Supplementary Information for Achieving Ultrahigh Energy Density and Excellent Stability in Carbon Pentazole

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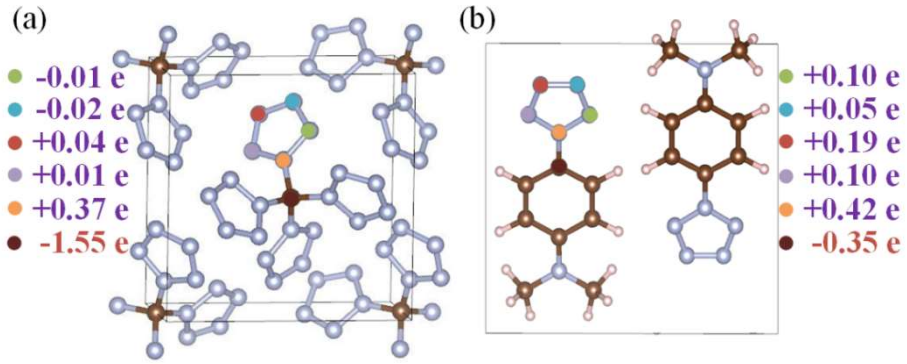

Fig. S1 The atomic Bader charge transfer of (a) the $P\bar{4}2_1c$-$C(N_5)_4$ and (b) the p-dimethylaminophenylpentazole at 35 GPa.

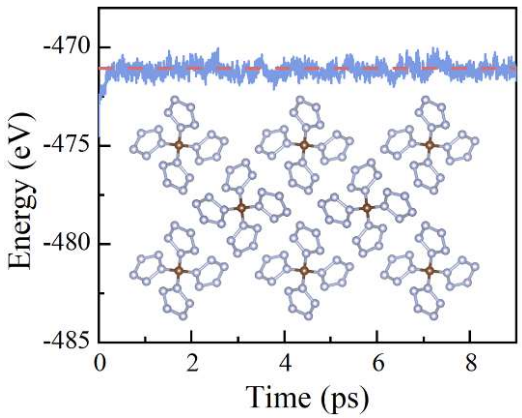

Fig. S2 Fluctuations of the total energies of $P\bar{4}2_1c$-$C(N_5)_4$ in AIMD simulations at 35 GPa and 300 K, the inserted images is the structures at the end of simulations.

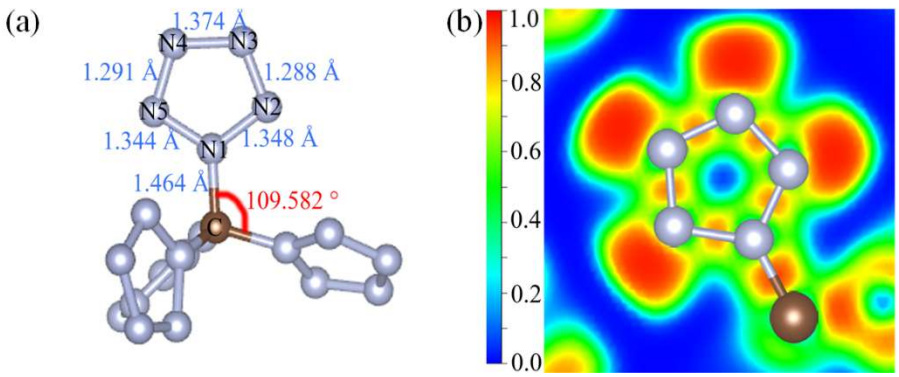

Fig. S3 Under atmospheric pressure, (a) the bond length and bond angle of C-N and N-N bonds, (b) sectional view of ELF along the C and the *cyclo*-N_5^- plane of $P\bar{4}2_1c$-$C(N_5)_4$.

