Composition	Structural motifs predicted at the Gupta potential level	Structural motifs predicted at the DFT level
$Pd_{17}Pt_{17}$	Marks Dh < low-symmetry pIh	Dh-cp(DT) < Marks Dh < low-symmetry pIh
$Pd_{18}Pt_{16}$	low-symmetry pIh < Marks Dh	Dh-cp(DT) < low-symmetry pIh
$Pd_{19}Pt_{15}$	Marks Dh < Dh-cp(DT) < TO < low-symmetry pIh	Dh-cp(DT) < Marks Dh
$Pd_{20}Pt_{14}$	incomplete 5-fold pIh < Dh-cp(DT) < TO < Marks Dh < Dh-cp(T) < low-symmetry pIh	Dh-cp(DT) < incomplete 5-fold pIh
$Pd_{21}Pt_{13}$	incomplete 5-fold pIh < TO < Marks Dh < Dh-cp(T) < low-symmetry pIh	Dh-cp(DT) < TO < incomplete 5-fold pIh
$Pd_{22}Pt_{12}$	Dh-cp(DT) < Marks Dh < TO < Dh-cp(T)	Dh-cp(DT)
$Pd_{23}Pt_{11}$	Dh-cp(T) < Dh-cp(DT) < Marks Dh < TO < low-symmetry pIh	Dh-cp(DT) < TO < Dh-cp(T)
$Pd_{24}Pt_{10}$	Dh-cp(T) < Dh-cp(DT) < TO < Marks Dh < low-symmetry pIh	Dh-cp(DT) < TO < low-symmetry pIh < Dh-cp(T)
$Pd_{25}Pt_{9}$	Dh-cp(T) < TO < Marks Dh < Dh-cp (DT)	Dh-cp(DT) < Dh-cp(T) < TO
$Pd_{26}Pt_{8}$	incomplete 6-fold pIh < TO < Marks Dh < Dh-cp(DT) < Dh-cp(T)	Dh-cp(DT) < TO
$Pd_{27}Pt_{7}$	incomplete 6-fold pIh < Marks Dh < TO < Dh-cp(T) < Dh-cp(DT)	Dh-cp(DT) < incomplete 6-fold pIh
$Pd_{28}Pt_{6}$	incomplete 6-fold pIh < TO < Dh-cp(T)	Dh-cp(DT) < incomplete 6-fold pIh
$Pd_{29}Pt_{5}$	incomplete 6-fold pIh < incomplete distorted 5-fold pIh < low-symmetry pIh	incomplete 6-fold pIh
$Pd_{30}Pt_{4}$	incomplete 6-fold pIh	incomplete 6-fold pIh

Table 1. Ordering of structural isomers of 34-atom PdPt clusters as a function of composition, for Gupta and DFT calculations.

