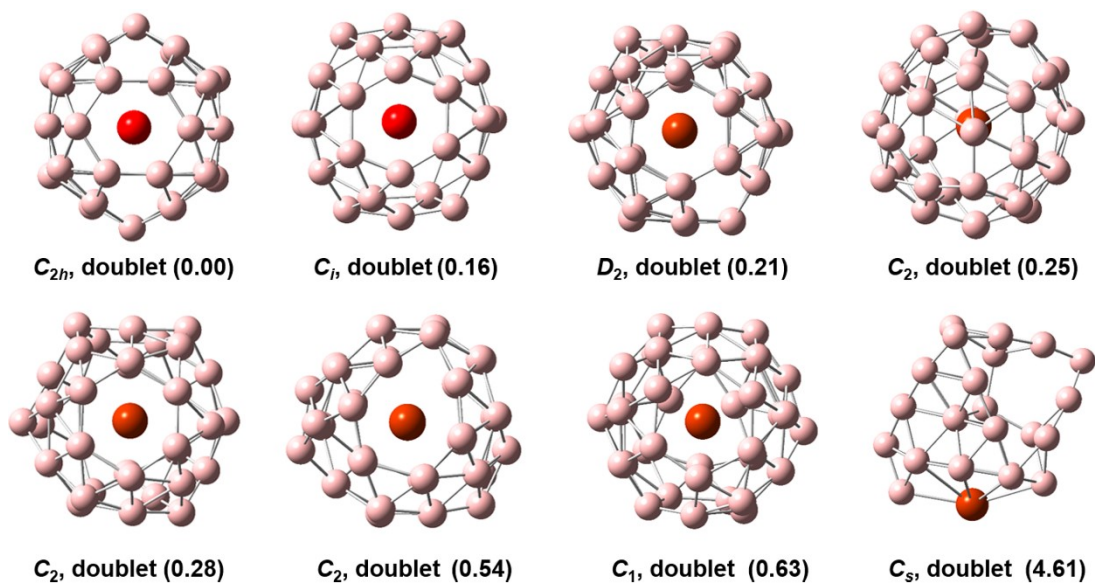# Electronic Supplementary Material (ESI)

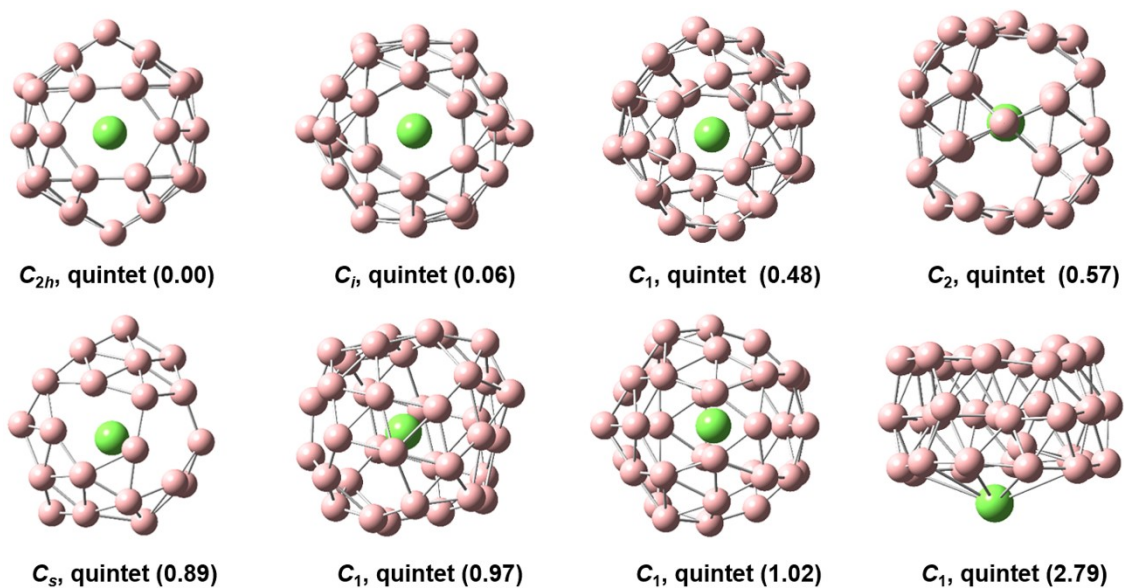**Actinide-doped boron clusters: from borophenes to borospherenes**

