Electronic Supplementary Information

1. The detailed atomic position of the predicted structures

A: CrN_4C_6

Primitive Cell

1.0

4.8384070396	0.0000000000	0.0000000000
-0.5138389787	6.5830143660	0.0000000000
0.0000000000	0.0000000000	16.0000000000

C	N	Cr
6	4	1

Direct

0.025575197	0.222872716	0.500000000
0.738839859	0.896706653	0.500000000
0.240954106	0.887305890	0.500000000
0.974424750	0.777127266	0.500000000
0.759045919	0.112694164	0.500000000
0.261160157	0.103293320	0.500000000
0.903438055	0.568670096	0.500000000
0.517620695	0.207704449	0.500000000
0.096562004	0.431329958	0.500000000
0.482379307	0.792295624	0.500000000
0.499999973	0.500000009	0.500000000

B: MnN_4C_6

Primitive Cell

1.0

4.8111515045	0.0000000000	0.0000000000
-0.5504593605	6.5168075854	0.0000000000
0.0000000000	0.0000000000	16.0000000000

C	N	Mn
6	4	1

Direct

0.028977453	0.222293841	0.500000000
0.737650496	0.897008616	0.500000000
0.240178036	0.885442650	0.500000000
0.971022538	0.777706173	0.500000000
0.759821945	0.114557336	0.500000000
0.262349497	0.102991361	0.500000000
0.898319768	0.566788000	0.500000000
0.519523257	0.213142991	0.500000000
0.101680301	0.433211995	0.500000000
0.480476688	0.786857005	0.500000000

0.499999997 0.499999998 0.500000000

C: FeN_4C_6

Primitive Cell

1.0

4.7858681679 0.0000000000 0.0000000000

-0.5439361689 6.4794524632 0.0000000000

0.0000000000 0.0000000000 16.0000000000

C N Fe

6 4 1

Direct

0.029100483 0.223348719 0.500000000

0.736636379 0.896582450 0.500000000

0.241313292 0.884277466 0.500000000

0.970899499 0.776651303 0.500000000

0.758686666 0.115722538 0.500000000

0.263363698 0.103417499 0.500000000

0.896762494 0.565546449 0.500000000

0.519144356 0.215724899 0.500000000

0.103237619 0.434453610 0.500000000

0.480855650 0.784275087 0.500000000

0.499999993 0.500000011 0.500000000

D: CoN_4C_6

Primitive Cell

1.0

4.7636513710 0.0000000000 0.0000000000

-0.5611254759 6.4265851910 0.0000000000

0.0000000000 0.0000000000 16.0000000000

C N Co

6 4 1

Direct

0.030136787 0.223934404 0.500000000

0.735221943 0.895952121 0.500000000

0.241838696 0.882962942 0.500000000

0.969863281 0.776065578 0.500000000

0.758161318 0.117037040 0.500000000

0.264778068 0.104047833 0.500000000

0.895903539 0.564242318 0.500000000

0.519772064 0.218594477 0.500000000

0.104096473 0.435757757 0.500000000

0.480227925 0.781405505 0.500000000

0.500000021 0.500000001 0.500000000

2. Four configurations of MN_4C_6 layer.

In a graphene sheet, we choose two neighboring benzene rings, and there are only two basic types of configurations, i.e., gray and blue circles in Fig. S1 (a) and gray and red circles in Fig. S1 (b). There, the dashed circles are the equivalent circles to the blue or red solid circle. In Fig. S1 (c) and (d), the structural fraction enclosed by a solid rectangle would be replaced by MN4 moiety. Then, after the replacement, two basic structural patterns are formed and shown in Fig. S1 (e) and (f).

Based on the two basic structural patterns, four kinds of MN_4C_6 monolayers are built, displayed in the Fig. S2 (a) – (d), in which the structure in Fig. S2 (b) is adopted in our manuscript. We take MnN4C6 as an example to calculate their energies and they are -95.2116 eV, **-96.5271 eV**, -96.0484 eV, -95.8902 eV per formula cell four the four kinds of MN_4C_6 (M = Mn) in Fig. S2 (a)-(d), respectively. This demonstrates that the MN_4C_6 (M = Mn) structure in the manuscript is the lowest energy structure.

