

## **SUPPORTING INFORMATION**

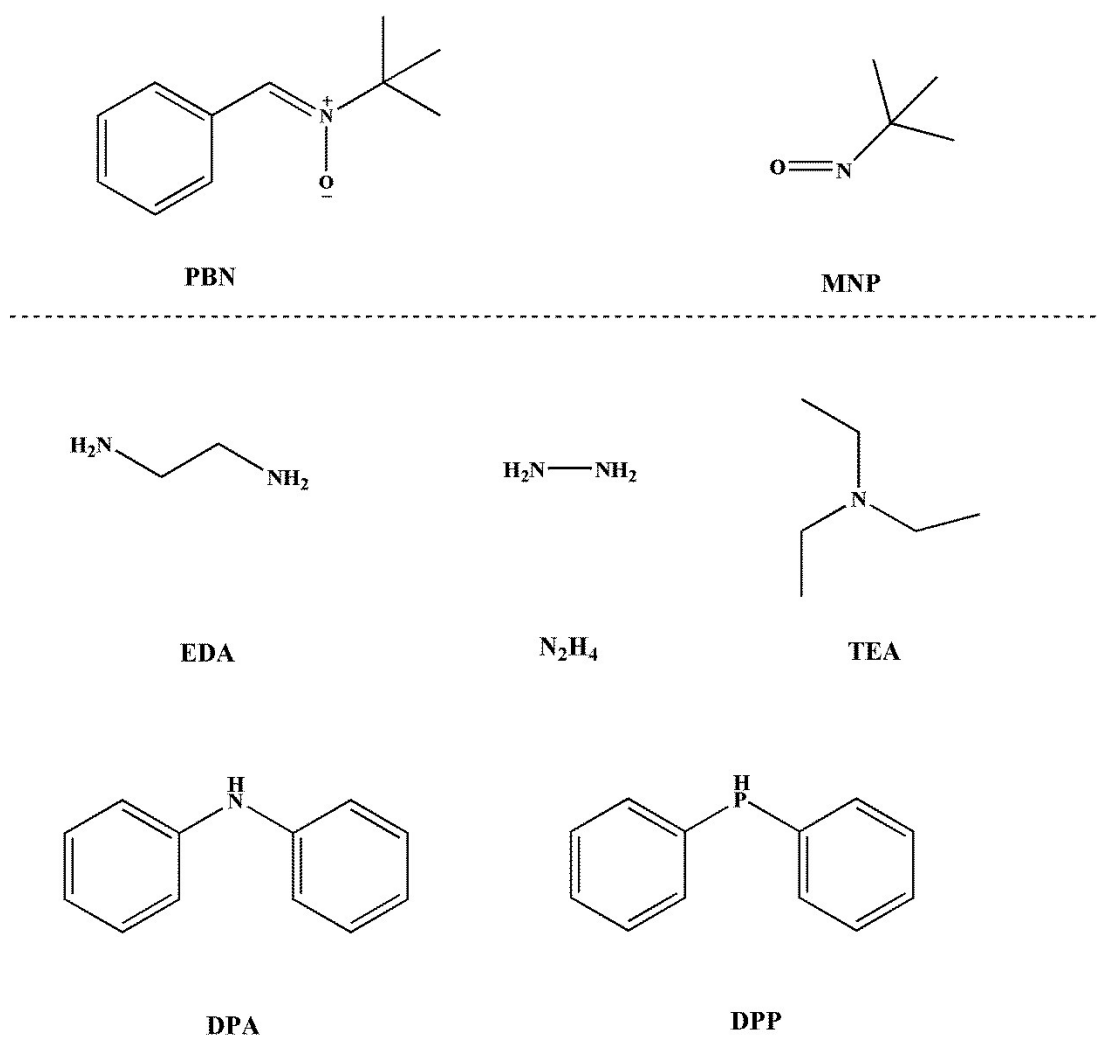
**OF**

### **Radical Mechanism of Nucleophilic Reaction Depending on Two-Dimensional