Abstract: Diamondoids are a new class of molecular carbon nanomaterials composed of several fused diamond unit cells. Here we explore their ability as a work-function lowering coating on metallic surfaces. In particular, we show dramatic work function reduction on gold coated nanowires, equivalent to the best reported cesium coatings. We explain this unexpected phenomenon through a relatively stable radical carbocation. In addition, we describe how these types of high-temperature stable, low-work function materials could play an important role in a newly described high-temperature stable solar cell. These devices, based on photon-enhanced thermionic emission, operate by harvesting both heat and light from solar illumination, allowing direct electric power generation at high temperatures. Such devices could be combined with traditional thermal conversion cycles for high-efficiency combined cycle solar harvesting.