Supplementary information

Remarkable nonlinear optical response of excess electron compounds: Theoretically designed alkali-doped aziridine M-$(C_2NH_5)_n$

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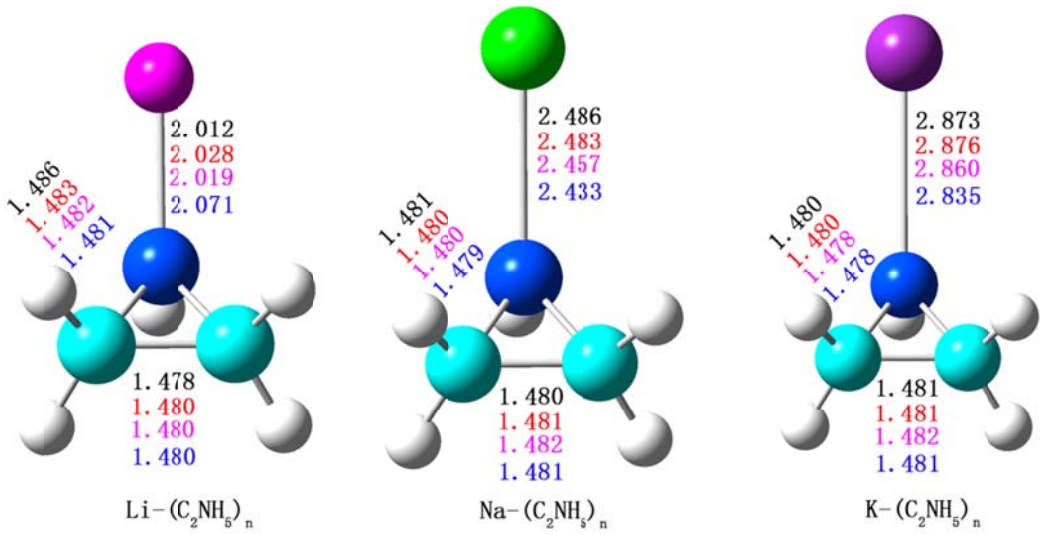

Fig.S1 Optimized bond lengths: M-N, C-N and C-C for M-$(C_2NH_5)_n$, in which black, read, magenta and blue colors represent compounds with *n* =1, 2, 3 and 4 respectively.