Structure**

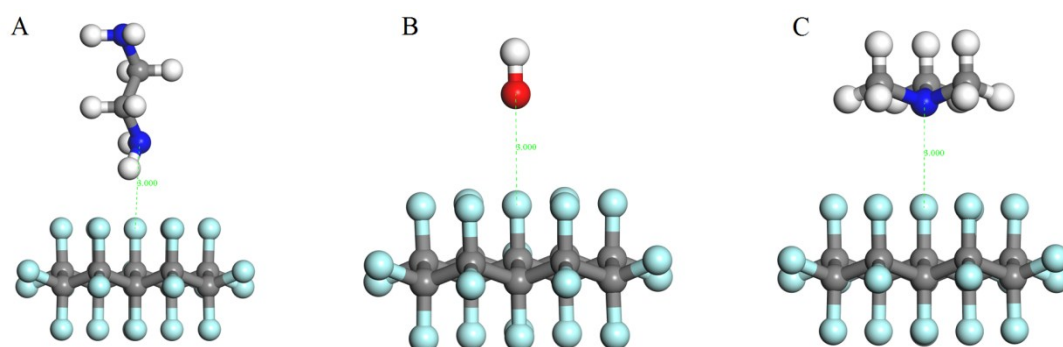
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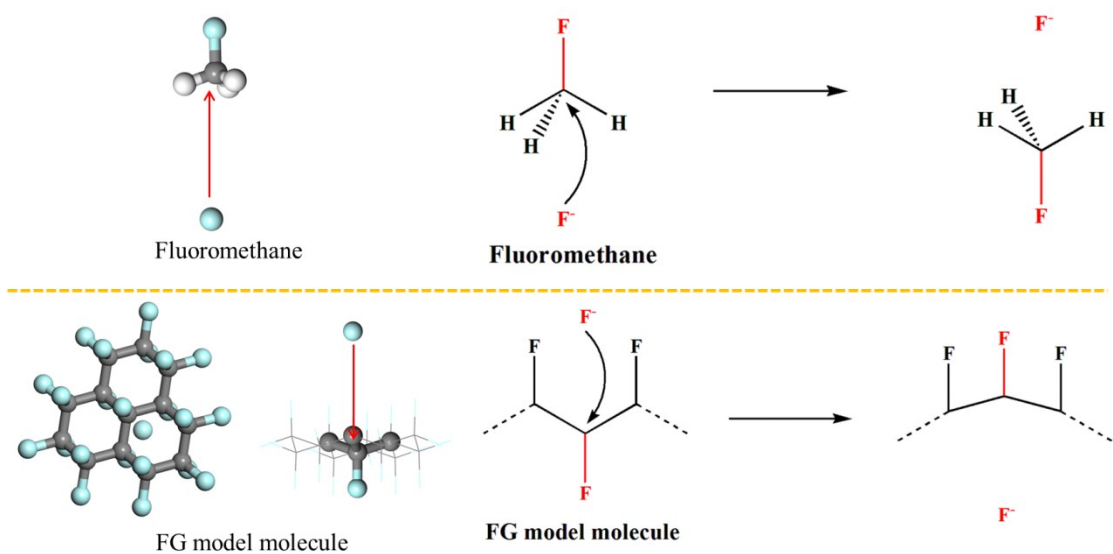


