Supporting Information

For

Pressure-induced structure, elasticity, intrinsic hardness and ideal strength of tetragonal C_4N

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The convergence tests results of pseudo-potential plane wave cutoff energy and k-points grids of tetragonal C_4N (t-C_4N) unit cell are shown in Fig. S1. In Fig. S1(a), structural energy of t-C_4N tends to a fixed value with E_{cut} over 800 eV, which indicates the test attains convergence and E_{cut} is set as 800 eV in following calculations. In Fig. S1(b), structural energy of t-C_4N tends to a fixed value when k-points grids are more than 8×8×8, which indicates k-points test attains convergence and 8×8×8 k-points grids are used in following calculations.

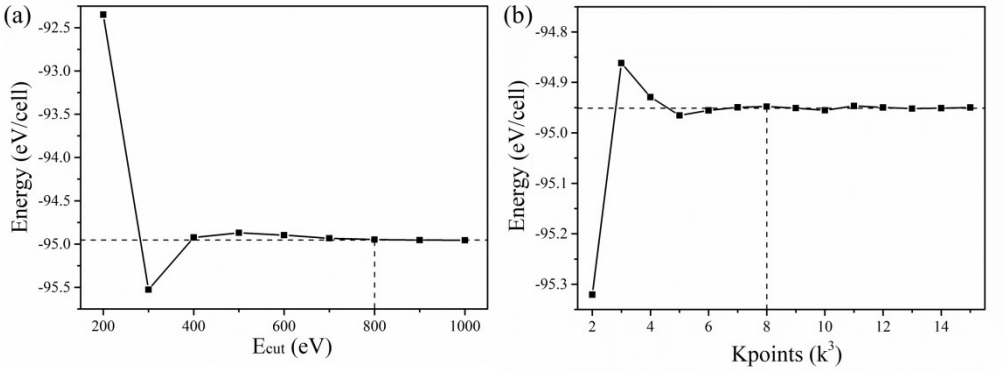

Fig. S1. Convergence tests of (a) cut-off energy and (b) Kpoints.

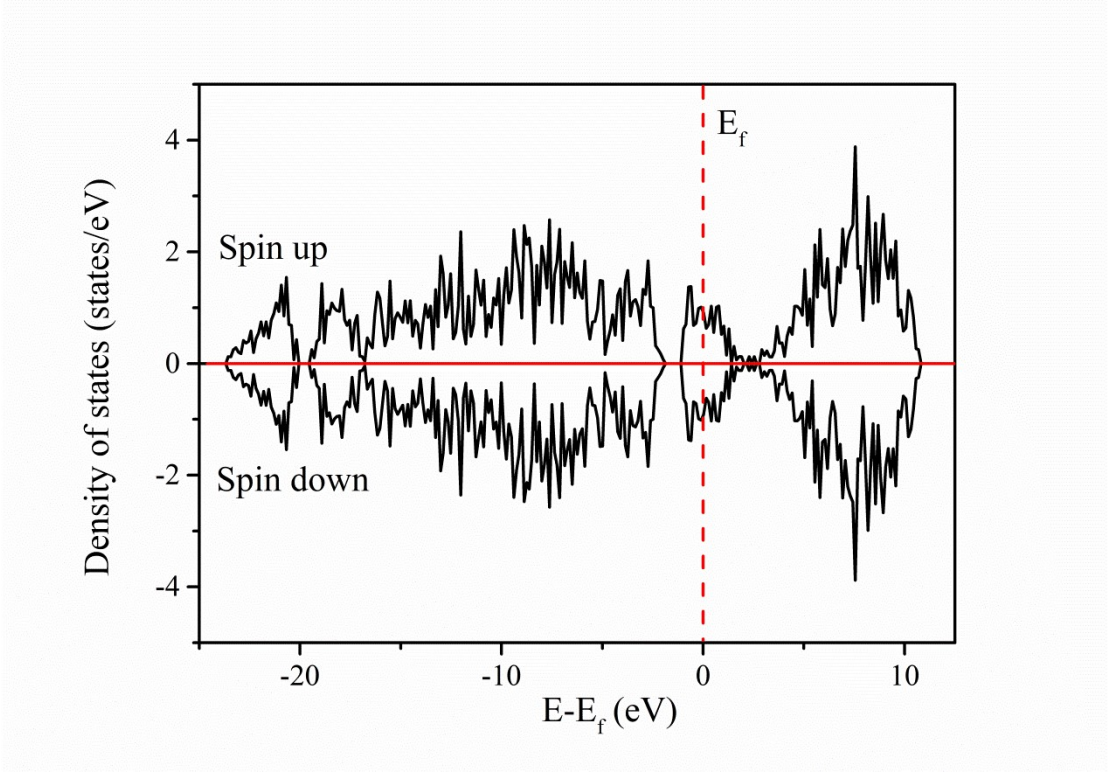

Fig. S2. Calculated spin-polarized density of states of t-C_4N. The red dotted line represents the Fermi level position.

