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Sulfuric Acid on Europa and the Radiolytic Sulfur Cycle

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A comparison of laboratory spectra with Galileo data indicates that hydrated sulfuric acid is present and is a major component of Europa's surface. In addition, this moon's visually dark surface material, which spatially correlates with the sulfuric acid concentration, is identified as radiolytically altered sulfur polymers. Radiolysis of the surface by magnetospheric plasma bombardment continuously cycles sulfur between three forms: sulfuric acid, sulfur dioxide, and sulfur polymers, with sulfuric acid being about 50 times as abundant as the other forms. Enhanced sulfuric acid concentrations are found in Europa's geologically young terrains, suggesting that low-temperature, liquid sulfuric acid may influence geological processes.

Europa is unique among Jupiter's moons. It is differentiated (1) with an icy crust that may melt from tidal heating to produce a subsurface ocean (2). The surface may be young and renewed by solid state convection (3) or ex-

trusion of solid or liquid material from below (4). Europa's surface exhibits bright, icy plains and younger, darker mottled terrain (4). Linear features with different morphologies crisscross the surface. These long (>1000 km), narrow (10 to 20 km) features often show bright central bands flanked by bands of darker material ("triple bands"). Because Europa is within Jupiter's energetic magnetosphere, the surface is subject to intense bombardment by high-energy plasma (e^- , H^+ , O^{n+} , and S^{n+}) (5–7) that can alter the

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surface by radiolysis and ion implantation (8-10).

Europa's surface consists mainly of H₂O ice and hydrated materials (11, 12), with minor amounts of SO₂, CO₂, and H₂O₂ (9, 10, 13). Hydrated compounds on Europa were suggested by shifts and distortions of H₂O absorption bands found in spectra obtained by Galileo's near-infrared mapping spectrometer (NIMS) (11, 12). These spectra vary between two extremes (end-members), from icelike, with a symmetric absorption band at 2 µm in wavelength, to predominately hydrated, with highly distorted H₂O bands (Fig. 1A). Most NIMS spectra of Europa's surface indicate mixtures of these two end-members. Although the hydrated material is ubiquitous on Europa, there is more of it within the linear features and the dark mottled terrain (12) of Europa's trailing (in relation to orbital motion) hemisphere, which suffers the greatest plasma bombardment and implantation (5).

It was suggested (12) that Europa's spectra are similar to those of hydrated salt minerals (Fig. 1B) such as epsomite (MgSO₄·7H₂O), magnesium hexahydrate (MgSO₄·6H₂O), and natron (Na2CO3.10H2O). Fits based on combining spectra of several salts were suggested to indicate a subsurface ocean (12), with salt minerals being formed on Europa's surface (14) by extrusion of subsurface brine that evaporates to form salt pans, similar to terrestrial deposits. However, identification of specific hydrates in the spectra is not unique (12). The presence of H₂O₂ and SO₂ in the icy surface of Europa suggests that sulfuric acid (H_2SO_4) , a common photochemical product in the atmospheres of Earth and Venus, might be formed on Europa. Here, we present laboratory spectra showing that NIMS observations of the europan surface are consistent with the presence of hydrated H₂SO₄ and show that H₂SO₄ is part of a radiolytic sulfur cycle.

Fig. 1. IR reflectance spectra of Europa and candidate surface materials. (A) NIMS Europa end-member spectra. Ice abundance is much greater than hydrate (solid line), and hydrate abundance is much greater than ice (line with circles). (B) Evaporite salt minerals (12). (C) Spectra of H₂SO₄. nH₂O for various n, grain size (d), and temperature (T): n = 8, d = 5 μ m, and T = 80 K, offset vertically by 0.10 (shortdashed line); n = 8, d =50 μ m, and T = 80 K, offset vertically by 0.05

Frozen H_2SO_4 hydrates occur as H_2 $SO_4 \cdot nH_2O_5$, where n = 1, 2, 3, 4, 6.5, and $\bar{8}$ (15-17). We investigated the n = 4, 6.5, and 8hydrates by freezing stoichiometric solutions and obtaining their infrared (IR) reflectance spectra. The solutions contained spectrally neutral diamond powder to produce diffusely reflecting media. Samples with various grain sizes (effective diameters of \sim 5 to \sim 50 μ m) were prepared, and spectra were obtained for temperatures between ~ 80 K and their melting points (\sim 220 to 230 K). The spectra (Fig. 1C) of H₂SO₄·6.5H₂O and H₂SO₄·8H₂O are similar and show little variation with temperature. The hydration bands of H2SO4·4H2O were found to be less distinct than those for the n = 6.5 and 8 hydrates and unlike those in Europa's spectra. Laboratory spectra of H₂SO₄·8H₂O are similar to the spectrum of Europa's hydrated material (Fig. 1D) and provide the best match yet found for any single compound (18).