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C₂ₕ, doublet (0.00)   **Cᵢ, doublet (0.16)**   **D₂, doublet (0.21)**   **C₂, doublet (0.25)**

C₂, doublet (0.28)   **C₂, doublet (0.54)**   **C₁, doublet (0.63)**   **Cₛ, doublet (4.61)**

Figure S1. Low-lying isomers of Pa@B$_{36}$ with the relative energies (eV) at the PBE0/RECP/6-311+G* level of theory. Light pink and red spheres represent B and Pa, respectively. All the energies have been corrected for zero-point energies.

C₂ₕ, quintet (0.00)   **Cᵢ, quintet (0.06)**   **C₁, quintet (0.48)**   **C₂, quintet (0.57)**

Cₛ, quintet (0.89)   **C₁, quintet (0.97)**   **C₁, quintet (1.02)**   **C₁, quintet (2.79)**

Figure S2. Low-lying isomers of Pu@B$_{36}$ with the relative energies (eV) at the PBE0/RECP/6-311+G* level of theory. Light pink and green spheres represent B and Pu, respectively. All the energies have been corrected for zero-point energies.
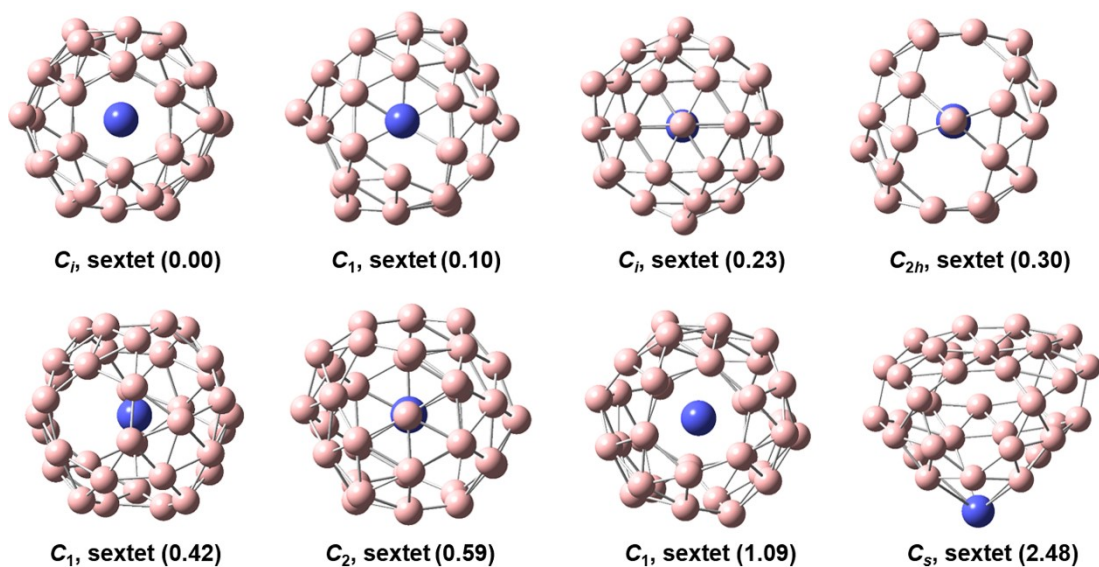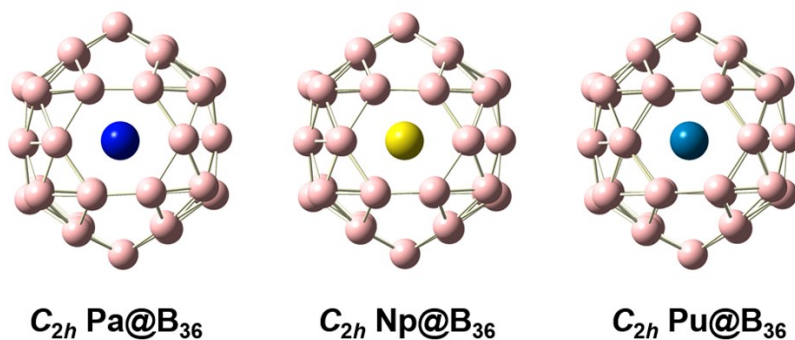
Figure S3. Low-lying isomers of Am@B_{36} with the relative energies (eV) at the PBE0/RECP/6-311+G* level of theory. Light pink and blue spheres represent B and Am, respectively. All the energies have been corrected for zero-point energies.
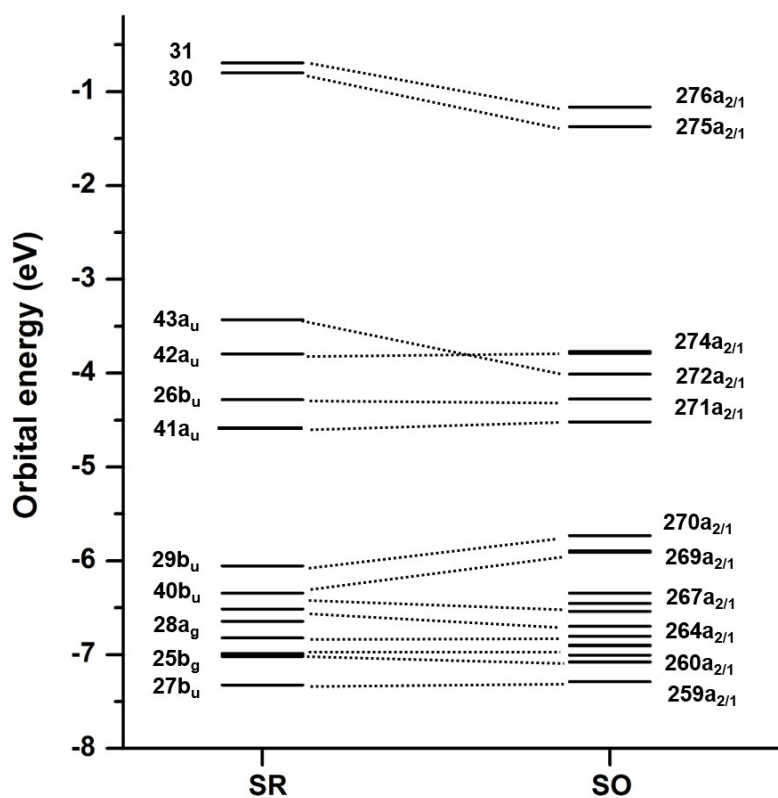

