute to aggregation when solution conditions change. Surface modification is therefore used to tune wettability and particle interactions [1,2].

Palmitic, stearic, and oleic acids combine a polar carboxyl group with a hydrophobic hydrocarbon chain. Palmitic acid is a saturated C16 acid, stearic acid a saturated C18 acid, and oleic acid an unsaturated C18 acid. Their structures offer useful comparators for studying chain length and unsaturation. Yet adding an acid to silica sol does not by itself prove chemical grafting: free acid, precipitated crystals, molecular aggregates, and reversibly adsorbed species may coexist [3–5].

Prior infrared studies associate fatty-acid carboxyl groups with surface silanols through hydrogen bonding [4,5]. Other studies show that oleic-acid adsorption can change the interfacial behavior of charged silica [6], that fatty-acid/silica coatings can be water-repellent [7], and that alumina activation can increase stearic-acid retention [8]. Because these studies use different materials and endpoints, their values should not be treated as measurements of the same system.

This review evaluates the plausible interactions and proposes measurements to distinguish surface association, colloidal stability, and film wettability.

## 2. Materials and Methods

### 2.1. Literature scope and proposed materials

A targeted, non-systematic review of peer-reviewed primary studies and authoritative reviews was conducted using combinations of “colloidal silica”, “silica sol”, “fatty acid adsorption”, “stearic acid”, “oleic acid”, “silanol”, “hydrophobic coating”, and “dispersion stability”. Studies were selected for mechanistic evidence, a defined silica–fatty-acid system, or relevant characterization methods. This is not an exhaustive review or meta-analysis; no new laboratory results are reported.

For a comparative experiment, use an aqueous silica sol with documented $SiO_2$ content, particle size, initial pH, conductivity, and counterions; analytical-grade palmitic, stearic, and oleic acids; deionized water; and a justified cosolvent such as ethanol. Include untreated silica, solvent-matched silica, and fatty-acid-only controls. First verify modifier solubility and confirm that the cosolvent does not destabilize the sol.

### 2.2. Experimental design and characterization

A screening design may compare acid identity, acid-to-silica mass ratio (for example, 0, 0.025, 0.050, and 0.100 g $g^{-1}$), and initial aqueous-phase pH (for example, 7.0, 8.5, and 9.5).

These are proposed design levels, not optimized conditions. Introduce solvent and pH adjustments incrementally and equally across controls. Keep mixing time and temperature constant, and record turbidity, crystallization, gelation, and phase separation. Separate unbound acid using a validated ultrafiltration or equivalent method with recovery controls. Retain one portion as a sol for stability tests; wash and dry another for surface analysis. Prepare coating films on identically cleaned substrates using the same deposition and drying protocol. The treatment sequence should be defined in advance: equilibrate the silica, add the modifier solution slowly under controlled stirring, maintain the selected temperature for a fixed contact time, and then separate the particle fraction.

Report the silica concentration and all liquid volumes so the final solvent fraction is identical across samples. A fatty-acid-only blank should pass through the same filtration or centrifugation steps to reveal acid retention by the membrane or vessel. If a visible precipitate forms, do not count it as modified silica; examine the separated solid and supernatant independently. These controls are particularly important for the saturated acids, whose apparent solubility can be sensitive to temperature and solvent composition.

Use FTIR to examine alkyl C–H bands and changes in carboxylic-acid/carboxylate and Si–OH signals, comparing neat acids, untreated silica, and washed samples. An alkyl band alone cannot establish grafting. TGA can estimate organic-associated mass loss after accounting for adsorbed water and silica dehydroxylation; XPS may add chemical-state evidence. Measure hydrodynamic size by DLS, electrophoretic mobility (reporting the model used to calculate zeta potential), and sedimentation under controlled concentration, pH, ionic strength, and temperature.

Interpret DLS distributions cautiously because small numbers of aggregates can dominate intensity-weighted values. Report the measurement temperature, dilution medium, equilibration time, number of runs, and whether the result is intensity-, volume-, or number-weighted.

Dilution itself can shift equilibria, so samples should be diluted in a medium matched for pH and ionic strength where feasible. Electrophoretic mobility should be reported alongside the calculated zeta potential and the conversion model, because a zeta-potential value is not independent of solution conductivity and viscosity.

Visual sedimentation observations are simple but should use the same vial geometry, fill volume, and storage temperature. Together, these measurements indicate whether surface treatment changes dispersion behavior; none alone establishes chemical bonding.