The small differences between the Europa and laboratory spectra (Fig. 1D) may be due to several effects. (i) The Europa spectrum also contains H₂O ice features, inclusion of which may improve the comparison, particularly in the 1.2- to 1.4-µm region. (ii) The wavelengths of reflectance minima for hydration bands are reduced by ion irradiation and partial dehydration. Shifts of $\sim 0.02 \ \mu m$ were produced by proton irradiation (19), and spectra of partially dehydrated sulfates exhibit shifts of 0.04 μ m (11). Such effects may be responsible for the small mismatch of band positions found in the Europa and laboratory spectra. (iii) The grain sizes of the laboratory samples may be different from those on Europa's surface. (iv) Other hydrates may also be present in Europa's spectrum.

With the above effects, the match to the NIMS spectra suggests that $H_2SO_4 \cdot nH_2O$ (n = 6.5 and 8) is a major component of Europa's optically sensed surface. The opti-

cal sampling depth is a few grain diameters [~60 μ m on Europa (9)], so we adopted 200 μ m as the effective sampling depth. The number of H₂SO₄ hydrated molecules for Europa's surface area (whose spectrum is given in Fig. 1D) is ~7 × 10¹⁹ cm⁻² (200 μ m)⁻¹. The average number of H₂SO₄ molecules for Europa's trailing hemisphere is ~3 × 10¹⁹ cm⁻² (200 μ m)⁻¹.

NIMS spectra of hydrated regions are featureless in the 3- to 5-um region. We measured the mid-IR diffuse reflectance of a frozen H₂SO₄·8H₂O sample and found no spectral features in this range, consistent with NIMS spectra. We also measured the ultraviolet (UV) properties of H₂SO₄·8H₂O. An absorption band was found at 190 nm, with weaker absorption extending to 240 nm. A UV spectrum of Europa's trailing side (13) shows a possible absorption onset at 225 nm, which could be due to H_2SO_4 hydrate absorption. However, SO₂ also has an absorption band beginning at these same wavelengths (20), and because a longer wavelength SO₂ band appears in Europa's UV spectrum, the 225-nm onset could be due to SO_2 alone. Therefore, Europa's UV spectrum is consistent with H_2SO_4 hydrates existing on Europa.

The H_2SO_4 concentration correlates with Europa's dark material. Sulfuric acid hydrates are visually colorless, so the dark material is a related compound acting as a source, sink, or both. Sulfur has been suggested as a constituent for Europa's dark material (4, 21). We compare the visible reflectance of Europa's trailing side (22) to the reflectance of polymerized sulfur produced by radiolysis and photolysis (23–25)





Fig. 2. Reflectance spectra of Europa's trailing side compared to photolyzed and radiolyzed sulfur compounds. We compare Europa's spectrum (22) (solid dots); photolyzed $H_2S + 10H_2O$ (23) (solid line); the transmission of ion-irradiated SO_2 (24), assumed here to approximate the reflectance when the same number of molecules is in the optical path (dashed line); and the SO_2^+ -irradiated H_2O ice (25), assumed to represent implanted and irradiated sulfur in H_2O ice (dotted line with circles).

(long-dashed line); n = 8, $d = 50 \,\mu\text{m}$, and $T = 140 \,\text{K}$ (solid line); and n = 6.5, $d = 50 \,\mu\text{m}$, and $T = 80 \,\text{K}$ (dotted line). (**D**) Comparison of Europa's hydrate spectrum [line with circles, from (A)] and H₂SO₄·8H₂O at $T = 140 \,\text{K}$ and $d = 50 \,\mu\text{m}$, normalized at 1.1 μm [solid line, from (C)].

(Fig. 2). The shape of Europa's spectrum (approximately bilinear, with a breakpoint at 0.5 µm) is similar to the laboratory spectra and suggests that altered sulfur compounds are present on the surface. Because the visible reflectance of these materials and Europa are about the same, we estimate that the amount of polymerized sulfur in Europa's optically sampled layer is $\sim 4 \times 10^{17}$ atoms cm⁻² (200 μ m)⁻¹ (24, 25). Therefore, we suggest three chemical reservoirs for Europa's sulfurous compounds: (i) polymerized sulfur, (ii) SO₂, with optically sensed surface densities of $\sim 2 \times 10^{17}$ cm^{-2} (200 µm)⁻¹ (13), and (iii) H₂SO₄ hydrate. Because extensive radiation dosage occurs to depths of 200 µm in geologically short times (6), these reservoirs are in radiation-induced equilibrium, with sulfur being continuously cycled among these forms.

