Carbon dioxide capture and sequestration will likely become an important technology in the next decades, as concern about climate change drives governments to consider regulating green-house gas emissions. Undoubtedly, one of the largest sources of human-generated CO2 is from coal-fired power plants. Thus, there has been a recent emphasis on assessing current CO2 separation technologies for application to dilute CO2 streams with large water contents, as well as developing new technologies for this separation. One of the most promising is adsorptive separation using amine-modified organic or inorganic polymer sorbents. Unfortunately, a significant drawback with sorbents of this type has been low CO2 sorbent capacities, requiring very large masses of (usually expensive) adsorbent. In this talk, we will describe a new, rationally designed hybrid aminopolymer-silica sorbent, deemed a hyperbranched aminosilica (HAS), with a very large, low temperature adsorption capacity. The material is low cost, owing to the simple method of synthesizing the adsorbent. In addition, the adsorbent can be used with effectively no change in capacity over multiple cycles (10 or more times), demonstrating the potential to have the durability needed for practical application. The sorbent is compared to the leading published supported amine sorbents. The structure of the sorbent and the attributes that endow it with excellent adsorption capacities are described.