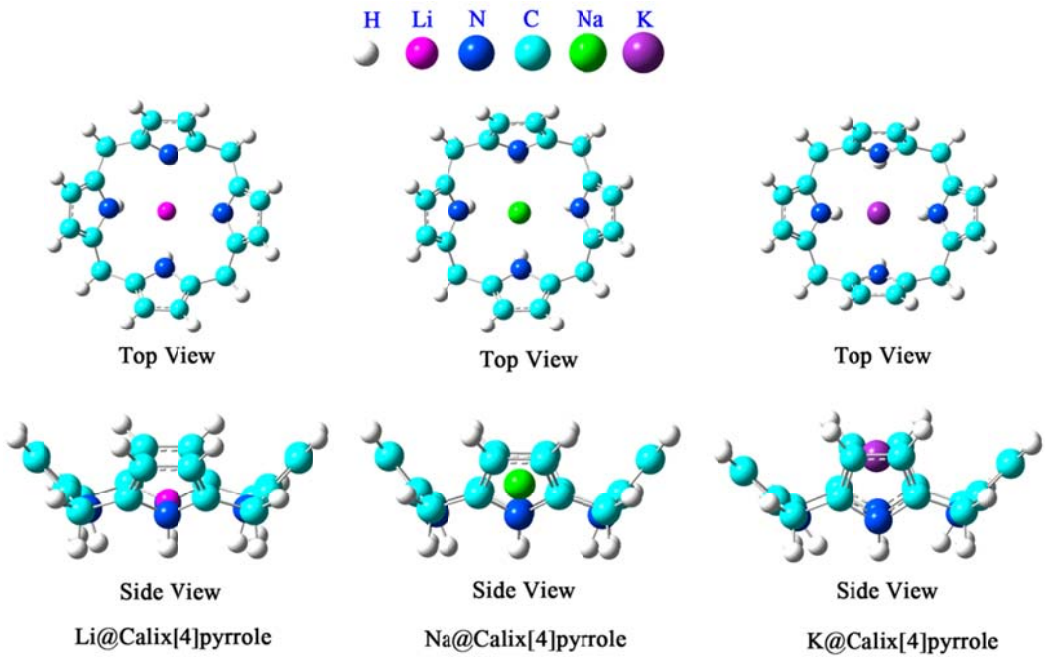

Fig.S2 Optimized geometrical structures of M-@Calix[4]pyrrole (M=Li, Na and K) at the B3LYP/6-311++G(d,p) level.

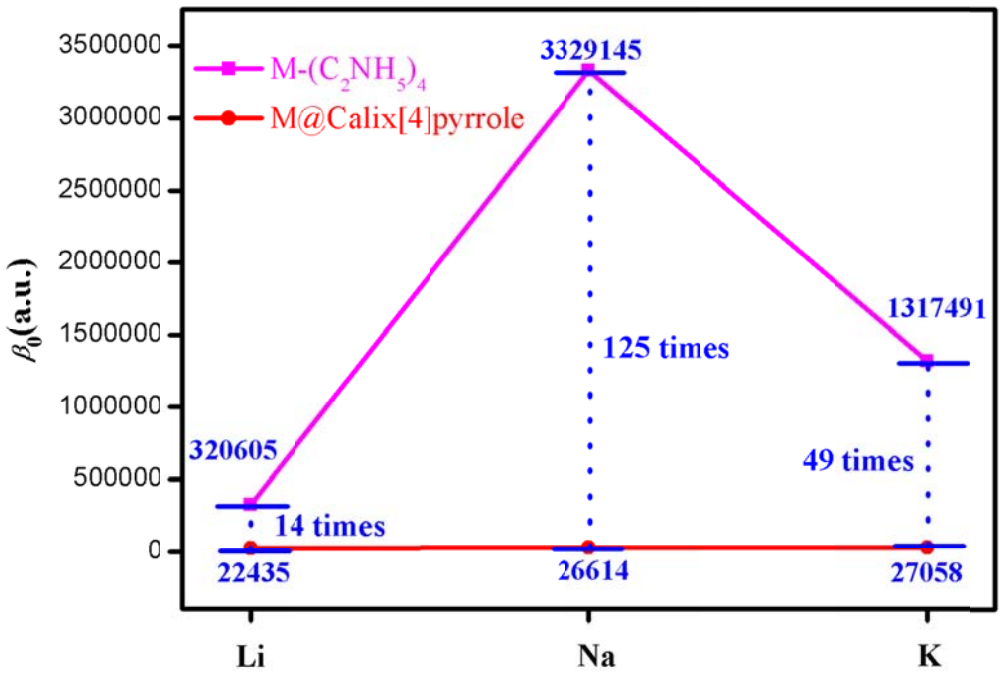

Fig.S3 The β_0 values of M-(C2NH5)$_4$ and M@Calix[4]pyrrole (M=Li, Na and K) calculated at the equal footing of theory.