Measure static water contact angle only on reproducibly prepared films and interpret it alongside surface roughness; it is not a direct measure of grafting on dispersed particles. Include short-term and seven-day storage, wash resistance, or redispersion tests. Where a validated free-acid assay is available, apparent uptake may be estimated as $q = (C_0 - C_e)V/m$, with $C_0$ and $C_e$ denoting initial and equilibrium acid concentrations, V the liquid volume, and m the dry silica mass. Correct for precipitation and extraction recovery. Use at least three independently prepared batches per condition and report dispersion measures and confidence intervals. No statistical tests were performed in this review. If particle size is followed during storage, report the ratio $R(t) = D_h(t)/D_h(0)$, where $D_h$ is the hydrodynamic diameter at time t, together with polydispersity and electrophoretic mobility. This normalized comparison helps distinguish initial size differences from subsequent growth, but should not replace the absolute size distribution. If specific surface area is measured independently, an approximate molecular surface density may be estimated from retained organic mass; this value remains conditional on the purity of the bound fraction and the assumed molecular mass and footprint. Predefine primary outcomes and statistical procedures before examining the data, and use comparisons among independently prepared batches rather than treating repeated instrument scans of one sample as independent replicates.

**3. Results: Critical Synthesis of Published Evidence**

On native silica, the evidence supports physical association more clearly than covalent attachment. Hasegawa and Low reported physical adsorption of stearic acid on silica, while observing stearate-layer formation on alumina under their studied conditions [3]. Marshall and Rochester found hydrogen-bond-associated adsorption of oleic and linolenic acids on silica; treatment could alter adsorption behavior [4]. Blyholder et al. related infrared changes in carbonyl and Si–OH bands to oleic-acid interaction with silica [5]. Until wash resistance and chemical-state evidence are shown, “fatty-acid-treated” or “fatty-acid-adsorbed silica” is the more defensible description.

In aqueous dispersions, Sadeghpour et al. reported that oleic-acid adsorption altered silica interfacial behavior while preserving particle charge and dispersion stability, enabling Pickering emulsions with droplets of approximately 200 nm [6]. This is an emulsion droplet size, not a silica primary-particle diameter.

The result suggests that acid loading and medium composition can modify interfacial affinity without necessarily causing bulk flocculation; it does not establish equivalent behavior for palmitic or stearic acid.

For coatings, Heale et al. reported a water contact angle of 142 ± 6° and a tilting angle of 16 ± 2° for a pipette-deposited system containing hydrophobic silica nanoparticles and octadecanoic acid [7]. The performance reflects both low surface energy and hierarchical roughness, so the values do not describe an aqueous sol or isolated silica particles.

Gan et al. found that alumina-coated silica had increased reactivity toward stearic acid; a reported 15% alumina coating corresponded to a 350% increase in grafting density relative to the reference and a 124° contact angle [8]. This activated material is not equivalent to unmodified silica sol.

**Table 1. Selected evidence and limits of interpretation**

| Study / system | Reported outcome | Interpretive limit |
|---|---|---|
| Hasegawa & Low [3] | Physical stearic-acid adsorption on silica; stronger layer formation on alumina. | Different oxide surfaces and non-aqueous conditions. |
| Sadeghpour et al. [6] | Oleic-acid-treated silica stabilized ~200 nm emulsion droplets. | Droplet size is not silica particle size. |
| Heale et al. [7] | C18 acid/silica coating: contact angle 142 ± 6°. | Film roughness and deposition affect wetting. |
| Gan et al. [8] | Alumina activation increased stearic-acid grafting density. | Activated silica is not native silica sol. |

Taken together, the reports answer different questions. Infrared adsorption studies address molecular association at oxide surfaces; emulsion work tests whether treated particles can stabilize an oil–water interface; coating studies measure the behavior of a dried, rough film; and alumina activation changes the chemical identity of the oxide surface. A contact angle from a textured coating therefore cannot be used as a proxy for fatty-acid coverage on suspended silica, just as emulsion droplet size cannot be interpreted as a particle-size measurement. These distinctions are essential when translating promising results into a sol formulation. The strongest inference across the studies is that fatty-acid chemistry can tune interfacial performance, while the durability and mechanism of attachment depend on material configuration and processing.