**Fig. S1** The chemical structures of mentioned reagents of radical trappers and nucleophiles.

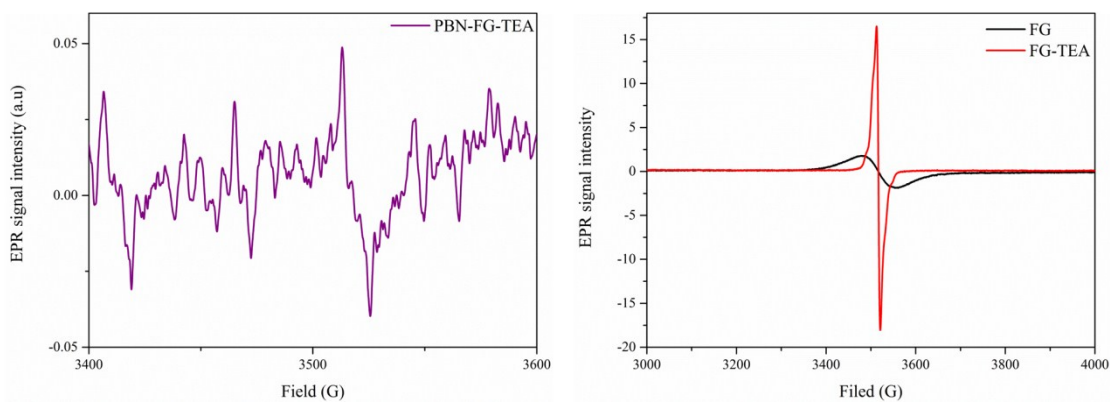


**Fig. S2** The three model structures for calculation of SET reaction for particular nucleophile EDA

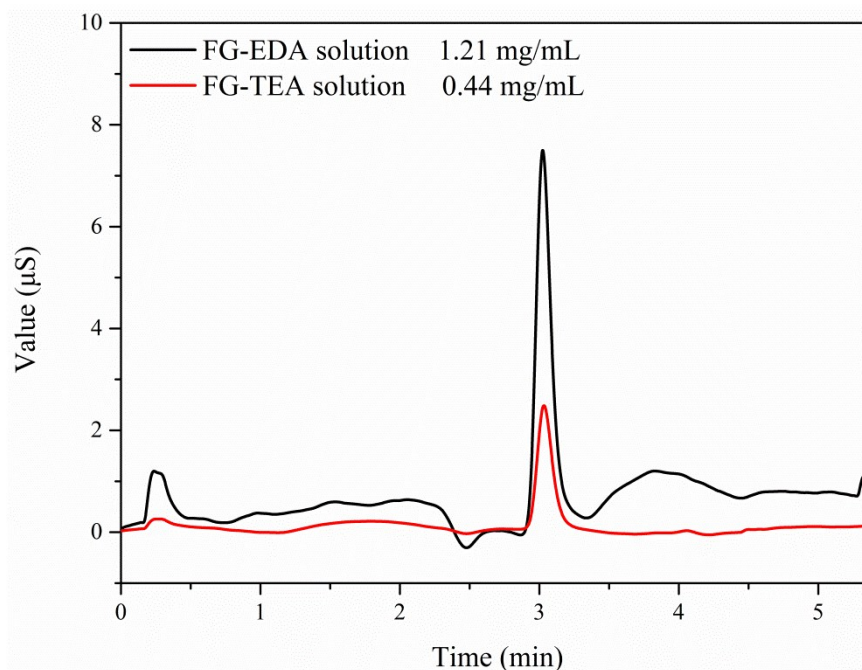
(A), OH<sup>-</sup> (B) and TEA (C).



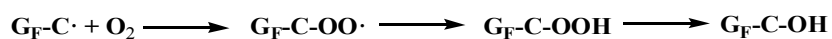
**Fig. S3** The schematic of S<sub>N</sub>2 nucleophilic attacking of FG model molecule and fluoromethane.