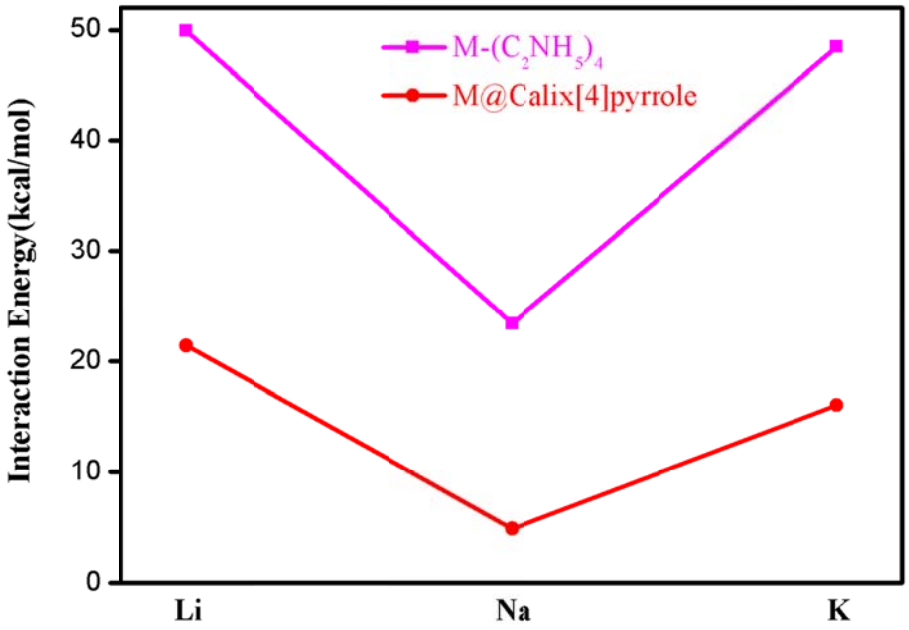

Fig.S4 The interaction energies of M-(C_2NH_5)$_4$ and M@Calix[4]pyrrole (M=Li, Na and K) calculated at the equal footing of theory.

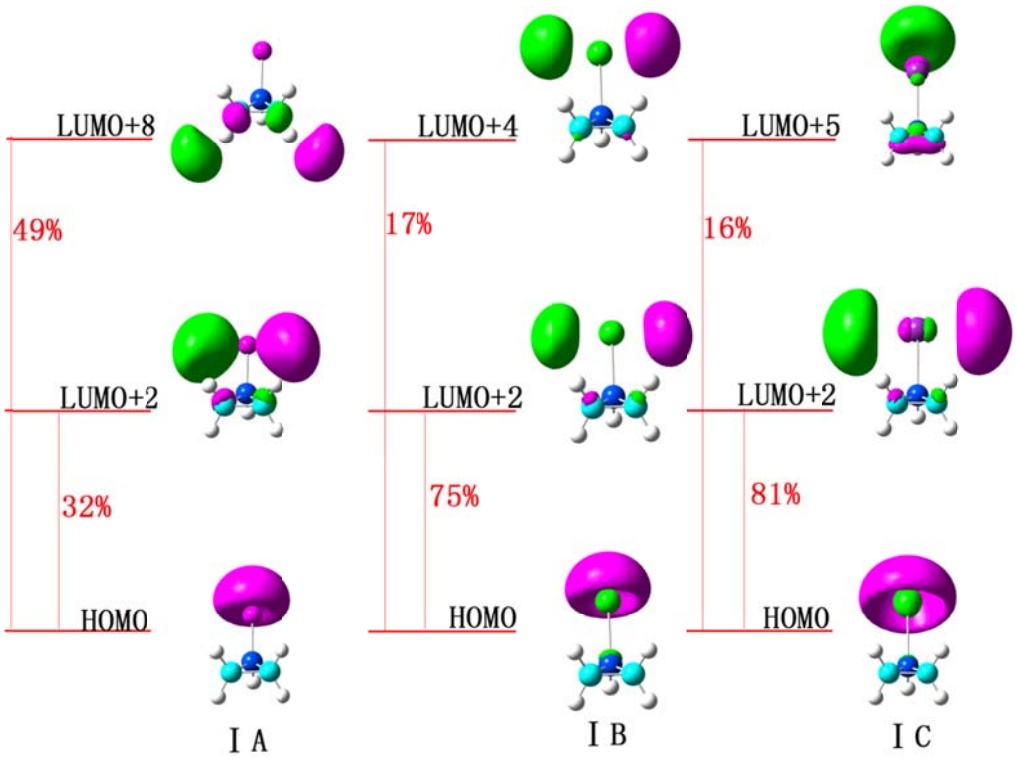

Fig.S5 The diagram of the LUMO orbitals and percentage of electron transition coefficient from HOMO for M-C_2NH_5(M=Li, Na and K).