The production of H_2SO_4 hydrate in this radiolytic cycle can be estimated with the Gvalue, the number of molecules produced per 100 eV of energy absorbed. Laboratory radiolysis of sulfur hydrosols produces sulfate (26), and irradiation of polymerized sulfur in Europa's ice may be similar. For hydrosols, $G(SO_4) \approx 0.6$ and applies to concentrations that are $<10^{-4}$ of Europa's polymerized sulfur concentration. However, these data apply to liquid suspensions and not to frozen mixtures. Irradiation of a 10:7 mixture of H₂O:SO₂ ice produced compounds whose complicated 2.5- to 25-µm spectra contain features that may be due to H_2SO_4 hydrates (27). Radiolysis of SO₂ ice produces sulfur, sulfate, and (predominately) SO_3 (28). In the presence of H_2O , SO_3 rapidly forms H_2SO_4 in an exothermic reaction (29). The G value for SO₂ in H₂O ice is not available, so we used the value for SO₃ production in pure SO₂ ice ($G \approx 5$) (28). With this G value, Europa's SO₂ concentration, and a value of \sim 5 \times $10^{10} \text{ ke} \tilde{\text{V}} \text{ s}^{-1} \text{ cm}^{-2}$ for Europa's irradiation flux (6), we obtained an H_2SO_4 production of $\sim 1 \times$ $10^9 \text{ s}^{-1} \text{ cm}^{-2}$. Irradiation of polymerized sulfur in H₂O ice and associative reactions of SO₂ with H_2O_2 can also produce H_2SO_4 , so the above rate is a lower limit. The optically observed H₂SO₄ surface concentration [$\sim 3 \times 10^{19} \text{ cm}^{-2}$ (200 μ m)⁻¹] can be formed from SO₂ and polymerized sulfur in $<10^4$ years.

Sulfuric acid is quite stable under irradiation. When hydrogen is removed from H_2SO_4 , it recombines quickly (30). Destruction of SO₄ is moderated by reactions between SO₂ and H₂O₂ molecules produced during radiolysis (31). The net destruction rate of H₂SO₄ can be estimated from rates for another acid sulfate, Li₂SO₄, which exhibits $G \approx 10^{-3}$ to 10^{-4} (32). These values are consistent with upper limits for H₂SO₄ at Europa's concentration levels (31). The high radiation stability of H₂SO₄, particularly for the n = 6.5 and 8 hydrates (31), implies large concentrations of H₂SO₄ in comparison to SO₂ and polymerized sulfur. The observations show that Europa's H₂SO₄ is more abundant than either SO₂ or polymerized sulfur by a factor of ~100 (assuming that the photon sampling depths are about the same). The *G* values, obtained for x-ray and gamma-ray irradiation, give lower bounds for destruction by energetic particles of ~5 × 10⁷ to 5 × 10⁸ s⁻¹ cm⁻². Heavy ions and UV radiation can also decompose H₂SO₄ in the upper few micrometers of the surface (δ), but *G* values and cross sections are not available.

The original source of sulfur may be S^{n+} ions implanted from the jovian plasma. The plasma influx, $\sim 6 \times 10^7$ sulfur ions cm⁻² s⁻¹ (7), provides a sufficient amount of sulfur to account for the three reservoirs in 10^4 years. However, implantation would be expected to produce a more uniform surface distribution than is observed, so if the ultimate source of sulfur is plasma implantation, then some geological process has acted to produce the observed nonuniform distribution. One possibility is the burial of implanted sulfur and its radiolytic products by solid state convection, forming a crust enriched with H₂SO₄, SO₂, and sulfur polymers. Subsequent convection of matter back to the surface may produce a nonuniform distribution of sulfurous material. Cracks in this crust can be filled by the extrusion of ice or liquid H₂O from below. This upwelling material can scour the crack walls, forming two layers of upward moving sulfurous till. The surface expression would be two dark bands and possibly a bright median of pure ice, accounting for the appearance of Europa's triple bands (4). Alternatively, much of the sulfurous material on Europa's surface may have been endogenic. Sulfuric acid formed internally could be geologically emplaced onto the surface. The extrusion of salt-rich brine (12) may have provided sulfate salts to the surface, where proton implantation can replace salt mineral cations to form H₂SO₄. Radiolysis of such evaporite salt minerals should also produce MgO, MgOH, Mg(OH)₂, Na₂O, and NaOH (14, 33). However, hydroxides exhibit 1.4-µm bands (34) that are not evident in NIMS spectra (Fig. 1A) and imply hydroxide concentration limits of <10%.

The melting point of H_2SO_4 solutions can be as much as 55 K lower than that of pure H_2O ice (15), so liquid H_2SO_4 may be formed more readily by tidal heating compared to melted brine. Enhanced H_2SO_4 concentrations are associated with geological structures whose formation may have been affected by the presence of liquid H_2SO_4 . Sulfuric acid can supercool to 150 K (16), possibly facilitating cryovolcanic activity. The acid is also an electrical conductor and may contribute to Europa's magnetic signature (35).

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