Figure S4. MO energy levels for C_{2h} Pu@B_{36} with scalar relativistic (SR) and spin-orbit (SO) coupling.

C_{2h} Pa@B$_{36}$ C_{2h} Np@B$_{36}$ C_{2h} Pu@B$_{36}$

Figure S5. Optimized structures of An@B$_{36}$ (An= Pa, Np and Pu) at the PBE0-D3/RECP/6-311+G* level of theory. Pink, dark blue, yellow, and blue spheres represent B, Pa, Np, and Pu, respectively.
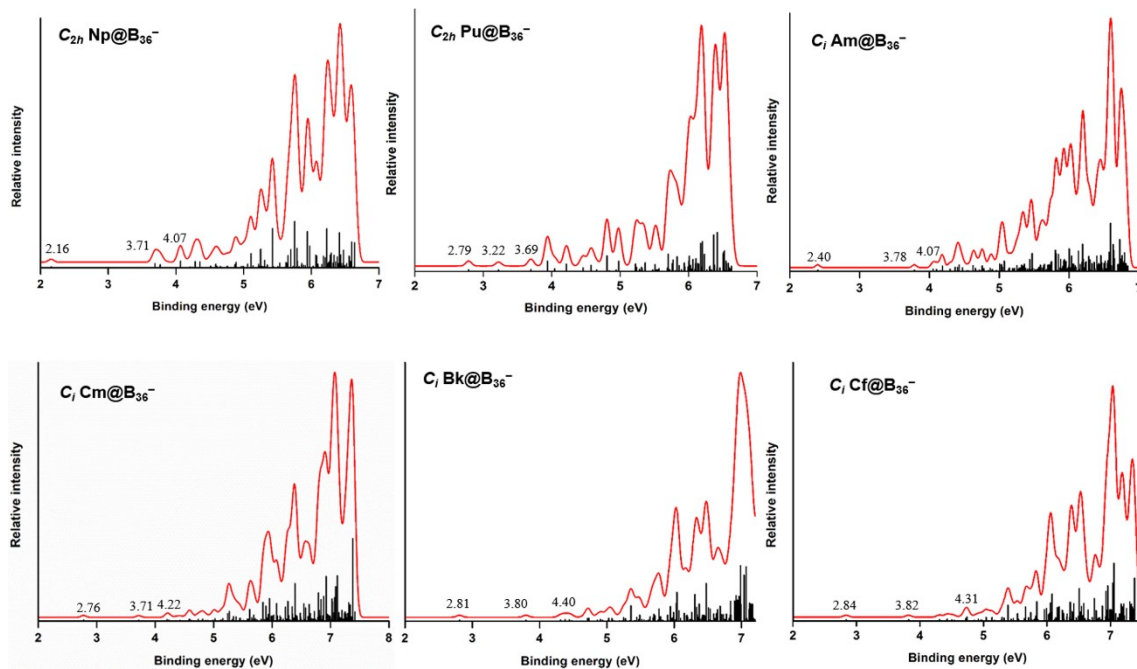