**Fig. S4** EPR spectrum of radical intermediate of PBN-FG-TEA reaction and FG derivative after treatment by TEA.



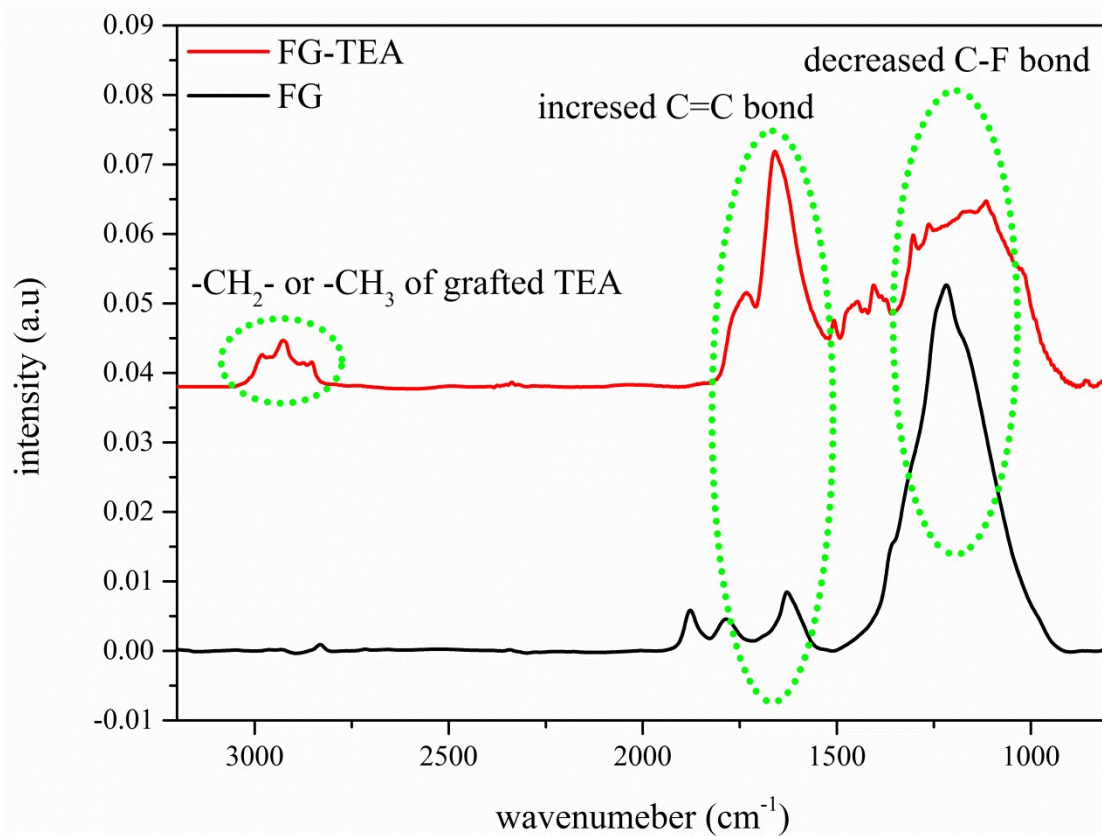
**Fig. S5** Ion chromatography curves of fluorine anion of FG-EDA and FG-TEA solution.



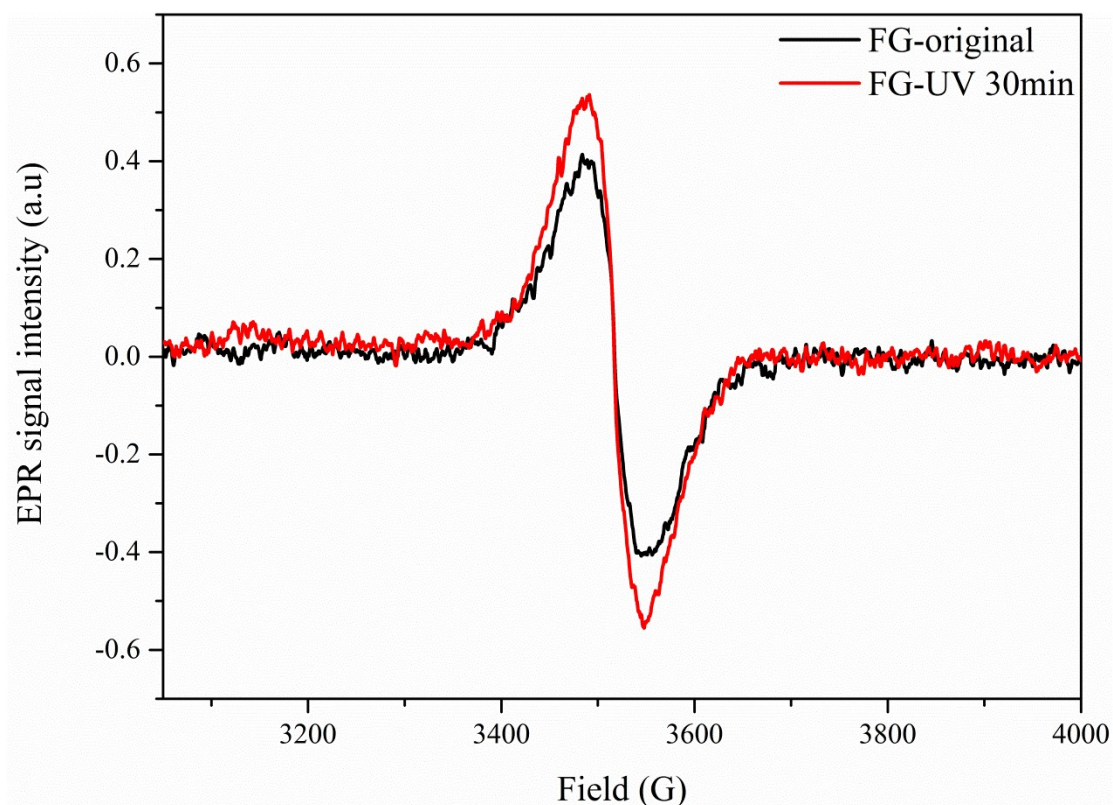
**Scheme S1** The reaction of spin center on graphene nanosheet with trace amount of oxygen purity.