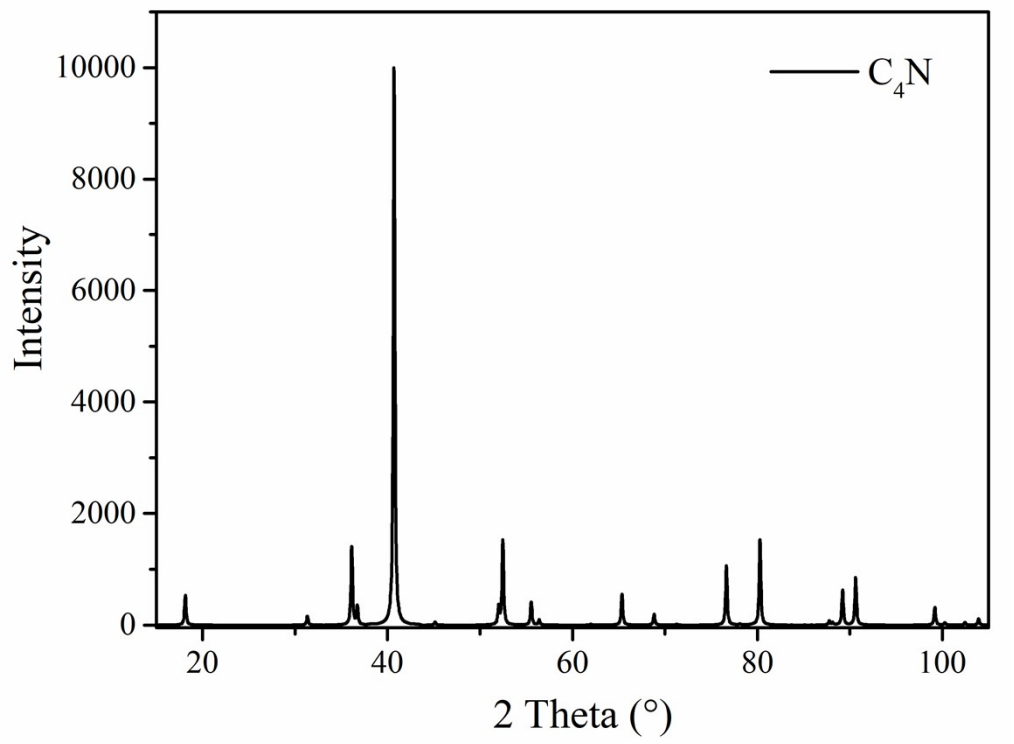

Fig. S3. Theoretical XRD (X-ray diffraction) spectrum simulation.

Table S1. Atomic coordinates and corresponding Wyckoff sites in Diamond ($Fd\bar{3}m$).

Atom type	Atomic number	Symmetric multiplicity	Wyckoff symbol	Fractional coordinates		
				x	y	z
C	C1	8	a	0.00000	0.00000	0.00000
	C2			0.00000	0.50000	0.50000
	C3			0.50000	0.00000	0.50000
	C4			0.50000	0.50000	0.00000
	C5			0.75000	0.25000	0.75000
	C6			0.25000	0.25000	0.25000
	C7			0.25000	0.75000	0.75000
	C8			0.75000	0.75000	0.25000

Table S2. Atomic coordinates and corresponding Wyckoff sites in t-C_4N ($P\bar{4}2_1m$).

Atom type	Atomic number	Symmetric multiplicity	Wyckoff symbol	Fractional coordinates		
				x	y	z
C	C1	2	a	0.00000	0.00000	0.00000
	C2			0.50000	0.50000	0.00000
	C3	2	c	0.50000	0.00000	0.64076
	C4			0.00000	0.50000	0.35924
	C5	4	e	0.24877	0.25123	0.81166
	C6			0.75123	0.74877	0.81166
	C7			0.74877	0.24877	0.18834
	C8			0.25123	0.75123	0.18834
N	N1	2	c	0.50000	0.00000	0.35759
	N2			0.00000	0.50000	0.64240

Table S3. Bond length of t-C_4N and diamond.

t-C_4N		Diamond	
Bond-type	Bond-length (Å)	Bond-type	Bond-length (Å)
C3-N1; C4-N2	1.38		
C5-N2; C6-N2 C7-N1; C8-N1	1.48		
C3-C5; C3-C6 C4-C8; C4-C7	1.51	All C-C	1.54
C1-C5; C1-C6; C1-C7 C1-C8; C2-C6; C2-C7 C2-C8; C2-C5	1.55		

