Aggregation of the natively unfolded protein α-synuclein (α-syn) is key to the development of Parkinson’s disease (PD). Some nanoparticles (NPs) can inhibit this process and may pave the way for treatment of PD. Using simulation strategies, we show here that α-syn self-assembly is electrostatically driven. Dimerization by head-to-head monomer contact is triggered by dipole-dipole interactions and subsequently stabilized by van der Waals (vdW) interactions and hydrogen bonds. Therefore, we hypothesized that charged nano-objects can interfere with this process and thus prevent α-syn fibrillation. In our simulations, positively and negatively charged graphenenanosheets or superparamagnetic iron oxide (SPIO) NPs first interacted with α-syn’s N/C terminally charged residues and then with hydrophobic residues in the NAC (non-amyloid-β component (61-95)) region. In the experimental setup, we demonstrated that the charged nano-objects have the capacity not only to strongly inhibit α-syn fibrillation (both nucleation and elongation) but also to disaggregate the mature fibrils. Through the α-syn fibrillation process, the charged nano-objects induced the formation of off-pathway oligomers.