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Synthesis of Mineral Semiconductor Thin Films: Toward Mineral-Based Photosynthesis

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Is there an Archaean TTG series?

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The Archaean continental crust is dominated by grey orthogneisses, typically described as being of TTG (Tonalite—Trondhjemite—Granodiorite) composition. TTG are juvenile, sodic ($\text{Na}_2\text{O} > \text{K}_2\text{O}$) granitoids with high La/Yb and Sr/Y ratios, reflecting melting of a garnet-present mafic source. However, the Archaean plutonic complexes are not exclusively made of TTG; they typically feature a large range of rock compositions, all with low Y (< 30 ppm) and Yb (< 3 ppm) contents and correlatively high Sr/Y or La/Yb ratios, ranging from “true” TTG to granodiorites or monzogranites, with $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratios up to 2 or 3; the potassic components can occur either as well-defined, individual plutons (“late potassic granites”) or as component of the “grey gneiss” complexes. TTG appear as the sodic end-member of a wider family of Archaean granitoids (Tonalite—trondhjemite—granodiorite—monzogranite, or TTGM).

This compositional spread reflects the melting of a range of sources, from mafic to felsic, yielding melts ranging from sodic to mildly potassic.

Furthermore, the TTG s.s. can be further subdivided in a high-silica, high Sr/Y (and La/Yb), high Na subseries (trondhjemites and (leuco-)tonalites, or TT), and a low-silica, lower Sr/Y and La/Yb, lower Na subseries (tonalites, occasionally diorites or granodiorites, TD). The TT subseries reflects low melt fractions and deep (20 kbar) melting of a mafic source; the TD series corresponds to shallower melting (12 kbar) and higher melt fractions.

The TTGM series that form the Archaean Grey Gneisses define a range of composition between at least three end-members: (1) tonalites, granodiorites and monzogranites corresponding to intracrustal melting of felsic lithologies (older grey gneisses); (2) tonalites and trondhjemites, formed by deep and cool melting of a mafic source; (3) tonalites and diorites, formed by shallow and hot melting of a mafic source. Between these three end-members, there is a complete range of composition with intermediate rock types corresponding to diverse sources or depth of melting.

The term of “TTG” therefore corresponds to the grouping of two of the three main end-members of a range of compositions; extracting TTG from the wider “TTGM” group creates an artificial boundary between petrogenetically related rock types. TTG (or, worse, TTGM) are not one single, uniform rock type, but to a collection of plutonic rocks whose individual origins must be discussed before any geodynamic inference can be made.

Synthesis of mineral semiconductor thin films: Toward mineral-based photosynthesis

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The use of semiconductors as photocatalysts for carbon reduction has been the subject of research interest over the last few decades in the disparate but fundamentally related areas of solar energy conversion and research on the origins of life and of photosynthesis. Here, we combine the photocatalytic properties of hematite (toward water oxidation) with those of tausonite, whose conduction band is sufficiently energetic to reduce bicarbonate to formate.

We have used two different synthetic pathways for SrTiO_3 . Dip-coated films of SrTiO_3 made by a polymeric precursor method [1] show photocurrent activity. We have also made hematite films (originally made by chemical vapor deposition) dip-coated with SrTiO_3 particles that also show photocurrent activity. One purpose of coating hematite with SrTiO_3 is to put the SrTiO_3 in the role of hole scavenger [2]; the valence band of SrTiO_3 is at slightly higher energy than the valence band of hematite, so that holes generated in the hematite can be quickly filled by electrons from the SrTiO_3 valence band and thus removed from the high recombination rates encountered in hematite. This is anticipated to increase the overall efficiency of water oxidation over that exhibited by hematite alone.

Previous studies have documented the ability of SrTiO_3 (tausonite) to photoelectrochemically reduce carbon. However, measured quantum efficiencies for carbon reduction by SrTiO_3 are very low, and have been mainly attributed to back-reaction of the reduced compounds with photogenerated valence band holes on the semiconductor surface. This research aims to test the hypothesis that SrTiO_3 semiconducting thin films may more efficiently photoreduce carbon via a photoelectrochemical tandem cell with hematite (Fe_2O_3), in which the Fe_2O_3 photoanode can oxidize water to O_2 and the photocathode (SrTiO_3) can reduce CO_2 to formate. The tandem cell will physically separate the sites of carbon reduction and water oxidation, as well as isolate the conduction band of the photocathode through dye-sensitization, thereby eliminating the problems of back-reactions on SrTiO_3 .

[1] Zanetti *et al.* (1997) *Materials Lett.* **31**, 173.

[2] Wang *et al.* (2007) *J. Phys. D: Appl. Phys.* **40**, 3925.