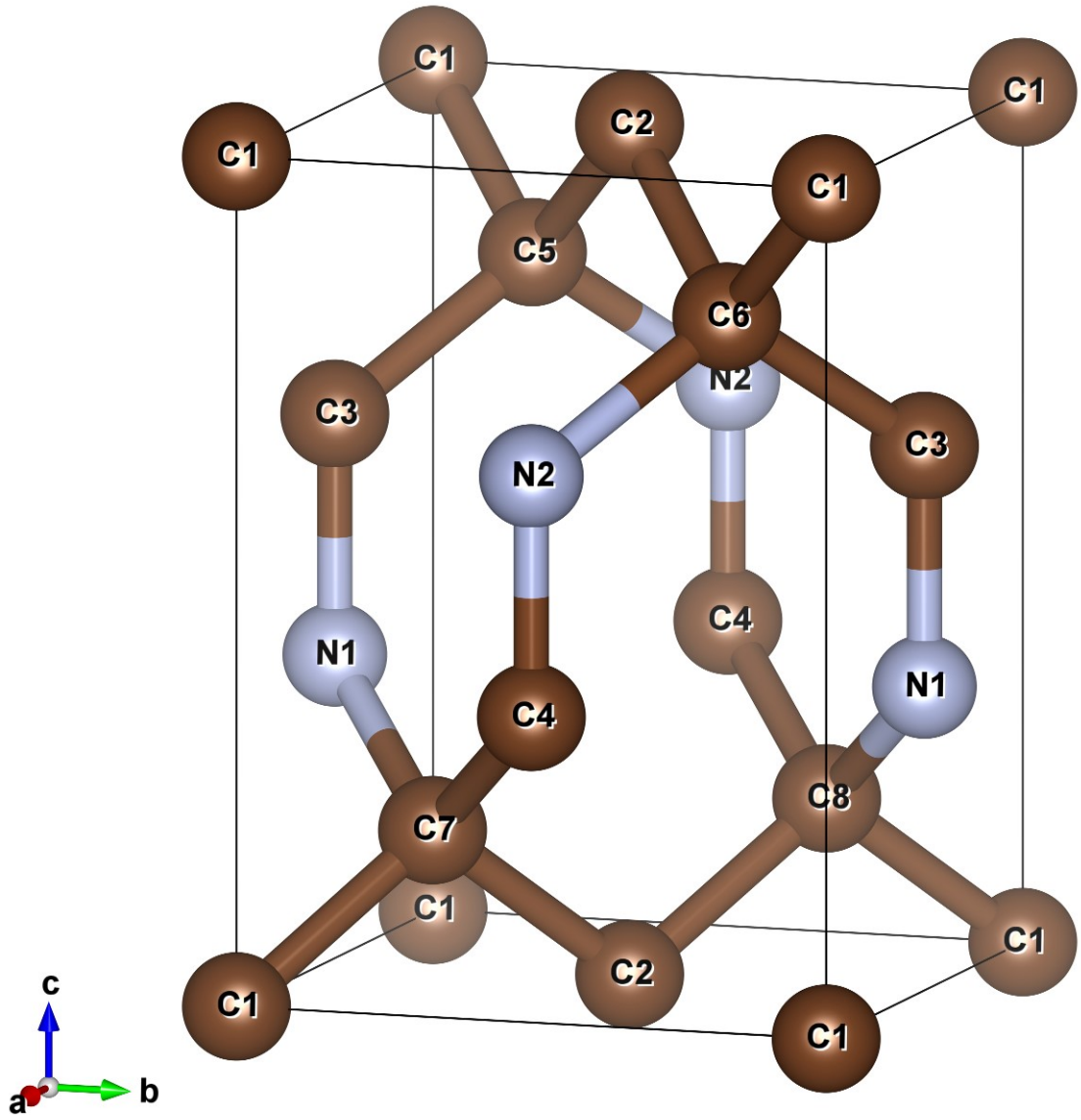

Fig. S4. The mark number of atoms in t-C_4N.