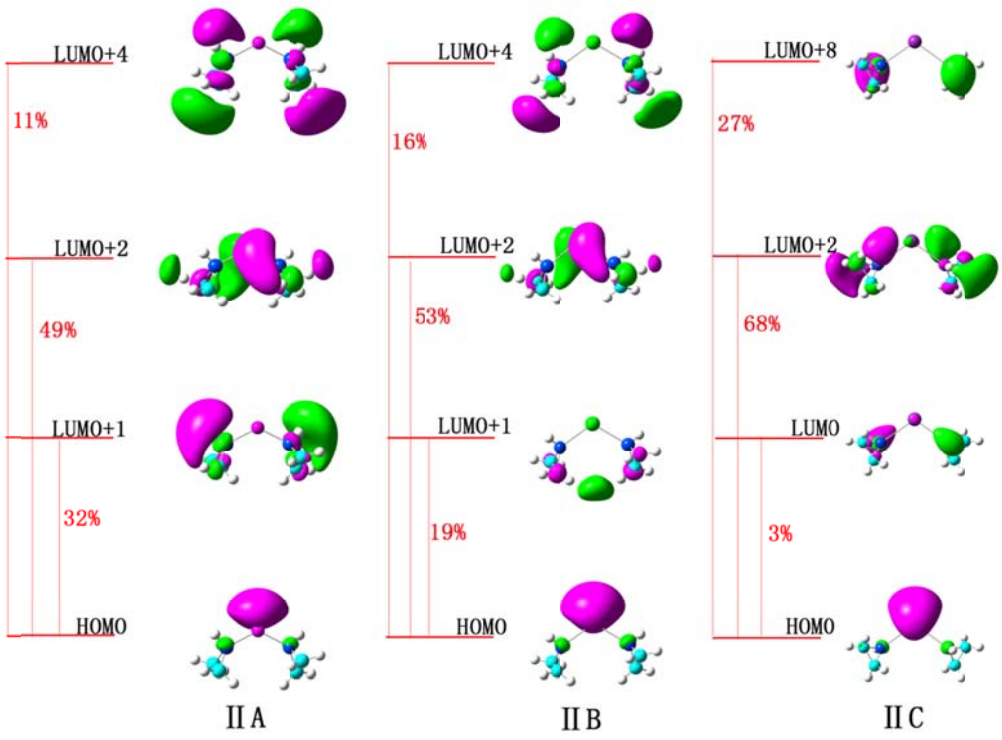

Fig.S6 The diagram of the LUMO orbitals and percentage of electron transition coefficient from HOMO for M-$(C_2NH_5)_2$(M=Li, Na and K).