**4. Discussion**

The likely interaction combines carboxyl-group association with surface silanols and exposure of hydrocarbon tails toward the surrounding phase. Attachment strength depends on hydrogen bonding, acid dissociation, solvent quality, tail association, and electrostatic conditions.

Carboxylate-rich species may also experience electrostatic repulsion from negatively charged silica, so irreversible ionic binding cannot be assumed [3–5,8]. The cis double bond in oleic acid may change chain packing relative to stearic acid, while palmitic acid provides a shorter-chain comparator. These are hypotheses for matched experiments, not established rankings of performance. Surface silanol density, acid ionization, ionic strength, counterion identity, and the amount of cosolvent can each shift the balance between adsorption and aggregation [1,9–11].

Consequently, a high nominal acid loading cannot be assumed to produce either greater retained coverage or a more stable sol. Near the silica isoelectric region or at elevated electrolyte concentration, electrostatic repulsion may weaken; an organic modifier or a solvent shift can further alter particle interactions. The optimum condition, if one exists, will therefore depend on the starting silica and the intended end use rather than on fatty-acid identity alone.

Successful modification should be evaluated with three linked criteria: (1) the sol retains controlled particle size and reproducible surface charge; (2) fatty-acid-derived species remain after a defined washing or separation step; and (3) standardized films show a reproducible wettability change that persists during storage or water exposure. Mass balance, FTIR, TGA, and a desorption test provide a minimum evidence set for persistent adsorption. A claim of covalent Si–O–C grafting requires structure-specific proof beyond an isolated carbonyl shift or alkyl band. Conditions that cause irreversible aggregation should be excluded before film comparisons.

Native silica and alumina-activated silica, if compared, need separate matched controls.

A practical sequence is to screen solvent compatibility first, then determine the maximum modifier loading that retains a uniform dispersion, and only then prepare films from the stable formulations. This sequence prevents a coating result from obscuring failure of the precursor sol.

The control set should include unmodified silica, solvent-only silica, acid-only samples subjected to the same separation, and an activated-silica reference only when activation is part of the study. Record sample recovery at each separation step so apparent uptake is not simply a consequence of acid precipitation or particle loss.

The main limitation is heterogeneity among published silica morphologies, solvents, substrates, and treatment routes, which prevents quantitative cross-study ranking. Future work should measure adsorption isotherms and desorption kinetics, compare all three acids under identical conditions, test relevant ionic strengths and aging, and connect surface chemistry to coating transparency, adhesion, and durability.

For waterborne formulations, a useful target is a balance between hydrophobicity and stable dispersibility, not maximum contact angle alone. The review is limited by the small number of studies that directly examine the same fatty acid on the same native silica sol using common protocols. Some cited findings concern silica gel, dried coatings, non-aqueous adsorption, or alumina-modified particles. Their measurements inform mechanism and experimental planning, but they do not supply a single performance benchmark.

Future reports should state the silica source, surface area, particle-size distribution, solution pH and conductivity, solvent fraction, mixing and aging conditions, and washing procedure. These details are necessary for reproducibility and for judging whether the measured organic fraction is associated with particles or remains as a separate phase.

Additional useful endpoints include coating adhesion, abrasion resistance, transparency, and repeated wet–dry cycling, particularly if the target application is a durable water-repellent surface.

For dispersion applications, accelerated aging should be selected cautiously and accompanied by real-time storage because elevated temperature can change both fatty-acid phase behavior and silica aggregation kinetics. Comparative claims should be limited to matched conditions and should report unsuccessful or unstable formulations as well as the best-performing samples. Such reporting would reduce selection bias and help identify the practical operating window rather than a single favorable result.

## 5. Conclusions

Higher fatty acids can modify the surface and interfacial behavior of colloidal silica, but direct treatment of unactivated $SiO_2$ is best described as adsorption unless durable chemical attachment is demonstrated. Published studies show oleic-acid-treated silica used in Pickering emulsions, high contact angles in silica–fatty-acid coatings, and increased stearic-acid attachment after alumina activation.

These distinct systems illustrate potential applications but cannot be ranked as equivalent silica-sol outcomes. A controlled comparison of palmitic, stearic, and oleic acids should vary pH, solvent composition, and modifier loading, and combine FTIR, DLS, zeta potential, TGA, mass balance, and standardized contact-angle tests with washing and storage evaluation. Such measurements are needed before making original experimental claims about durable modification.

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