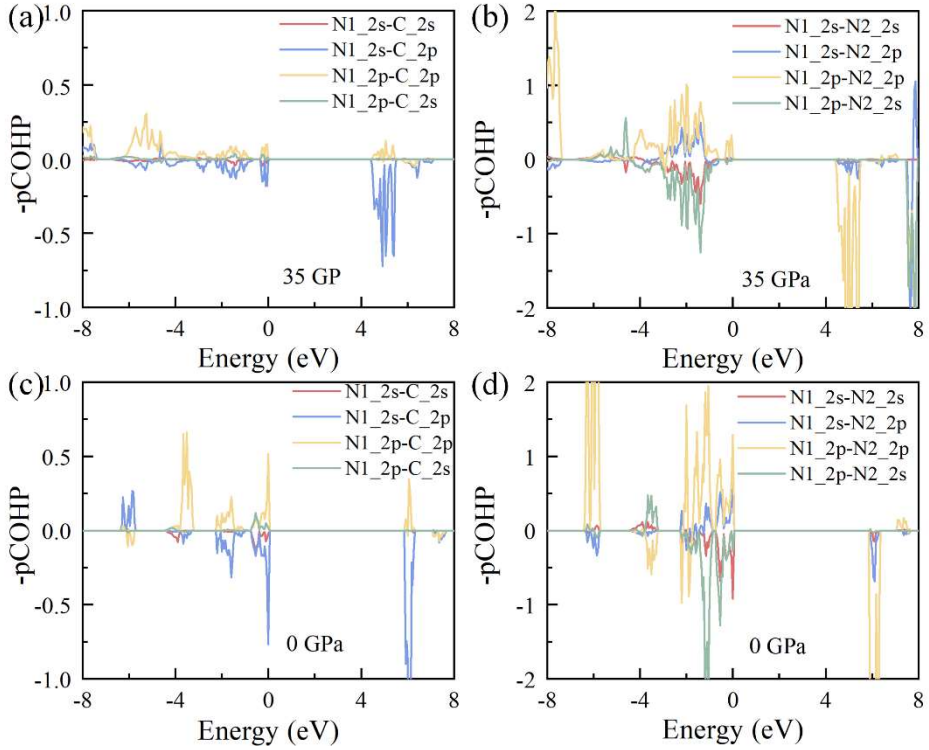

Fig. S4 The -pCOHP of (a) C-N1 and (b) N1-N2 in the $P\bar{4}2_1c$-$C(N_5)_4$ under 35GPa, the -pCOHP of (c) C-N1 and (d) N1-N2 in the $P\bar{4}2_1c$-$C(N_5)_4$ under atmospheric pressure.

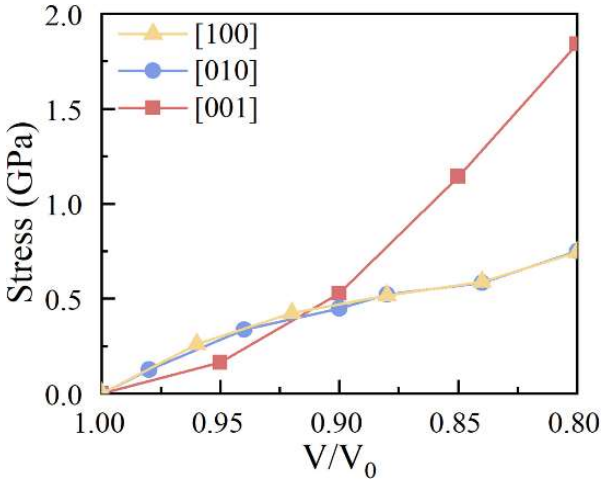

Fig. S5 Principal stress as function of compression ratio V/V_0 under uniaxial loadings. The results indicate that $P\bar{4}2_1c$-$C(N_5)_4$ possesses higher volume compression ratio (20%) than that of the known 1-diamino-2,2-dinitroethylene (FOX-7) (~8%).[1]