**Table S1** Elemental composition analysis of FG, derivative FG-TEA and G-TEA.

Sample	C (wt%)	H (wt%)	N (wt%)	Increased H / Increased N	Increased Group
FG	39.87	1.03	< 0.1	/	/
FG-TEA	52.53	3.16	3.13	9.33	-N(CH <sub>2</sub> CH <sub>3</sub> ) <sub>2</sub>
G-TEA	71.98	2.69	< 0.1	/	/



**Fig. S6** FT-IR spectra of FG and FG-TEA samples.



**Fig. S7** The EPR spectra of FG and FG samples under ultraviolet radiation for 30min.

**Table S2** The chemical composition of derivative FG-FM and FG-FM under ultraviolet radiation for 30min based on XPS calculation.

Sample	C (at%)	N (at%)	O (at%)	F (at%)
FG-FM	60.66	2.86	14.06	22.42
FG-FM-UV	61.31	3.55	14.26	20.88

### Supplementary data

#### Atomic coordinated of calculated structures

**FG model molecule for defluorination calculations (Fig. 1):**

	\$coordinates		
C	-4.89142107440126	-16.48966061824218	4.32169426673277
C	-3.67593826661441	-15.55902934775437	1.72713599593280
C	-3.89476458913298	-12.59408496621525	1.50791171598957
C	-2.39747657704688	-11.34004869357084	3.76018804549774
C	-3.59135235647604	-12.19708240126655	6.38971605917841
C	-3.43144650778676	-15.21005196318254	6.64548728900024
C	-2.68571629313255	-11.71779650832991	-1.07207586687707
C	-4.17308834721270	-12.96129102504427	-3.37765966394863
C	-4.01221251722127	-15.97856047734397	-3.18140434176082
C	-5.17498697054624	-16.87180182136445	-0.53196808980951
C	-2.58984528061615	-8.33930187267240	3.57832243028255
C	-1.40647181853282	-7.40177886738277	0.95361635327626
C	-2.88261258207607	-8.72351406822903	-1.33613186579047
F	-4.76711445337245	-19.07008418549229	4.51836607888935
F	-7.42685187644139	-15.98969938295007	4.44300307159032
F	-1.18981166261516	-16.31587277529828	1.64079771063338
F	0.10895217107486	-12.02988736112821	3.70644484488446
F	-6.03159161781500	-11.38180855878989	6.66183068554773
F	-2.34878614882332	-11.07893387384701	8.36925297350692
F	-4.46240227433468	-15.97937528945455	8.89096125452423
F	-0.98131432420332	-16.01945315480317	6.78330275707330
F	-0.18408855107862	-12.41385726544972	-1.20881750185906
F	-3.18439371652548	-12.17246925139128	-5.63886575064128
F	-6.63684682809852	-12.17894802434300	-3.48270008493722
F	-1.58697302691932	-16.81851413734388	-3.48119217419633
F	-5.29258104816331	-17.07845214524117	-5.14299828385580
F	-5.04403736125516	-19.45235046391760	-0.34014342138409
F	-7.71297491218418	-16.38023291648522	-0.43178303097099
F	-5.02442853119177	-7.50341714639840	3.83798373125255
F	-1.34468647832406	-7.21934700140341	5.55522132473510
F	1.13192921723605	-7.87163575072081	0.83878179925582
F	-1.56621402638830	-4.82605676109739	0.76153704614522
F	-1.88878544200528	-7.93509234959670	-3.59523901288910
F	-5.33820029915771	-7.91502838277555	-1.43421242168126
F	-6.39930151353373	-11.88074808530662	1.60139522584293
	\$end		