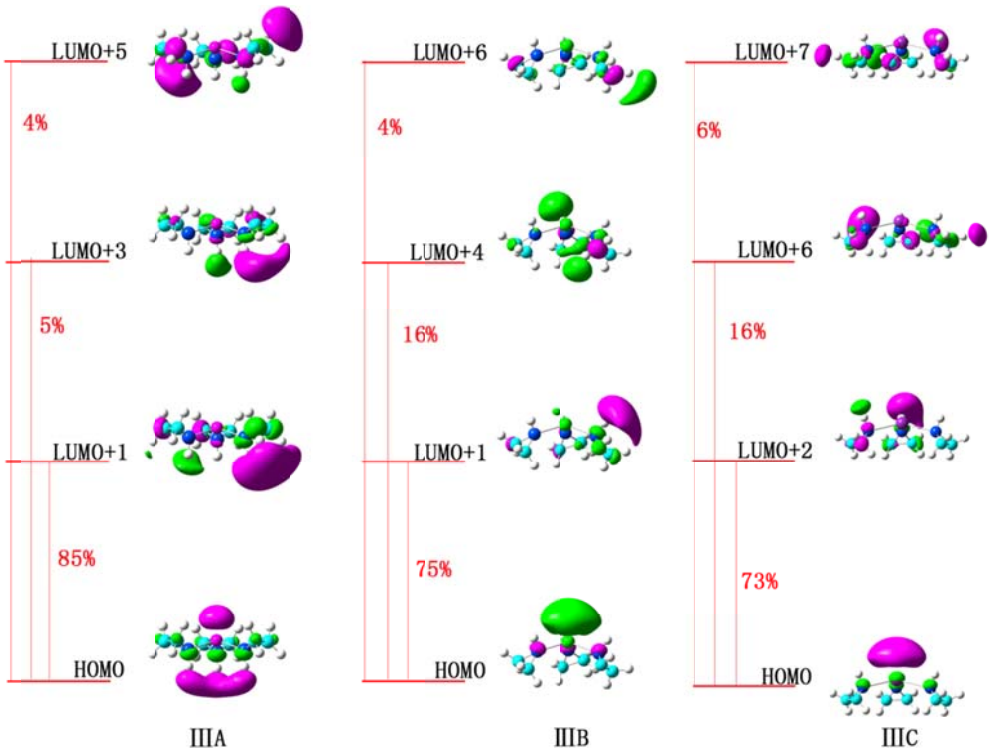

Fig.S7 The diagram of the LUMO orbitals and percentage of electron transition coefficient from HOMO for M-$(C_2NH_5)_3$(M=Li, Na and K).

Table S1. Cartesian coordinates (in angstrom) of all geometries optimized at the B3LYP/6-311++G(d,p) for alkali-doped aziridine M-$(C_2NH_5)_n$ (n=1-4, M=Li, Na and K),

Table S1.1: Li-C_2NH_5 .

Atom	X	Y	Z
C	-0.13244500	-0.65426700	0.73920600
C	-0.13244500	-0.65426700	-0.73920600
H	-1.04401600	-0.92382600	1.25921500
H	0.78574100	-0.86368200	1.27538400
H	-1.04401600	-0.92382600	-1.25921500
H	0.78574100	-0.86368200	-1.27538400
H	-1.06377400	1.04674800	0.00000000
N	-0.13244500	0.63483200	0.00000000
Li	1.36559100	1.97854900	0.00000000

Table S1.2: Na-C_2NH_5.

Atom	X	Y	Z
C	-0.55229200	-1.20532100	0.74016000
C	-0.55229200	-1.20532100	-0.74016000
H	-1.46160100	-1.48994000	1.25738200
H	0.36630600	-1.41484400	1.27687800
H	-1.46160100	-1.48994000	-1.25738200
H	0.36630600	-1.41484400	-1.27687800
H	-1.48408700	0.48647800	0.00000000
N	-0.55229200	0.07790300	0.00000000
Na	1.28802100	1.74923800	0.00000000

Table S1.3: K-C_2NH_5 .

Atom	X	Y	Z
C	-0.88997300	-1.67442800	0.74038800
C	-0.88997300	-1.67442800	-0.74038800
H	-1.79917300	-1.96292400	1.25631400
H	0.02747000	-1.88639400	1.27825000
H	-1.79917300	-1.96292400	-1.25631400
H	0.02747000	-1.88639400	-1.27825000
H	-1.82388700	0.01110000	0.00000000
N	-0.88997300	-0.39283100	0.00000000
K	1.17246200	1.60686800	0.00000000

Table S1.4: Li-$(C_2NH_5)_2$.

Atom	X	Y	Z
C	-2.21028600	-0.71163300	0.62533700
C	-2.52123700	-0.21761800	-0.73480700
H	-3.01565400	-0.78280600	1.34704200
H	-1.40235700	-1.41894500	0.77007900
H	-3.54364400	0.05957900	-0.96408000
H	-1.93956400	-0.56253100	-1.58128800
H	-2.40592300	1.35864500	0.60839700
N	-1.78512900	0.64898700	0.21624100
C	2.21016400	-0.71201000	-0.62494100
C	2.52142200	-0.21717400	0.73483300
H	3.01536400	-0.78359600	-1.34679200
H	1.40221700	-1.41942900	-0.76906700
H	3.54387700	0.06018600	0.96370000
H	1.93995800	-0.56159400	1.58165800
H	2.40576000	1.35828400	-0.60927900
N	1.78507100	0.64884500	-0.21655800
Li	-0.00000100	1.58599800	-0.00022800

Table S1.5: Na-$(C_2NH_5)_2$.