Table S4. Bond angle of t-C_4N and diamond.

t-C_4N		Diamond	
Bond-type	Bond-angle (º)	Bond-type	Bond-angle (º)
C1-C5-C2; C2-C6-C1 C2-C7-C1; C2-C8-C1 C5-C1-C6; C7-C1-C8 C6-C2-C5; C7-C2-C8	106.91		
C1-C5-C3; C3-C5-C2 C2-C6-C3; C1-C6-C3 C2-C7-C4; C1-C7-C4 C2-C8-C4; C4-C8-C1	109.14		
C1-C5-N2; N2-C5-C2 C2-C6-N2; C1-C6-N2 C2-C7-N1; N1-C7-C1 C2-C8-N1; C1-C8-N1	109.56		
		All C-C-C	109.47
C5-C1-C7; C5-C1-C8 C6-C1-C7; C6-C1-C8 C6-C2-C7; C6-C2-C8 C7-C2-C5; C8-C2-C5	110.77		
C5-C3-C6; C8-C4-C7	112.34		
C3-C5-N2; C3-C6-N2	112.38		
N1-C7-C4; C4-C8-N1	112.38		
C7-N1-C8; C5-N2-C6	112.43		
C3-N1-C7; C3-N1-C8	123.78		
C4-N2-C5; C4-N2-C6	123.78		
C5-C3-N1; N1-C3-C6	123.83		
C8-C4-N2; N2-C4-C7	123.83		

Table S5. Dihedral-angle of t-C_4N and diamond.

t-C_4N		Diamond	
Bond-type	Dihedral angle (º)	Bond-type	Dihedral-angle (º)
C6-C3-C5-C1; C7-C4-C8-C2 C6-C3-C5-C2; C7-C4-C8-C1	58.25		
C2-C5-N2-C6; C1-C7-N1-C8 C1-C5-N2-C6; C2-C7-N1-C8	58.49		
C8-C1-C5-C2; C6-C2-C7-C1 C5-C2-C8-C1; C8-C2-C6-C1	58.7		
C7-C1-C5-N2; C5-C2-C7-N1 C6-C2-C8-N1; C7-C2-C6-N2	58.89		
C8-C2-C6-C3; C8-C1-C5-C3 C6-C2-C7-C4; C5-C2-C8-C4	59.22		
C7-C2-C6-C1; C7-C1-C5-C2 C5-C2-C7-C1; C6-C2-C8-C1	59.76		
C6-C1-C5-C3; C8-C2-C7-C4 C7-C2-C8-C4; C5-C2-C6-C3	61.55		
C5-C2-C6-N2; C6-C1-C5-N2 C8-C2-C7-N1; C7-C2-C8-N1	61.89		
C5-C3-N1-C8; C6-C3-N1-C7 C8-C4-N2-C5; C7-C4-N2-C6 C5-C3-N1-C7; C6-C3-N1-C8 C8-C4-N2-C6; C7-C4-N2-C5	90		All C-C-C-C 60/180
C1-C5-N2-C4; C2-C7-N1-C3 C2-C5-N2-C4; C1-C7-N1-C3	121.51		
N1-C3-C5-C2; N2-C4-C8-C1 N1-C3-C5-C1; N2-C4-C8-C2	121.75		
C8-C1-C5-N2; C6-C2-C7-N1 C5-C2-C8-N1; C8-C2-C6-N2	177.34		
C7-C2-C6-C3; C7-C1-C5-C3 C5-C2-C7-C4; C6-C2-C8-C4	177.67		
C6-C1-C5-C2; C8-C2-C7-C1 C7-C2-C8-C1; C5-C2-C6-C1	179.47		
C7-C4-C8-N1; C3-C5-N2-C6 C4-C7-N1-C8; C6-C3-C5-N2 N1-C3-C5-N2; N2-C4-C8-N1 C3-C5-N2-C4; C4-C7-N1-C3	180		

Table S6. Crystal structural parameters a, b, c, ratio c/a and volumes V under increasing pressure.

	Space group	Pressure (GPa)	a=b (Å)	c (Å)	c/a	V (Å^3)
C_4N	$P\bar{4}2_1m$	0	3.47416	4.85415	1.397215442	58.588719
		50	3.34556	4.74088	1.417066201	53.063515
		100	3.25132	4.66058	1.433442417	49.267446
		150	3.17679	4.5963	1.446837846	46.385773
		200	3.11505	4.54251	1.458246256	44.078497
		250	3.06093	4.50008	1.470167563	42.162482
		300	3.01367	4.46088	1.480215153	40.514598
		350	2.97962	4.43473	1.48835422	39.372091

Table S7. Minimum and maximum of bulk module B, shear module G and Young's module E under applied pressure.

Pressure	Bulk module (GPa)		Shear module (GPa)		Young's module (GPa)	
(GPa)	B_{min}	B_{max}	G_{min}	G_{max}	E_{min}	E_{max}
0	368.85	582.38	215.02	478.29	599.98	1281.26
50	473.81	779.03	228.83	564.05	665.72	1516.44
100	570.48	944.16	235.59	633.91	709.09	1723.71
150	656.43	1105.94	238.11	693.05	735.31	1903.96
200	735.04	1272.11	238.87	753.50	752.89	2080.17
250	808.12	1438.47	239.31	815.61	766.58	2245.76
300	876.66	1615.12	238.80	937.01	775.17	2394.66
350	929.74	1751.41	235.73	1033.97	773.50	2490.14

Table S8. Cauchy pressure (C_{12}-C_{44}) and Pugh's modulus ratio (G/B) under applied pressure.

