

Figure S1: Schematic representation of the signaling cascade by two redox-regulated nano switches.

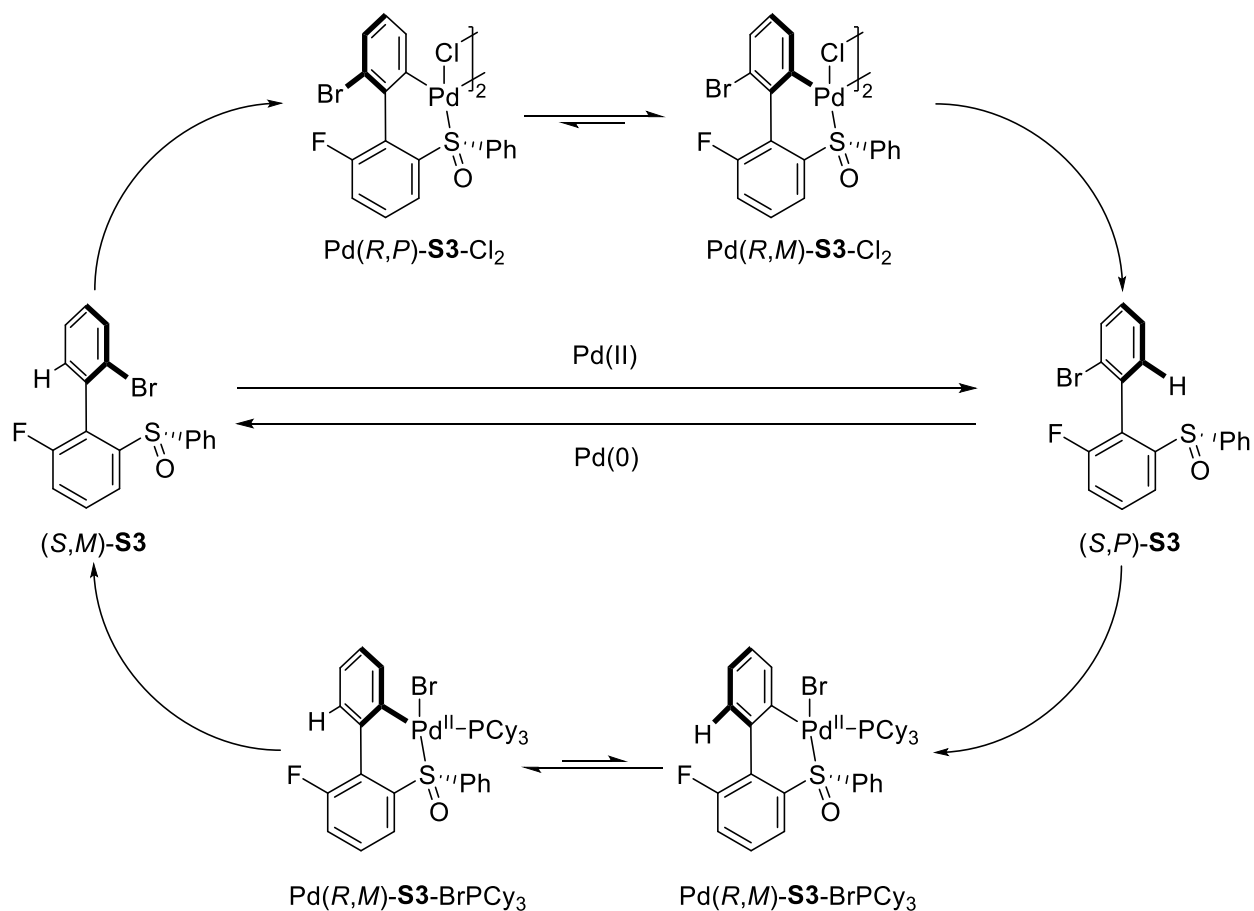


Figure S2: Sequential operation of a Pd-fueled molecular motor. Conditions: (i) $\text{Pd}(\text{OAc})_2$ (1.5 eq.) and TFA (2.0 eq.) in 1,2-dichloroethane (1,2-DCE) at 80 °C for 22 hours, followed by ligand exchange with LiCl (20 eq.) in acetone for 4 hours at room temperature to afford $\text{Pd}(R,M)\text{-S3-Cl}_2$. (ii) *trans,trans*-Dibenzylideneacetone (dba, 2.0 eq.) in 1,2-DCE for 15 minutes followed by sodium triacetoxyborohydride ($\text{NaBH}(\text{OAc})_3$, 3.0 eq.) for an additional 20 minutes at room temperature affords $(S,P)\text{-S3}$. (iii) $\text{Pd}_2(\text{dba})_3$ (0.75 eq.) and tricyclohexylphosphine (PCy_3 , 2.0 eq.) in THF for 22 hours at 40 °C affords $\text{Pd}(R,M)\text{-S3-BrPCy}_3$. (iv) *N*-Bromosuccinamide (NBS, 4.0 eq.) in DCM at room temperature for 22 hours affords $(S,M)\text{-S3}$.

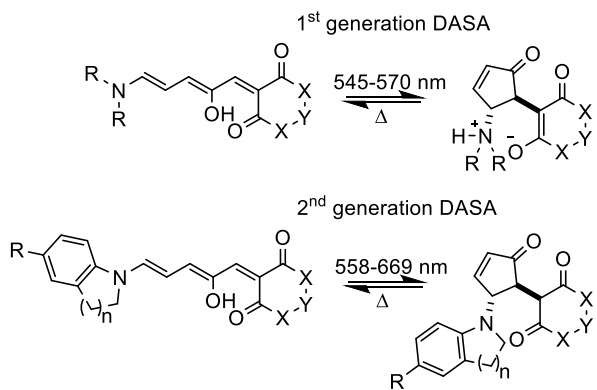


Figure S3: Generalized structures of DASAs with amine (first generation) or aniline (second generation) donors and Meldrum's ($X = O$ and $Y = C(CH_3)_2$) or barbituric ($X = NMe$ and $Y = CO$) acid-based acceptors.