**SET reaction for EDA (step 1 of Scheme 1 and Table 1):**

Scoordinates

C	-3.20727996852732	-4.54711497564740	1.85508264899755
C	-0.26619378560737	-4.45282452817222	2.26590886290676
C	0.80118836629236	-1.69712682948364	1.86888484389736
C	-0.61214303713171	0.29191100419172	3.58345600736138
C	-3.55248037450066	0.18174647743164	3.17000479000975
C	-4.64318236283353	-2.54336038387842	3.47360420381882
C	3.75505573642515	-1.58027995713530	2.23794721660376
C	5.14925528043922	-3.57056538043947	0.52937448416666
C	4.08191599319719	-6.29863902672809	0.87111983205608
C	1.15190847519392	-6.42880681474728	0.55841852968926
C	0.46388162413286	3.03800973698318	3.21414765467447
C	3.39886025757026	3.16946232711804	3.47464557519452
C	4.79273769020743	1.16975509218206	1.81755533944093
F	-4.07572375752807	-6.86346694807440	2.53740447357904
F	-3.79345452530965	-4.14436371075865	-0.61284850401952
F	0.15426667538571	-5.18459050769085	4.73678180360521
F	-0.20389472146180	-0.30468696820154	6.09278533390690
F	-4.17786592445602	1.02327380875186	0.81662926176790
F	-4.70568151307093	1.68520661661116	4.89692594108968
F	-7.09939295065350	-2.51489031809984	2.73147198046345
F	-4.54407481071367	-3.21834145394381	5.94574819131648
F	4.33028343479828	-2.20335064738959	4.70763247766073
F	7.63642715431917	-3.62386419683665	1.15915808632929
F	4.94623557965206	-2.93731696164721	-1.95235223459098
F	4.72756809050402	-7.17286648505083	3.19454508905318
F	5.15338189994481	-7.81331421046902	-0.90091385129618
F	0.40618443049551	-8.79383697186228	1.21956148872010
F	0.59855820419998	-6.05514307042037	-1.92045741815607
F	-0.16238804174898	3.95598003140615	0.89289216798601
F	-0.54572772429753	4.58394885398190	4.99363413440479
F	4.04410934243505	2.83444087044574	5.93225012893923
F	4.17000657617888	5.50477285973102	2.73551259286391
F	7.27953769055241	1.24443461955675	2.43594585460557
F	4.54820596367224	1.87381951407345	-0.64880944982675
F	0.31824812286493	-1.05003422607707	-0.63099931589117
N	-4.62818679076797	7.38963432323227	-8.57669551781840
C	-3.99086816457441	5.15799692861604	-7.07776700235784
C	-1.31690649139345	5.10789699713954	-5.96587253299760
N	-0.83152697247634	2.66490056491299	-4.76660539139000
H	-3.43139162825037	7.51907545486462	-10.08746931851057
H	-4.35079097325330	8.99203239082495	-7.53485251372053
H	-4.24331362570461	3.46302114225513	-8.25411176984873
H	-5.37730825123431	5.00069374491665	-5.52788869455978



H	-1.08667345695152	6.76117565907848	-4.68709739455772
H	0.06362351082561	5.35548538024835	-7.50838443324740
H	-2.05943304578905	2.39999402105772	-3.30128202489857
H	0.93444280083877	2.63011616070145	-3.99462162742100

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**SET reaction for OH<sup>-</sup> (step 1 of Scheme 1 and Table 1):**