Pressure (GPa)	Cauchy pressure (C_{12}-C_{44}) (GPa)	Pugh's modulus ratio (G/B) (GPa)
0	-99.47	0.87
50	-31.78	0.77
100	47.98	0.69
150	128.04	0.63
200	206.65	0.59
250	280.99	0.56
300	354.05	0.54
350	429.38	0.52

Table S9. Comparison of mechanical properties of t-C_4N and diamond under different pressures.

Pressure (GPa)	Bulk module (GPa)		Shear module (GPa)		Young's module (GPa)		Hardness (Tian's) (GPa)		Pugh's ratio		Poisson's ratio	
	DO	t-C_4N	DO	t-C_4N	DO	t-C_4N	DO	t-C_4N	DO	t-C_4N	DO	t-C_4N
0	435.07	426.88	520.28	371.37	1115.99	863.66	94.45	51.81	1.19	0.87	0.07	0.16
50	559.57	553.91	612.19	424.56	1345.79	1014.5	95.78	49.32	1.09	0.77	0.1	0.19
100	670.81	667.44	683.32	461.23	1530.34	1124.6	95.46	46.49	1.02	0.69	0.12	0.22
150	773.78	771.23	741.44	489.14	1685.86	1211.3	94.34	43.96	0.96	0.63	0.14	0.24
200	870.99	869.49	791.51	515.42	1822.47	1291.1	93.03	42.25	0.91	0.59	0.15	0.25
250	964.07	962.78	835.35	542.46	1944.45	1370.1	91.56	41.35	0.87	0.56	0.16	0.26
300	1053.97	1053.5	874.67	570.31	2055.42	1449.4	90.06	40.93	0.83	0.54	0.17	0.27
350	1141.08	1123.1	909.98	582.97	2156.65	1490.9	88.52	39.64	0.80	0.52	0.19	0.28

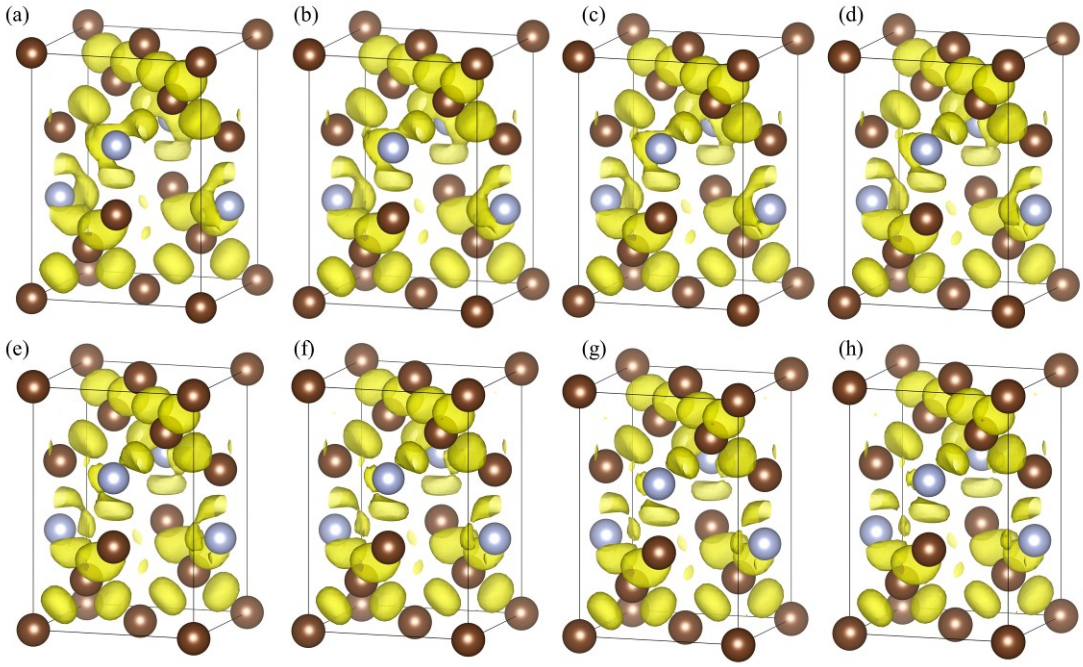

Fig. S5. ELF isosurface (ELF = 0.8) changes under applied pressure from 0 GPa to 350 GPa. (a) 0 GPa; (b) 50 GPa; (c) 100 GPa; (d) 150 GPa; (e) 200 GPa; (f) 250 GPa; (g) 300 GPa; (h) 350 GPa

