Supporting Information

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Table S1 E_fs (eV) of certain configurations observed in the NEMD simulation of V_ns, for n = 4, 6, and 8. The optimizations were performed with both REBO2 potential and DFT (in parentheses). The 5-, rotated 6-, 7-, and 8- membered rings and hole-structures are emphasized with yellow, purple, green, red, and dark purple colors, respectively. The haeckelites are distinguished by merged (M-) or separated (S-) with the total number of 5|7 pairs (see the main text).

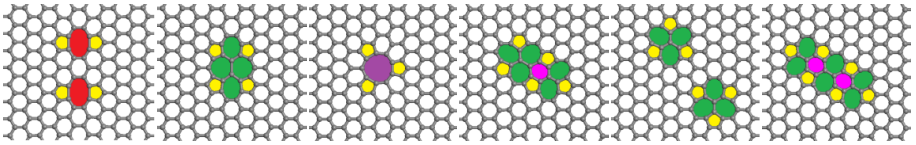
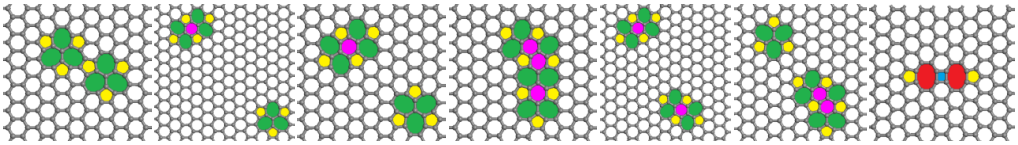
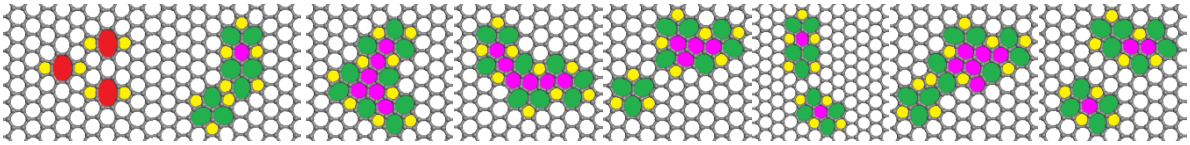
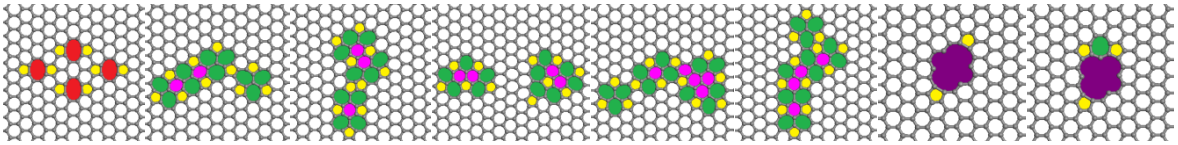
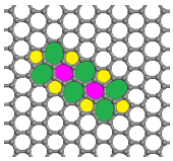
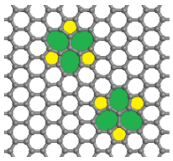
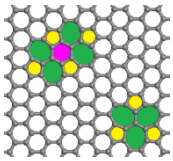
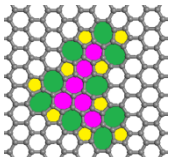
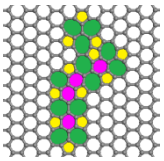
V_ns	Representative configurations and E_fs
V_4	 2-585 M-4-57 5559 M-5-57 S-6-57 M-6-57 16.51(16.15) 13.01(11.77) 10.80(10.28) 11.70(10.64) 13.22(12.27) 11.36(9.96)  S-6-57 S-7-57 S-7-57 M-7-57 S-8-57 S-8-57 58-4-85 12.89(12.09) 13.98(12.59) 14.09(12.72) 13.28(10.65) 14.45(12.50) 15.25(13.43) 12.66(11.36)
V_6	 3-585 S-8-57 M-9-57 M-9-57 S-9-57 S-9-57 S-10-57 S-10-57 22.74(21.86) 17.40(15.60) 16.85(14.49) 18.79(16.17) 19.16(17.10) 19.69(17.69) 20.18(17.81) 20.79(18.22)
V_8	 4-585 S-10-57 S-10-57 S-11-57 S-12 -57 S-12-57 hole hole 29.23 (27.65) 22.86 (20.77) 23.71(21.24) 25.64 (23.26) 24.63(21.74) 17.18(18.27) 17.52(18.28) 17.51(18.31)

Table S2 The area of pentagon and heptagon of the 57 pair of certain haeckelites, and the resulted area compensation by 57 pair relative to two hexagons in the perfect graphene.

MVs	V_4			V_6	V_8
Configurations					
Average area of pentagons ($Å^2$)	3.59	3.61	3.62	3.56	3.66
Average area of heptagons ($Å^2$)	7.76	7.87	7.75	7.67	7.72
$S_5 + S_7 = n\ S_6$	2.15	2.18	2.16	2.13	2.16
Compensation(%)	45	54	56	39	48

Note: In the perfect graphene, C-C length is 1.424 Å, and the area of a hexagon is 5.27 $Å^2$.

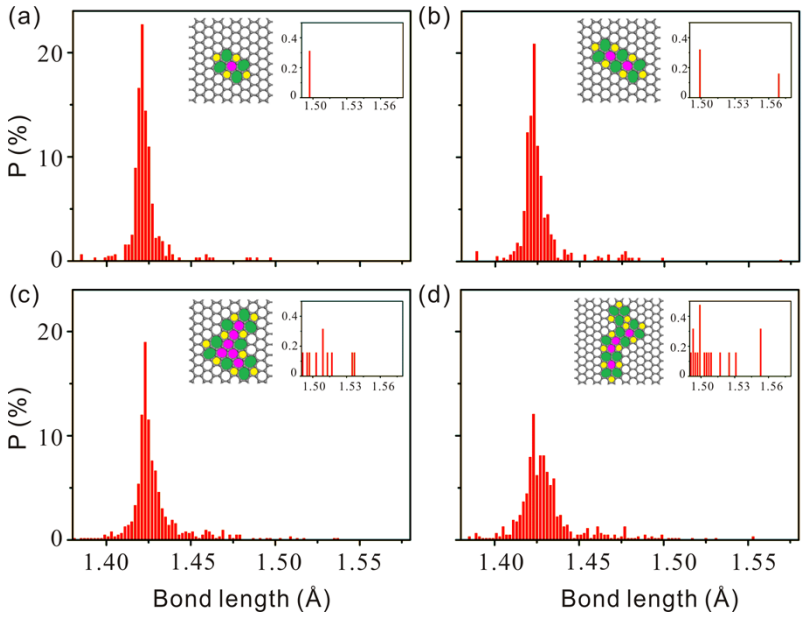


Fig. S1 Probability distributions of bond lengths in representative configurations of V_ns: V_2 (a), V_4 (b), V_6 (c), and V_8 (d). The configurations and partial enlarged regions with bond length exceeds 1.49 Å are also shown as insets.