	Scoordinates		
C	0.02923763284630	-4.92074174370322	0.10144608630981
C	1.43210498209913	-2.46159122958210	1.00951208989592
C	0.02997989270176	0.00179106730346	0.07921091487789
C	-2.84835881107071	0.00223258291530	0.87253606462374
C	-4.22890854014078	-2.46821728022911	-0.03879450698371
C	-2.83394605699827	-4.92350724450358	0.82070737578331
C	1.42661530313958	2.47785132322255	0.98448567581437
C	4.29051447353069	2.46669386428536	0.18147778294310
C	5.69341696224081	0.01236591233796	1.03003193991664
C	4.29543687560477	-2.45218255910577	0.20566099744823
C	-4.23601713424353	2.45846225507941	-0.06733885411681
C	-2.84410965416427	4.92569022588756	0.76257878034834
C	0.01957486979521	4.92279296393313	0.04288202049687
F	1.11180207498081	-6.96211977728652	1.21011922080300
F	0.21028672763850	-5.17718799503525	-2.45014354839636
F	1.40564355724141	-2.52367783748861	3.61499160575718
F	-3.00697029374040	0.01454820775135	3.47393448448177
F	-4.41290492171739	-2.51234942258780	-2.59804759339799
F	-6.58849991995914	-2.52881397892579	0.96161130830013
F	-3.95092472867314	-6.92949826667026	-0.31983036976240
F	-3.06522331441262	-5.18719701071451	3.36092705562898
F	1.39651931322361	2.56620376366366	3.58931900927582
F	5.47845623891708	4.45881516218914	1.27065784821586
F	4.51735493746247	2.66385707704403	-2.37098066657711
F	5.95434655478932	0.02350221201173	3.57938161232698
F	8.02728634246772	0.00910690011143	-0.03477219280132
F	5.48681626446186	-4.43208589699910	1.31247793522706
F	4.52194645227152	-2.67718695946534	-2.34707930094408
F	-4.42531924505922	2.46865176228031	-2.62367710790315
F	-6.59473306630914	2.52238804082680	0.93981290737776
F	-3.07621753502768	5.21995842342499	3.30005873525799
F	-3.96567716668573	6.91437017000128	-0.40167915144845
F	1.09747803410131	6.97999606675428	1.12988074466793

F	0.20058177645166	5.14437526870157	-2.50750437456291
F	0.10882970737333	-0.01011512569090	-2.53887965804290
O	-1.07500389168308	0.39326565610297	-8.10866961846876
H	0.41858530465651	-0.48044657973015	-7.42630525237275

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**SET reaction for TEA (step 1 of Scheme 1 and Table 1):**

	Scoordinates		
C	-0.05050761721124	-5.02374336185227	2.72438252361717
C	1.33628842393075	-2.49818594795768	3.44474990002725
C	-0.13550722997235	-0.11309605889778	2.43881597253394
C	-2.98297611987791	-0.10893847811936	3.31709347299050
C	-4.35308279719290	-2.64448708946557	2.59814913972104
C	-2.88867128969719	-5.02723979737942	3.53234437320935
C	1.25291068751368	2.42686387242283	3.13999804241003
C	4.08503946670713	2.41013569362122	2.24451512982652
C	5.55902061910940	0.02798400365279	3.16585849656548
C	4.16841193456776	-2.49685741757948	2.54943512771544
C	-4.43771296660684	2.26836490782607	2.29192667266691
C	-3.06146890859269	4.79892407906391	2.91746804943853
C	-0.21945894613344	4.79661245268425	2.12287148817634
F	1.09938880799589	-6.98436293092159	3.91801934339838
F	0.05517585301124	-5.43264350831338	0.19100326495012
F	1.40073554354204	-2.41848014971245	6.05645964261165
F	-3.06808099794308	0.05304346148536	5.92426489844600
F	-4.64202002630911	-2.83372004764344	0.05302446217890
F	-6.67660598357725	-2.68539739429006	3.69098386539967
F	-4.01021228180222	-7.10849843158061	2.53317810865940
F	-3.04084160685478	-5.16654879726729	6.08914157344132
F	1.31563297661192	2.67316227367666	5.74173609896505
F	5.27872946905067	4.47867577936143	3.18182340931602
F	4.22778701414177	2.48486799726713	-0.32375014551553
F	5.93478172321929	0.19131072150093	5.69740157071783
F	7.84630661574212	-0.00440750461735	1.99975259214243
F	5.43031070188331	-4.38982761462486	3.73603023458092
F	4.31866120716928	-2.88393260112780	0.01058824130300
F	-4.72334850592171	2.13435878558057	-0.26041843108033
F	-6.76653877363080	2.36258631793673	3.36733924219096
F	-3.23233225484925	5.25883904711794	5.43449217068968
F	-4.24750802401712	6.69645691779288	1.65877349796107
F	0.85404435182831	6.92779667222964	3.06657009954519