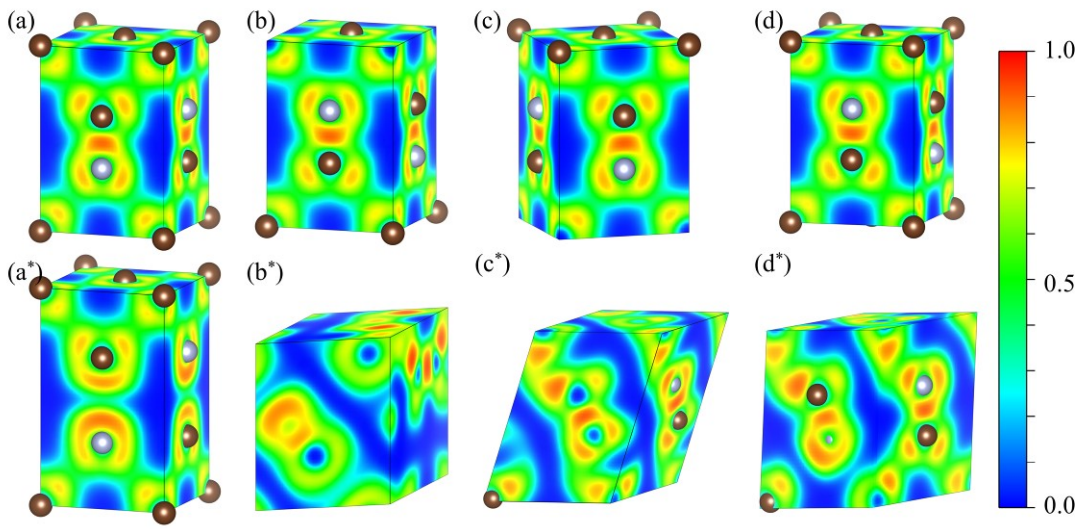

Fig. S6. ELF projections in (100), (010) and (001) crystal faces of tensile t-C_4N.
(a) (001) orientation 0% strain; (b) (100) orientation 0% strain; (c) (110) orientation 0% strain; (d) (111) orientation 0% strain; (a^*) (001) orientation 16.09% strain; (b^*) (100) orientation 36.13% strain; (c^*) (110) orientation 22.02% strain; (d^*) (111) orientation 11.57% strain.

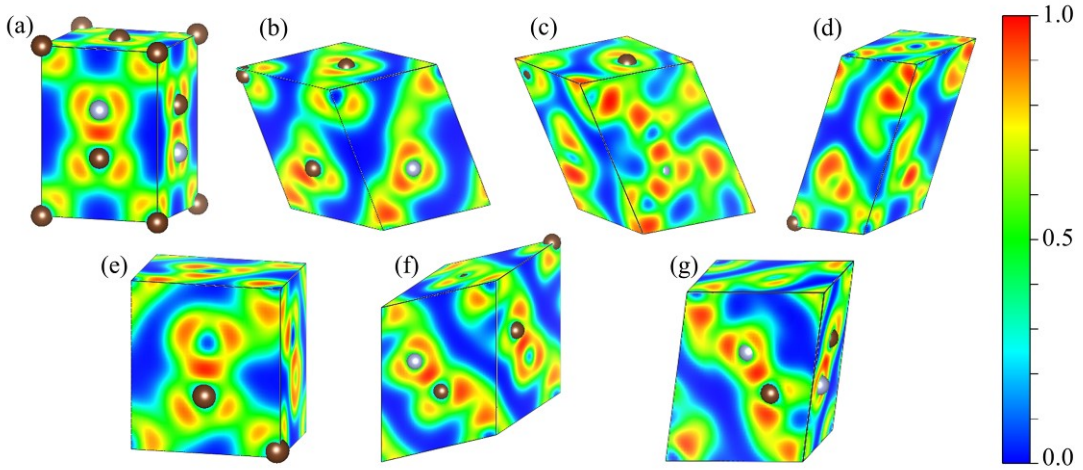

Fig. S7. ELF projections in (100), (010) and (001) crystal faces of shear t-C_4N.
(a) 0% strain; (b) (111)[$0\bar{1}1$] orientation 23.13% strain; (c) (111)[$1\bar{1}0$] orientation 34.52% strain; (d) (111)[$\bar{1}\bar{1}2$] orientation 29.91% strain; (e) (111)[$11\bar{2}$] orientation 27.62% strain; (f) (111)[$\bar{1}01$] orientation 23.13% strain; (g) (111)[$\bar{1}10$] orientation 39.23% strain.