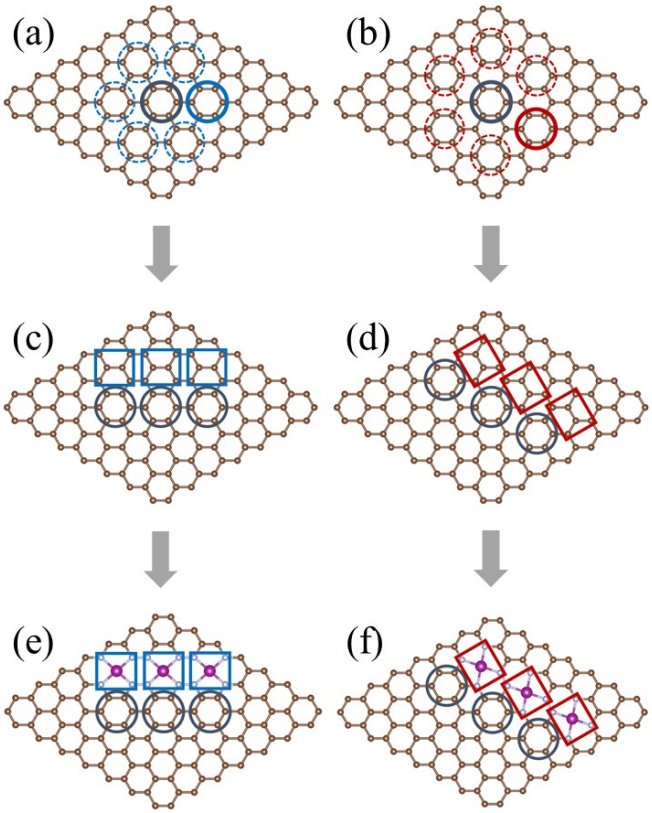

Fig. S1 Flow Chart to form the two basic patterns of arrangement for benzene ring and MN_4 moieties in the graphene sheet.

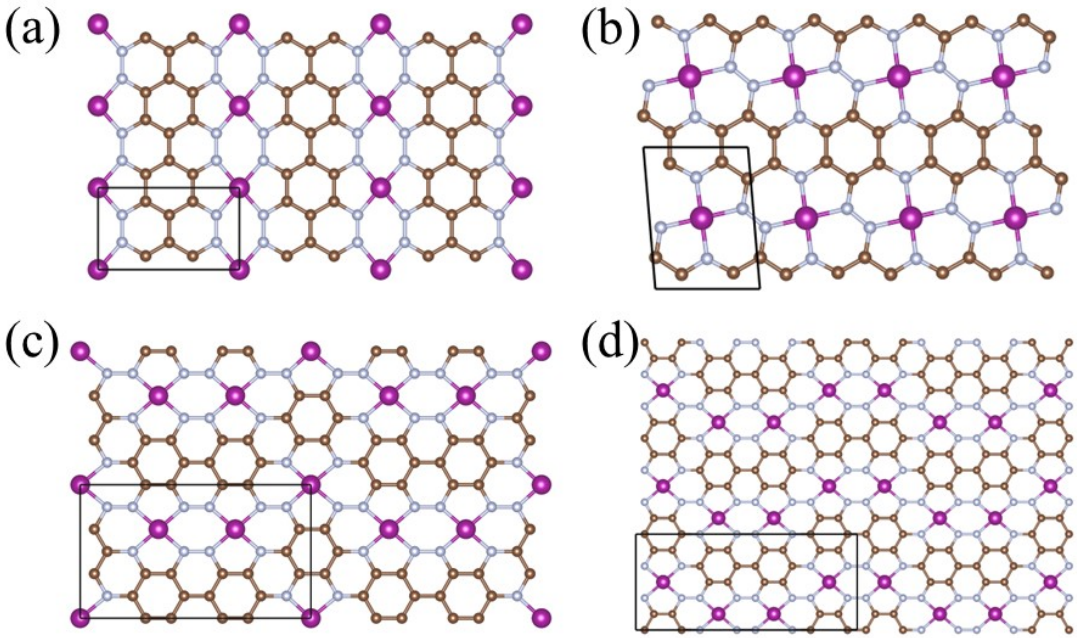

Fig. S2 Four kinds of MN_4C_6 configurations composed of MN_4 and benzene ring moieties, derived from the two basic structural patterns in Fig. S1(e) and (f). The unit cells are marked by the solid lines.