Atom	X	Y	Z
C	2.35678500	-1.07106000	-0.54871000
C	2.79006400	-0.43687800	0.71789100
H	3.10914200	-1.29928900	-1.29541900
H	1.49447400	-1.72787700	-0.55476100
H	3.84416900	-0.22191600	0.85279100
H	2.24260600	-0.63012300	1.63341000
H	2.68827800	0.95624900	-0.80673400
N	2.04903300	0.35355200	-0.29039000
C	-2.35782900	-1.06907700	0.55128600
C	-2.78840400	-0.43934900	-0.71845800
H	-3.11180300	-1.29484300	1.29711200
H	-1.49542500	-1.72572200	0.56153800
H	-3.84224600	-0.22504600	-0.85643100
H	-2.23889300	-0.63566800	-1.63209000
H	-2.69019000	0.95905000	0.80159600
N	-2.04973300	0.35467800	0.28872300
Na	0.00010000	1.72597800	-0.00012700

Table S1.6: K-$(C_2NH_5)_2$.

Atom	X	Y	Z
C	-3.58639300	-0.11207700	0.20725800
C	-2.81827300	-1.37844800	0.21896200
H	-4.48647300	-0.06048700	-0.39569400
H	-3.58511700	0.52674600	1.08358200
H	-3.18563400	-2.20923300	-0.37377800
H	-2.26023500	-1.65955700	1.10514600
H	-2.43931700	-0.29456100	-1.49856100
N	-2.28710200	-0.19712600	-0.49633100
C	3.58637500	-0.11223300	-0.21038100
C	2.81901300	-1.37911000	-0.21586000
H	4.48698700	-0.05747200	0.39149500
H	3.58389800	0.52276700	-1.08946800
H	3.18743400	-2.20708700	0.38014200
H	2.26032300	-1.66441300	-1.10028200
H	2.44103400	-0.28803600	1.49730800
N	2.28779700	-0.19502100	0.49479000
K	-0.00063700	1.47513500	0.00058000

Table S1.7: Li-$(C_2NH_5)_3$.

Atom	X	Y	Z
C	3.08137800	-0.85950400	-0.18136700
C	2.36483000	-2.15482700	-0.17926000
H	3.85659000	-0.69881400	0.56038600
H	3.20640400	-0.31684900	-1.11081500
H	2.64160000	-2.89510200	0.56395100
H	1.97036400	-2.55136100	-1.10724300
H	1.68354800	-0.92963400	1.34045800
N	1.68718000	-0.93326900	0.31420500
C	-0.79427900	3.09792500	-0.18265000
C	0.68581400	3.12240200	-0.17705300
H	-1.32169000	3.68967200	0.55805600
H	-1.32486200	2.93619900	-1.11341600
H	1.18788800	3.73109300	0.56753100
H	1.22837500	2.97836600	-1.10379900
H	-0.04012800	1.92059700	1.34060100
N	-0.03650700	1.92590000	0.31433900
C	-2.28787400	-2.23728300	-0.18002500
C	-3.04833200	-0.96724300	-0.17990800
H	-2.53866200	-2.98752300	0.56241200
H	-1.88073300	-2.61894500	-1.10876500
H	-3.82802500	-0.83408500	0.56260300
H	-3.19251900	-0.42798200	-1.10856100
H	-1.64775400	-0.99135700	1.34079600
N	-1.65202200	-0.99393100	0.31454800
Li	-0.00006100	-0.00133500	-0.28809000

Table S1.8: Na-$(C_2NH_5)_3$.

Atom	X	Y	Z
C	-2.38857900	-2.16510600	0.72870800
C	-1.34657900	-3.08871200	0.22235300
H	-3.42856100	-2.41209500	0.54559200
H	-2.20654800	-1.59094800	1.63005700
H	-1.66136800	-3.97664600	-0.31471700
H	-0.40680600	-3.18506200	0.75365100
H	-2.08006900	-1.90326700	-1.30319900
N	-1.55803900	-1.78063300	-0.43387400
C	-0.67906300	3.16882800	0.70146800
C	-2.00645300	2.68773200	0.25226800
H	-0.40475300	4.19625600	0.48875900
H	-0.22959300	2.75102200	1.59512200
H	-2.65467000	3.37940100	-0.27439200
H	-2.52094000	1.91914500	0.81733600
H	-0.67058300	2.73483900	-1.32412300
N	-0.78153700	2.23409200	-0.44077700
C	3.08120200	-0.99132000	0.71870200
C	3.34494200	0.38637200	0.24165000
H	3.82513800	-1.75552000	0.52209400
H	2.49162500	-1.14661900	1.61506900
H	4.27091700	0.58016400	-0.28841700
H	2.94607700	1.23240400	0.78925100
H	2.70524900	-0.81804900	-1.31120700
N	2.33039400	-0.44958400	-0.43540500
Na	0.00512200	-0.00172000	-1.08739700