The POSCARS of t-C_4N and diamond structures in Figure 1

t-C_4N

```
   1.00000000000000
     3.5130000114000000         0.0000000000000000         0.0000000000000000
     0.0000000000000000         3.5130000114000000         0.0000000000000000
     0.0000000000000000         0.0000000000000000         4.8892998695000003
   C     N
     8     2
Direct
  0.0000000000000000  0.0000000000000000 -0.0000000000000000
  0.5000000000000000  0.5000000000000000 -0.0000000000000000
  0.5000000000000000  0.0000000000000000  0.6404776886635993
 -0.0000000000000000  0.5000000000000000  0.3595223113364007
  0.2484396071159711  0.2515603928840289  0.8116394231067676
  0.7515604218840278  0.7484395781159722  0.8116394231067676
  0.7484395781159722  0.2484396071159711  0.1883605618932347
  0.2515603928840289  0.7515604218840278  0.1883605618932347
  0.5000000000000000  0.0000000000000000  0.3573275417917833
  0.0000000000000000  0.5000000000000000  0.6426724282082142
```

Diamond

```
   1.0
        3.5559999943          0.0000000000          0.0000000000
        0.0000000000          3.5559999943          0.0000000000
        0.0000000000          0.0000000000          3.5559999943
   C
   8
Direct
     0.000000000           0.000000000           0.000000000
     0.000000000           0.500000000           0.500000000
     0.500000000           0.500000000           0.000000000
     0.500000000           0.000000000           0.500000000
     0.750000017           0.250000000           0.750000017
     0.250000000           0.250000000           0.250000000
     0.250000000           0.750000017           0.750000017
     0.750000017           0.750000017           0.250000000
```

The POSCARS of t-C_4N structures under different pressures

t-C_4N (50 GPa)

1.00000000000000

3.3455572799516173 0.0000000000000000 0.0000000000000000

0.0000000000000000 3.3455572799516173 0.0000000000000000

0.0000000000000000 0.0000000000000000 4.7408813765841558

C N

8 2

Direct

0.0000000000000000 0.0000000000000000 0.0000000000000000

0.5000000000000000 0.5000000000000000 0.0000000000000000

0.5000000000000000 0.0000000000000000 0.6410808634470868

0.0000000000000000 0.5000000000000000 0.3589191365529132

0.2479810595170306 0.2520189404829694 0.8118651602049383

0.7520189694829682 0.7479810305170318 0.8118651602049383

0.7479810305170318 0.2479810595170306 0.1881348247950640

0.2520189404829694 0.7520189694829682 0.1881348247950640

0.5000000000000000 0.0000000000000000 0.3573834667796234

0.0000000000000000 0.5000000000000000 0.6426165032203741

t-C_4N (100 GPa)

1.00000000000000

3.2811315271242059 -0.0000000344688479 0.0000000019707171

-0.0000000344652765 3.2811315438409698 -0.0000000012739228

0.0000000014536558 -0.0000000017250822 4.6889624505861160

C N

8 2

Direct

0.9999999993082227 0.9999999997179430 0.9999999856412956

0.5000000000884768 0.5000000000093507 0.9999999861925986

0.5000000008659171 0.9999999991994670 0.6413252388405510

0.9999999983969730 0.5000000020898128 0.3586747465409701

0.2479997146929591 0.2520002860337556 0.8118770822885537

0.7520002844915936 0.7479997130801266 0.8118770827277828

0.7479997315342075 0.2479997320912091 0.1881229255562076

0.2520002695732799 0.7520002685671443 0.1881229235906758

0.5000000020373605 0.9999999993902193 0.3577243080508055

0.9999999990110169 0.4999999998209645 0.6422756605705615

t-C_4N (150 GPa)

1.00000000000000

3.2030181027177762 0.0000002095625297 0.0000000037275832

0.0000002095635420 3.2030180983579810 -0.0000000046890749

0.0000000039422891 -0.0000000068089987 4.6223560585460746

C N

8 2

Direct

0.9999999970279987 0.0000000005529088 0.0000000978710659

0.5000000017639721 0.4999999997429043 0.0000000989624027

0.4999999984287768 0.9999999981760510 0.6413404474420332

0.0000000023610909 0.5000000004601191 0.3586595546174323

0.2478326271329578 0.2521673727542293 0.8119783598948089

0.7521673761958567 0.7478326249993117 0.8119783592821008

0.7478326190755027 0.2478326219637808 0.1880214466939591

0.2521673795335673 0.7521673810245346 0.1880214464903815

0.4999999943571964 0.9999999997430606 0.3581319221805259

0.0000000041230734 0.5000000005831211 0.6418682065653059

t-C_4N (200 GPa)