F	-0.11168259288916	4.90344619352984	-0.44517606614967
F	-0.15214331675071	-0.27995239208438	-0.18855439910036
N	-0.04692302029118	-0.41570352522823	-7.11028413395160
C	-0.22990304857626	-0.86443397724640	-9.81183372041009
C	-1.36985623569109	1.87083709161120	-6.35840431584965
C	2.55990041776757	-0.44359122276851	-6.24632420206256
H	0.65934330292214	-2.68142607436715	-10.28377797821826
H	0.71107751490496	0.63761153563267	-10.94755872279903
H	-2.22600113149090	-0.94150603773506	-10.37902417168869
H	-3.35850395241548	1.75959375233171	-6.94700026753306
H	-0.53518811509636	3.60438411003469	-7.20975085497767
H	-1.31028108892251	2.07424326595736	-4.29737417558496
H	3.70981749390180	1.10558158979585	-7.08663868069959
H	3.44683234231220	-2.25162762432399	-6.75781829605577
H	2.62716036259058	-0.24707253700956	-4.18450214182957

\$end

**S<sub>N</sub>2 substitution of fluoromethane (Fig. S2 and Table 2):**

\$coordinates			
C	1.45512706194565	-0.08848793191455	-0.03888692026040
H	0.84129088500801	-2.00432158897497	0.48584190088482
H	0.78091808673179	0.39448250744663	-1.94546255392145
H	0.77654438931481	1.30142331707532	1.34986427310448
F	4.09623030387687	-0.03499576251971	-0.05064555210056
F	-7.95011072498740	0.43189945888728	0.19928885229311

\$end

**S<sub>N</sub>2 substitution of FG model molecule (Fig. S2 and Table 2):**

\$coordinates			
C	-4.22643369244653	-2.53941742874465	-0.50417259011714
C	-1.24949304222758	-2.50033332690146	-0.56845941370988
C	-0.19939205779570	0.29136609410467	-0.49439680443624
C	-1.26950222588837	1.80743446142371	1.84444304115902
C	-4.24630596544956	1.75699868404084	1.90270470354881
C	-5.32549474067412	-0.99220898837694	1.76550569059389
C	2.78436833621861	0.34364351920047	-0.56467210964785
C	3.83830503004722	-1.15432854728672	-2.91148432615005
C	2.76884189095167	-3.90802335927048	-3.05821240403272

C	-0.18586665486087	-4.00217273302125	-2.90993587700432
C	-0.22265082772911	4.59358255304520	1.90033191807284
C	2.73180241797367	4.68584959327957	1.77585405195231
C	3.82238723761760	3.13238557929222	-0.49317406092651
F	-5.02962389097652	-4.95666157284943	-0.25234353686192
F	-5.19637887437159	-1.56695966318435	-2.67131752492317
F	-0.44556565208760	-3.77131382128673	1.54803476846928
F	-0.47018593945845	0.67224252242875	4.03915470730245
F	-5.21845533526517	3.09155198601005	-0.06150778479442
F	-5.06047726422613	2.80203236786855	4.09361260611565
F	-7.86774864833238	-0.84337611841844	1.47653078411819
F	-4.81124742910036	-2.21345071196327	3.95190664359577
F	3.71181966629013	-0.84823995221073	1.54803048068071
F	6.38666615648597	-1.32186997611349	-2.72661025938496
F	3.24484277092002	0.04664687839199	-5.09989656190992
F	3.75618048440721	-5.29750691221565	-1.15428584514547
F	3.49273955166325	-4.93105026396575	-5.29372825365064
F	-0.87307690802017	-6.46030990040511	-2.70925976164505
F	-1.12304985715761	-3.05761613307933	-5.10353020478183
F	-1.14532103165165	5.95203717151457	-0.07167543542296
F	-0.94168868732395	5.71819184999567	4.08466532739055
F	3.70250463552275	3.78800723290709	3.96477957512225
F	3.44996467296849	7.12832661916287	1.48963089662969
F	6.36924555368627	3.07638353990886	-0.23812366524534
F	3.23461325065815	4.37616918441946	-2.65899705293960
F	-1.04520546182220	1.49949241729516	-2.65399227581401
F	2.85888253145462	-4.39750285066509	6.81459055379261

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