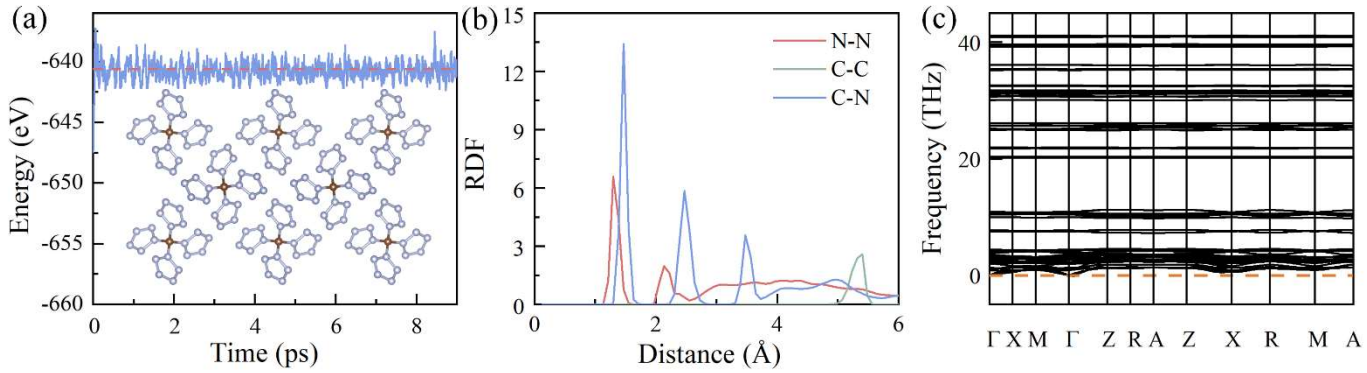

Fig. S6 Under atmospheric pressure, (a) evolution of total energy as a function of time at 600 K, and snapshots of the final structures of $P\bar{4}2_1c$-$C(N_5)_4$; (b) RDF during AIMD simulations at 600 K, (c) phonon spectra of $P\bar{4}2_1c$-$C(N_5)_4$.

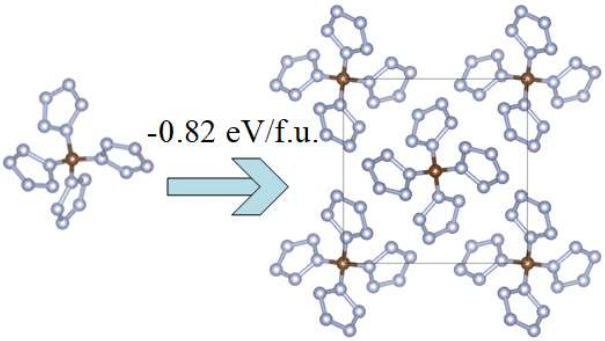

Fig. S7 Assembling the $C(N_5)_4$ single molecules to $P\bar{4}2_1c$-$C(N_5)_4$ crystal will release energy of 0.82 eV/f.u..