1.00000000000000

3.1150532554549040 0.0000000000000000 0.0000000000000000

0.0000000000000000 3.1150532554549040 0.0000000000000000

0.0000000000000000 0.0000000000000000 4.5425095376956106

C N

8 2

Direct

0.0000000000000000 0.0000000000000000 0.0000000000000000

0.5000000000000000 0.5000000000000000 0.0000000000000000

0.5000000000000000 0.0000000000000000 0.6413121992840374

0.0000000000000000 0.5000000000000000 0.3586878007159626

0.2476199643001280 0.2523800356998720 0.8121929654786797

0.7523800646998708 0.7476199353001292 0.8121929654786797

0.7476199353001292 0.2476199643001280 0.1878070195213226

0.2523800356998720 0.7523800646998708 0.1878070195213226

0.5000000000000000 0.0000000000000000 0.3585666529298805

0.0000000000000000 0.5000000000000000 0.6414333170701170

t-C_4N (250 GPa)

1.00000000000000

3.0609269158268400 0.0000000000000000 0.0000000000000000

0.0000000000000000 3.0609269158268400 0.0000000000000000

0.0000000000000000 0.0000000000000000 4.5000800606079183

C N

8 2

Direct

0.0000000000000000 0.0000000000000000 0.0000000000000000

0.5000000000000000 0.5000000000000000 0.0000000000000000

0.5000000000000000 0.0000000000000000 0.6411712344304590
0.0000000000000000 0.5000000000000000 0.3588287655695410
0.2474622518829506 0.2525377481170494 0.8122486636918040
0.7525377771170483 0.7474622228829517 0.8122486636918040
0.7474622228829517 0.2474622518829506 0.1877513213082054
0.2525377481170494 0.7525377771170483 0.1877513213082054
0.5000000000000000 0.0000000000000000 0.3591177609047023
0.0000000000000000 0.5000000000000000 0.6408822090952953

t-C_4N (300 GPa)

1.00000000000000
3.0136698649734619 -0.0000012990584413 0.0000000093541754
-0.0000012990479598 3.0136698584805468 -0.0000000134838198
0.0000000161083811 -0.0000000236319370 4.4608758938770912
C N
8 2
Direct
0.9999999983374863 0.0000000031271057 0.9999999567494129
0.4999999952030265 0.4999999996911484 0.9999999566950777
0.5000000024491840 0.0000000046860080 0.6411191584888485
0.9999999968642399 0.5000000031103369 0.3588808742686496
0.2473768080508307 0.2526231913927077 0.8122852939307492
0.7526231997974975 0.7473767976848791 0.8122852968474561
0.7473767869973429 0.2473767917122842 0.1877147726256183
0.2526232114431295 0.7526232134661299 0.1877147707105777
0.5000000036325432 0.9999999958886150 0.3595451780458916
0.9999999972247267 0.4999999992407780 0.6404546816377135

t-C_4N (350 GPa)

1.00000000000000
2.9796195229131861 0.0000025583865974 -0.0000000002834292
0.0000025583860184 2.9796195277915078 -0.0000000170267606
0.0000000037649978 -0.0000000302614490 4.4347265402772935
C N
8 2
Direct
0.0000000003625829 0.9999999999107771 0.9999999723079966
0.4999999958784400 0.5000000034450025 0.9999999706013796
0.5000000036840930 0.9999999993415116 0.6410059376032464
0.0000000023417286 0.5000000038690970 0.3589938605153975
0.2473409459678777 0.2526590520499070 0.8123588853387815
0.7526590568619582 0.7473409369518365 0.8123588859298252
0.7473408359854261 0.2473408412604599 0.1876413073705336
0.2526591623650773 0.7526591718420974 0.1876413125153746

0.5000000051329891 0.0000000052365721 0.3599378982304984
0.9999999914198199 0.4999999860927389 0.6400619095869757

CCDC number of the t-C_4N crystal structure

Data Block Name: data_C_4N-P42
Unit Cell Parameters: a 3.5130 b 3.5130 c 4.8893 P-421m
Deposition Number: 2121547

Explanation to the A-alerts in checkCIF file

The A-alerts are all about experimental crystal descriptions. In this work, the t-C_4N crystal structure was predicted by CALYPSO code theoretically and has not been prepared experimentally at present. Therefore, the diffractometer model, parameters, experimental descriptions of atoms, bonds and angles are not provided, which leads to that the relevant tests cannot be performed (as A-alerts described in checkCIF file). The checkCIF report is for guidance only. If used as part of a review procedure for publication, it should not replace the expertise of an experienced crystallographic referee.