Structure	Lowest energy motif (Gupta potential)	Symmetry (Gupta level)	Gupta potential total energy (eV)	Lowest energy motif (DFT)	Symmetry (DFT)	DFT PW91 Total energies (A.U) S=0 ; σ=0.01	DFT PW91 Total energies (A.U) S > 0 ; σ=0.005
Pd_{34}	incomplete decahedral	C_2	-116.967894	incomplete decahedral	C_2	-4352.492034156192	-4352.444876253685
$Pd_{30}Pt_4$	incomplete 6-fold pIh	D_{2h}	-127.968157	incomplete 6-fold pIh	D_{2h}	-4318.443049910242	-4318.397471476259
$Pd_{29}Pt_5$	incomplete 6-fold pIh	C_{2v}	-130.138409	incomplete 6-fold pIh	C_{2v}	-4309.934855082618	-4309.889122950146
$Pd_{28}Pt_6$	incomplete 6-fold pIh	D_{2h}	-132.302592	Dh-cp(DT)	C_s	-4301.444036731449	-4301.403614709368
$Pd_{27}Pt_7$	incomplete 6-fold pIh	C_{2v}	-134.315159	Dh-cp(DT)	C_s	-4292.938769106509	-4292.898805772955
$Pd_{26}Pt_8$	TO	C_{2v}	-136.284248	Dh-cp(DT)	C_s	-4284.431829852023	-4284.391822635112
$Pd_{25}Pt_9$	Dh-cp(T)	C_{3v}	-138.315822	Dh-cp(DT)	C_s	-4275.921890940306	-4275.882165266052
$Pd_{24}Pt_{10}$	Dh-cp(T)	T_d	-140.434769	Dh-cp(DT)	C_s	-4267.408526578860	-4267.369129019902
$Pd_{23}Pt_{11}$	Dh-cp(T)	C_s	-142.164285	Dh-cp(DT)	C_s	-4258.891008762675	-4258.851121347474
$Pd_{22}Pt_{12}$	Dh-cp(DT)	C_s	-144.001080	Dh-cp(DT)	C_s	-4250.378179742616	-4250.337835326187
$Pd_{21}Pt_{13}$	incomplete 5-fold pIh	C_{5v}	-145.922977	Dh-cp(DT)	C_s	-4241.858504438187	-4241.818391393768
$Pd_{20}Pt_{14}$	incomplete 5-fold pIh	C_s	-147.543196	Dh-cp(DT)	C_s	-4233.340608207724	-4233.300170211292
$Pd_{19}Pt_{15}$	Marks Dh	C_1	-149.179717	Dh-cp(DT)	C_s	-4224.823966556000	-4224.784850225449
$Pd_{18}Pt_{16}$	low-symmetry pIh	C_s	-150.885159	Dh-cp(DT)	C_s	-4216.303286573002	-4216.264630390280
$Pd_{17}Pt_{17}$	Marks Dh	C_1	-152.560451	Dh-cp(DT)	C_s	-4207.786361388469	-4207.747954974639
Pt_{34}	incomplete decahedral	C_2	-177.876096	incomplete decahedral	C_2	-4062.930477842208	-4062.890479334899

Table 2. Comparision of global minimum structures for 34-atom PdPt clusters for the Gupta potential and DFT calculations. Total DFT energies are in atomic units (A.U), while Gupta potential energies are given in eV.

Structure	Lowest energy structural motif (Gupta potential)	Symmetry	Gupta potential total energy (eV)
$Pd_{33}Pt_1$	Marks Dh	C_s	-119.702171
$Pd_{32}Pt_2$	low-symmetry pIh	C_1	-122.470613
$Pd_{31}Pt_3$	low-symmetry pIh	C_1	-125.288139
$Pd_{16}Pt_{18}$	low-symmetry pIh	C_1	-154.101708
$Pd_{15}Pt_{19}$	Marks Dh	C_s	-155.890715
$Pd_{14}Pt_{20}$	Marks Dh	C_1	-157.511383
$Pd_{13}Pt_{21}$	low-symmetry pIh	C_1	-158.722074
$Pd_{12}Pt_{22}$	Marks Dh	C_1	-160.610847
$Pd_{11}Pt_{23}$	Marks Dh	C_s	-162.111451
$Pd_{10}Pt_{24}$	Marks Dh	C_1	-163.580682
Pd_9Pt_{25}	Marks Dh	C_1	-165.033719
Pd_8Pt_{26}	Marks Dh	C_1	-166.362200
Pd_7Pt_{27}	Marks Dh	C_1	-167.839772
Pd_6Pt_{28}	Marks Dh	C_1	-169.335320
Pd_5Pt_{29}	Marks Dh	C_1	-170.783213
Pd_4Pt_{30}	Marks Dh	C_1	-172.222597
Pd_3Pt_{31}	Marks Dh	C_1	-173.652960
Pd_2Pt_{32}	Marks Dh	C_1	-175.061020
Pd_1Pt_{33}	incomplete decahedral	C_1	-176.481095

Table 3. PdPt cluster composition range not analyzed in this work at the DFT level.