Table S1.9: K-$(C_2NH_5)_3$.

Atom	X	Y	Z
C	-1.21140800	3.61061900	-0.51465100
C	0.25947800	3.78297600	-0.56242700
H	-1.80372500	4.37547200	-0.02384800
H	-1.71839400	3.08437600	-1.31550600
H	0.68914400	4.66778000	-0.10500800
H	0.82204000	3.38192100	-1.39788300
H	-0.34784800	3.10841600	1.29174500
N	-0.33876800	2.76666500	0.32926400
C	-2.54832400	-2.82864100	-0.57497100
C	-3.44280900	-1.64879400	-0.51881600
H	-2.88915000	-3.75497500	-0.12500500
H	-1.86874400	-2.95009900	-1.41088600
H	-4.40519600	-1.75530900	-0.02975700
H	-3.41364600	-0.91218400	-1.31374300
H	-2.48391600	-1.93782800	1.28627900
N	-2.23040500	-1.70011600	0.32588500
C	3.72916200	-0.76139300	-0.57421200
C	3.17519900	-2.13399300	-0.50725800
H	4.69922600	-0.57538600	-0.12576100
H	3.48488200	-0.12239600	-1.41526400
H	3.76026900	-2.90157700	-0.01205400
H	2.52826300	-2.49320100	-1.29956800
H	2.93114400	-1.14876200	1.29028000
N	2.59745500	-1.06029800	0.32902600
K	0.00262600	-0.01235000	0.91214500

Table S1.10: Li-$(C_2NH_5)_4$.

Atom	X	Y	Z
C	2.51754100	2.15309400	0.67169200
C	1.35034500	3.02243900	0.39552600
H	3.44574400	2.35798600	0.14692600
H	2.62982400	1.69336500	1.64669800
H	1.47095500	3.83153200	-0.31792100
H	0.61165600	3.19442200	1.16966900
H	1.64707700	1.66786600	-1.13278100
N	1.39048900	1.64507100	-0.13827300
C	-2.31382800	1.38544600	-1.69238400
C	-2.53153900	-0.05234100	-1.97394900
H	-2.25586700	2.06984200	-2.53318100
H	-2.68338700	1.81987000	-0.77068700
H	-2.62584200	-0.36305800	-3.00990500
H	-3.06258600	-0.66986000	-1.25865400
H	-0.57740400	0.49116900	-2.35021000
N	-1.20376200	0.42351300	-1.53601100
C	1.20394000	-2.94845800	-0.90137100
C	2.40426300	-2.29498400	-0.33027000
H	1.28832700	-3.41309400	-1.87864800
H	0.46651900	-3.39084600	-0.24177400
H	3.32012600	-2.30366500	-0.91323400
H	2.54095100	-2.26106000	0.74424500
H	1.53647500	-1.10917600	-1.77752100
N	1.28976500	-1.47458700	-0.84915900
C	-1.65077500	-1.45511700	2.19501400
C	-1.96631300	-0.02610000	2.41844300
H	-1.47529500	-2.08389600	3.06138700
H	-2.05152300	-1.96578200	1.32752300
H	-2.00965000	0.33710000	3.43972700
H	-2.59752600	0.50200300	1.71397100
H	0.05606100	-0.37988000	2.63783300
N	-0.63934000	-0.41713100	1.89207300
Li	0.12782900	0.01107600	0.00663800

Table S1.11: Na-$(C_2NH_5)_4$.