Figure S6. Simulated photoelectron spectrum of An@B$_{36}^-$ (An = Np, Pu, Am, Cm, Bk and Cf) at the PBE0/RECP/6-311+G* level of theory.
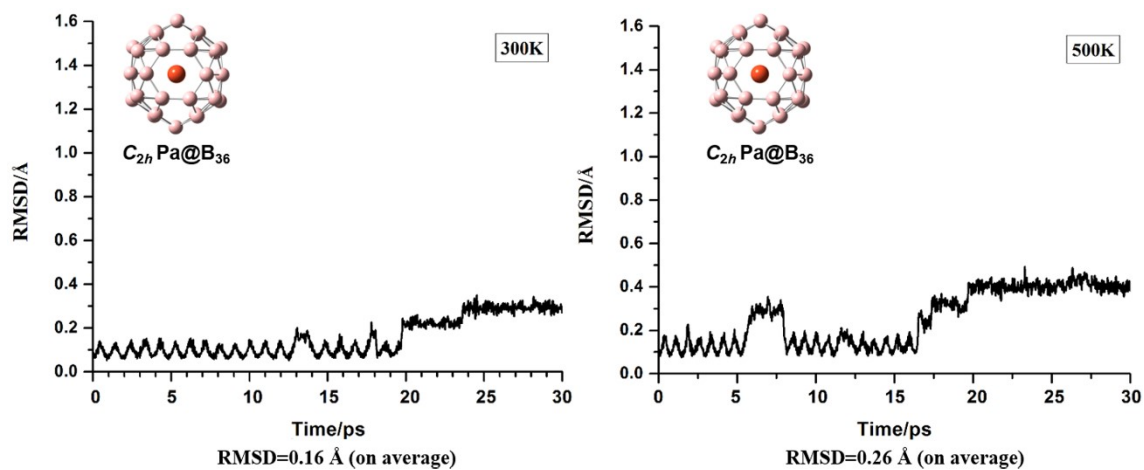
Figure S7. Molecular dynamics simulations of C_{2h} Pa@B$_{36}$ at 300K and 500K for 30 ps.
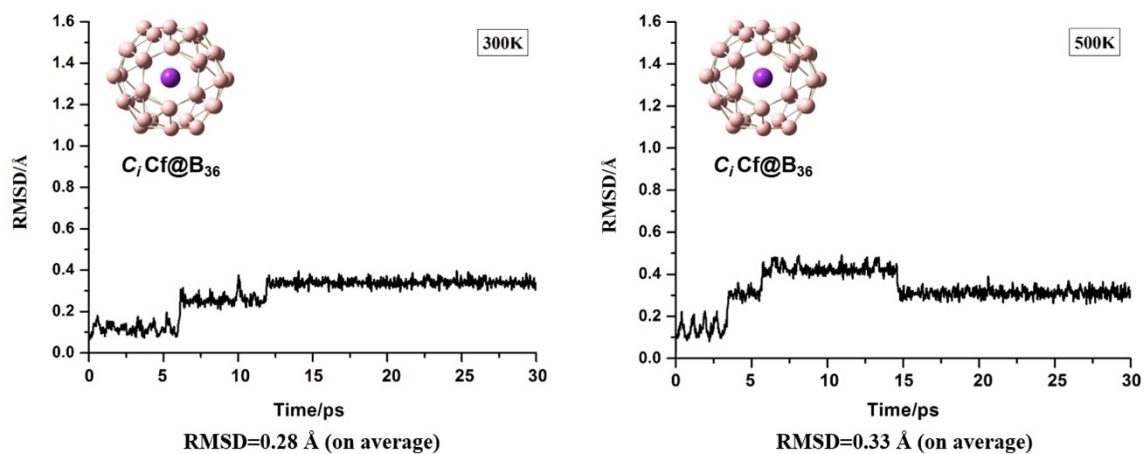
Figure S8. Molecular dynamics simulations of C_i Cf@B$_{36}$ at 300K and 500K for 30 ps.