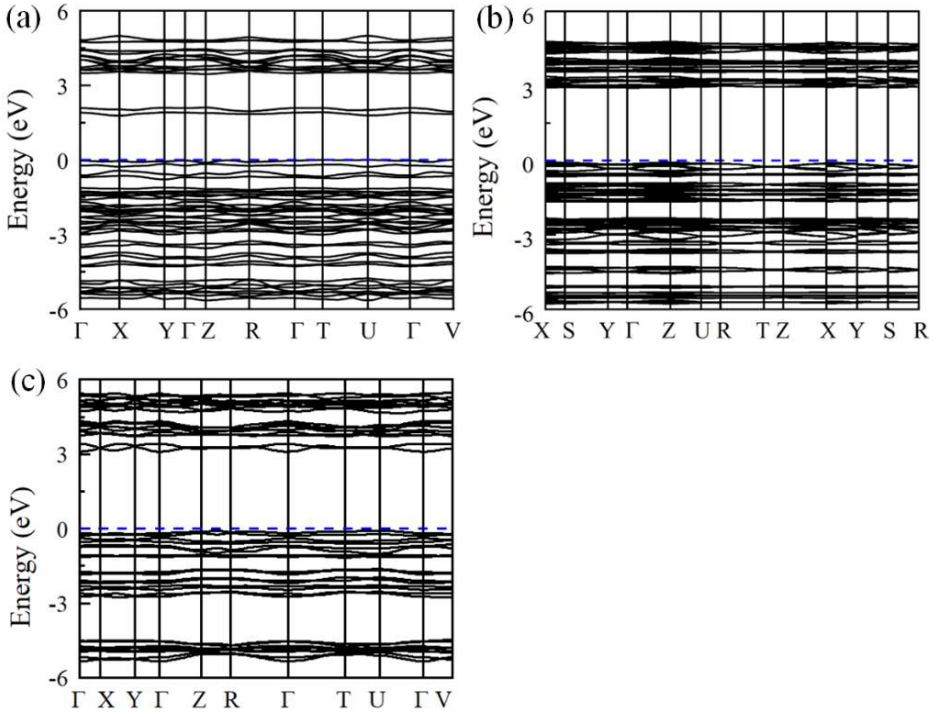

Fig. S8 Under atmospheric pressure, the band structures of (a) $P\bar{1}$-C_2N_{16}, (b) *Pbcn*-C_2N_{14} and (c) $P\bar{1}$-$C(N_3)_4$, the calculations are at HSE06 level.

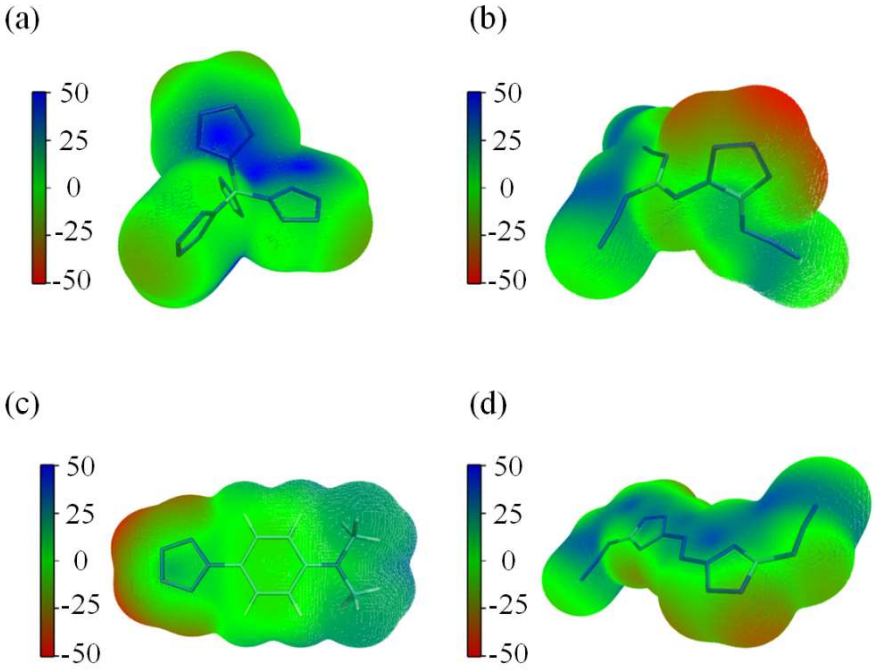

Fig. S9 Electrostatic potential (ESP) of $P\bar{4}2_1c$-$C(N_5)_4$, *Pbcn*-C_2N_{14}, $P\bar{1}$-C_2N_{16} and $P2_1/m$-$C_8N_6H_{10}$, the unit of error bar is kcal/mol.

Tab. S1 Heat of formation (HOF) of CN compounds, the known explosives of TNT and HMX.

Compounds	HOF (kJ/mol)
$P\bar{4}2_1c$-$C(N_5)_4$	2006.69
$P\bar{1}$-C_2N_{16}	1700.70
Pbcn-C_2N_{14}	1495.00
$P\bar{1}$-$C(N_3)_4$	1293.20
TNT	-63.12
HMX	102.41

References

1. Y. Su, J. Fan, Z. Zheng and J. Zhao, *Prog. Nat. Sci.*, 2019, **29**, 329-334.