Atom	X	Y	Z
C	2.73296200	2.25156700	0.98359700
C	1.74959500	3.18527200	0.38812000
H	3.75433600	2.27889900	0.61745500
H	2.62136800	1.93546300	2.01446500
H	2.08939600	3.86177700	-0.38937000
H	0.92163900	3.54899600	0.98584500
H	2.11384100	1.58923400	-0.86869200
N	1.69771400	1.74491500	0.05655900
C	-1.93143200	1.29264800	-2.66400000
C	-2.05608500	-0.16765900	-2.87659100
H	-1.59826000	1.90562700	-3.49537800
H	-2.58142600	1.79321200	-1.95544200
H	-1.80933400	-0.56644900	-3.85538300
H	-2.79736400	-0.73307300	-2.32347400
H	-0.08291800	0.37921100	-2.63073200
N	-0.94197400	0.37026700	-2.06682300
C	1.33666000	-3.35417000	-0.51289700
C	2.41637800	-2.72769600	0.28403300
H	1.60288800	-3.83989500	-1.44604200
H	0.46587700	-3.76288900	-0.01312900
H	3.43124000	-2.77668400	-0.09750600
H	2.33204600	-2.67975200	1.36367500
H	1.89776700	-1.53892600	-1.31974100
N	1.45694000	-1.88130200	-0.45830800
C	-2.60427700	-0.99850300	2.30758500
C	-2.67023000	0.47789700	2.39403900
H	-2.69341100	-1.57178800	3.22415800
H	-2.95561100	-1.50225500	1.41452500
H	-2.80521700	0.93040600	3.37062400
H	-3.06952000	1.04887000	1.56383700
H	-0.81641500	-0.21567100	2.97687200
N	-1.37660800	-0.19067600	2.12417300
Na	0.02592200	-0.01298800	0.13190100

Table S1.12: K-$(C_2NH_5)_4$.

Atom	X	Y	Z
C	-2.21102000	3.26664700	-0.67036800
C	-1.43126600	3.68979900	0.51594000
H	-3.29423600	3.30493600	-0.61299600
H	-1.79867900	3.40220400	-1.66386000
H	-1.97432100	4.02297400	1.39456300
H	-0.45080900	4.13416300	0.38639600
H	-2.20264200	1.79385300	0.77251400
N	-1.52115700	2.25746600	0.16154600
C	0.83925600	-0.38641000	3.72620500
C	0.66342100	-1.80302200	3.33048400
H	0.23187700	-0.00178700	4.53937300
H	1.80603600	0.09059200	3.61043900
H	-0.06571500	-2.39950400	3.86958900
H	1.50194200	-2.35882200	2.92609300
H	-0.86408900	-0.63247700	2.59156400
N	0.15034700	-0.72222800	2.46218200
C	-2.22946100	-3.10437400	-1.30902500
C	-2.83236400	-1.99460900	-2.08314800
H	-2.87967200	-3.70239800	-0.67854800
H	-1.36216900	-3.62511900	-1.69905000
H	-3.89982600	-1.82273200	-1.98751900
H	-2.40429600	-1.70662000	-3.03669200
H	-2.58733300	-1.46810300	-0.10500400
N	-1.98849500	-1.70298800	-0.90435100
C	3.87371300	-0.60293200	-1.58127500
C	3.89642000	0.86203100	-1.36505100
H	4.33610700	-0.99714100	-2.48002000
H	3.88476900	-1.27342200	-0.72940500
H	4.37451300	1.48482200	-2.11372000
H	3.92410100	1.25730100	-0.35589100
H	2.43823900	0.33670500	-2.72455800
N	2.63203100	0.18748100	-1.73449300
K	0.15604900	0.02419400	-0.27263300

Table S2 The electronic properties (μ_0, α_0 and β_0 values) and interaction energy between alkali metal atom and Calix[4]pyrrole for M-Calix[4]pyrrole.

Compounds	μ_0	α_0	β_0	E_{int}
Li-Calix[4]pyrrole	1.15	389	22435	-49.12
Na-Calix[4]pyrrole	1.93	425	26614	-23.43
K-Calix[4]pyrrole	2.62	435	27058	-16.06