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 (Na2O>K2O) granitoids with high La/Yb (Tonalite—Trondhjemite—Granodiorite) composition. TTG orthogneisses, typically described as being of TTG monzogranites, with K2O/Na2O ratios up to 2 or 3; the ratios, ranging from “true” TTG to granodiorites or (< 3 ppm) contents and correlatively high Sr/Y or La/Yb of rock compositions, all with low Y (< 30 ppm) and Yb exclusively made of TTG; they typically feature a large range of compositions; extracting TTG from the wider “TTGM” group of two of the three main end-members of a range of diverse sources or depth of melting.

Between these three end-members, there is a complete range of composition between at least three end-members: (1) tonalites, granodiorites and monzogranites (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 tawsomite, whose conduction band is sufficiently energetic to reduce bicarbonate to formate.

We have used two different synthetic pathways for SrTiO3. Dip-coated films of SrTiO3 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 SrTiO3 particles that also show photocurrent activity. One purpose of coating hematite with SrTiO3 is to put the SrTiO3 in the role of hole scavenger [2]; the valence band of SrTiO3 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 SrTiO3 valence band and thus removed from the high recombination rates encoutered 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 SrTiO3 (tawsomite) to photoelectrochemically reduce carbon. However, measured quantum efficiencies for carbon reduction by SrTiO3 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 SrTiO3 semiconducting thin films may more efficiently photoreduce carbon via a photoelectrochemical tandem cell with hematite (Fe2O3), in which the Fe2O3 photoanode can oxidize water to O2 and the photocathode (SrTiO3) can reduce CO2 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 SrTiO3.