In order to slow down the climate changes caused by human activities and control the carbon dioxide CO2 concentration in atmosphere, various CO2 separation and capture technologies have been investigated and applied. Solid sorbents for CO2 capture are considered as a potential alternative to replace the current energy consuming MEA (monoethanolamines) solution technology. In this work, we investigate the effects of Lewis-base modifiers on CO2 absorption/desorption abilities of well-known silica-supported amine-based sorbents. The primary amine-based polymers consisted of branched poly(ethylene amine) (PEI) and the Jeffamine® T403, having aliphatic and ether-containing backbones respectively. These polymers were dispersed on 14nm nanosilica with or without the addition of Lewis-basic polymer modifiers, poly(vinyl acetate) (PVAc), poly(methyl methacrylate) (PMMA) and poly(ethylene glycol) (PEG). And the sorbents’ CO2 absorption/desorption performance were investigated. The greatest effect for increasing the CO2 capture capacity was seen for PEG additives while other modifiers lowered the capture capacity. Meanwhile, PVAc and PMMA as well as T-403 and HMW PEG were found have positive influence on absorption kinetics. In desorption study, Lewis-base modifiers led to faster desorption rates and higher sorbent regeneration ability at relatively low temperature (45°C) without the need for further heating. Thus, a combination of amine containing, Lewis-base groups containing materials has the potential to be used in concert to fine tune the capacity, mass transfer, regeneration ability and kinetics for amines-based CO2 sorbents.