Table S1. Spin states, spin contamination $\langle S^2 \rangle$, An-B bond distances (Å), and WBIs of the An-B bonds for the An@B$_{36}$ (An= Pa, Np, Pu) complexes at the PBE0-D3/6-311+G*/RECP level of theory.

Species	Spin states	$\langle S^2 \rangle$	An-B Bond Distances	An-B Bond WBIs
C_{2h} Pa@B$_{36}$	doublet	0.76	2.707 - 3.067	0.253-0.435(0.316)
C_{2h} Np@B$_{36}$	quartet	3.87	2.716 - 3.090	0.208-0.382(0.272)
C_{2h} Pu@B$_{36}$	quintet	6.44	2.716 - 3.084	0.186-0.361(0.249)

Table S2. The molecular orbital composition analysis of Pa (%) for Pa@B$_{36}$ at the theoretical level of PBE0/6-311+G*/RECP.

Species	Orbitals	5f	6d
	SOMO	50.16	
	HOMO	23.66	
	HOMO-1	28.54	
	HOMO-2	23.38	
	HOMO-3	14.92	
Pa@B$_{36}$	HOMO-4	14.79	
	HOMO-5		2.11
	HOMO-6		
	HOMO-7	1.15	
	HOMO-8		2.28
	HOMO-9		16.64

Table S3. QTAIM analysis of An@B$_{36}$ (An= Pa, Np, Pu) at the PBE0-D3/6-311+G*/RECP theoretical level.

Species	ρ	H	$\nabla^2\rho$	DI$_{total}$
C_{2h} Pa@B$_{36}$	0.04170	-0.00765	0.07041	6.098
C_{2h} Np@B$_{36}$	0.03852	-0.00648	0.07299	5.771
C_{2h} Pu@B$_{36}$	0.03734	-0.00598	0.07321	5.443

Table S4. The first vertical detachment energies (VDE, eV) and adiabatic detachment energies (ADE, eV) of An@B$_{36}$ at the PBE0/6-311+G*/RECP level of theory.

Species	VDE	ADE
C_{2h} Pa@B$_{36}$	2.42	2.30
C_{2h} Np@B$_{36}$	2.16	2.07
C_{2h} Pu@B$_{36}$	2.79	1.96
C_i Am@B$_{36}$	2.40	2.30
C_i Cm@B$_{36}$	2.77	2.65
C_i Bk@B$_{36}$	2.81	2.69
C_i Cf@B$_{36}$	2.84	2.71

Table S5. Formation energy (ΔE, kcal/mol) of An@B_{36} (An = Pa, Np, Pu, Am, Cm, Bk, Cf) at the PBE0/6-311+G*/RECP level of theory.

Reactions	ΔE
Pa + B_{36} → Pa@B_{36}	-225.1
Np + B_{36} → Np@B_{36}	-157.6
Pu + B_{36} → Pu@B_{36}	-112.9
Am + B_{36} → Am@B_{36}	-75.8
Cm + B_{36} → Cm@B_{36}	-88.2
Bk + B_{36} → Bk@B_{36}	-80.0
Cf + B_{36} → Cf@B_{36}	-67.3

Table S6. The formation energy (ΔE, kcal/mol) of An@B_{36} (An= Pa, Np, Pu) at the PBE0-D3/6-311+G*/RECP level of theory.

Reactions	ΔE
Pa + B_{36} = Pa@B_{36}	-292.1
Np + B_{36} = Np@B_{36}	-245.5
Pu + B_{36} = Pu@B_{36}	-211.9