Supplementary Material

Obtaining excellent optical molecules by screening superalkali-doped cyclo[2*n*]carbon, $M_3O@C_{2n}$ (M = Li, Na, and K, *n* = 5-10)

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Table S1. Optimized Cartesian coordinates for studied $M_3O@C_{2n}$ at the ωB97XD/ 6-311+G(2d) level of theory

$Li_3O@C_{10}$ (charge = 0, spin multiplicity = 2)			
Atom	*x*	*y*	*z*
C	-1.99468917	0.47294619	0.20524408
C	-1.16433183	1.40965362	0.16373882
C	-0.03843763	2.11925883	0.16362562
C	1.10766573	1.53866446	0.12601042
C	2.13381982	0.73703077	0.17524987
C	1.87386714	-0.53990505	0.16362311
C	1.37782238	-1.72346341	0.23387214
C	0.06088297	-1.91503277	0.22464472
C	-1.17916455	-1.74035828	0.24558558
C	-2.18828742	-0.85403606	0.29112249
O	0.06956586	0.03183582	3.10122491
Li	0.29291644	1.60735319	2.54196904
Li	1.27623474	-1.02947158	2.58950891
Li	-1.42060694	-0.53501158	2.53188919

$Na_3O@C_{10}$ (charge = 0, spin multiplicity = 2)			
Atom	*x*	*y*	*z*
C	-2.00278910	0.46848998	0.06171547
C	-1.16924619	1.40267777	0.03949550
C	-0.04510717	2.11582160	0.04266955
C	1.09800823	1.52887384	0.01958166
C	2.12686837	0.73014662	0.06909737
C	1.86052376	-0.54577723	0.05517757
C	1.36918936	-1.73201336	0.11017391
C	0.05143765	-1.91835579	0.09837800
C	-1.18916279	-1.74727636	0.10069078
C	-2.19642492	-0.85902351	0.13030041
O	0.08852269	0.04661250	3.44667237
Na	1.57760008	-1.23483966	2.84424657
Na	-1.73740527	-0.65521562	2.79725780
Na	0.36529733	1.97284020	2.78663394

$K_3O@C_{10}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	-2.07949349	0.20088877	-0.09227738
C	-1.36698255	1.23147449	-0.10054346
C	-0.34332570	2.07948157	-0.10010874
C	0.86867187	1.66033742	-0.10705368
C	2.00487132	1.01908868	-0.03683149
C	1.90717826	-0.28253800	-0.03595577
C	1.57440510	-1.52250110	-0.00389078
C	0.29188853	-1.87833146	-0.02301998
C	-0.96059734	-1.87717758	-0.02529180
C	-2.08291165	-1.14218484	-0.02382212
O	0.11260840	0.04983317	3.54029921
K	0.71279529	2.21339027	2.96429852
K	-2.08052975	-0.51586565	2.99961268
K	1.66245961	-1.60782795	3.05833512

$Li_3O@C_{12}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.01958216	2.50050635	0.24442812
C	1.15683874	1.93887676	0.22954988
C	2.22749150	1.16413392	0.22857616
C	2.34364652	-0.09152903	0.14701535
C	2.23226348	-1.41456406	0.14607183
C	1.17723565	-2.11834568	0.12140983
C	-0.02874660	-2.65882306	0.12143390
C	-1.17535386	-2.12888398	0.08468080
C	-2.26480124	-1.37190810	0.13703140
C	-2.34748030	-0.10650934	0.16660017
C	-2.21268368	1.20713571	0.21853216
C	-1.18149306	1.93708600	0.19202666
O	-0.01133274	-0.16429804	2.81119355
Li	-0.16808105	1.46792820	2.40918470
Li	-1.33607839	-1.09885046	2.34079876
Li	1.47269232	-0.82522652	2.35165941

$Na_3O@C_{12}$ (charge = 0, spin multiplicity = 2)

atom	*x*	*y*	*z*
C	0.01700555	2.50958257	0.10664701
C	1.15219023	1.94243867	0.09969744
C	2.22481271	1.17068171	0.09072375
C	2.33574619	-0.08651450	0.02058192
C	2.23605235	-1.41014219	0.00168826
C	1.17783141	-2.11025784	-0.01032434
C	-0.02659700	-2.65413989	-0.01520651
C	-1.17182481	-2.11955837	-0.03761608
C	-2.26810532	-1.37180110	-0.00124712
C	-2.34507236	-0.10592964	0.04089552
C	-2.21349728	1.20818142	0.08780256
C	-1.17896683	1.93429547	0.07248416
O	-0.00936515	-0.17778281	3.25896938
Na	1.78129577	-1.00204040	2.68005834
Na	-1.63052334	-1.29314890	2.66781623
Na	-0.17728923	1.80656435	2.75291987

$K_3O@C_{12}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.12045190	2.25003757	0.48962237
C	1.23854945	1.65652564	0.41998126
C	2.29271501	0.86718295	0.29440475
C	2.35185310	-0.37749910	0.07375612
C	2.20740761	-1.68455850	-0.09500232
C	1.13162612	-2.35307321	-0.14816840
C	-0.08746363	-2.86386239	-0.20441485
C	-1.21178467	-2.28543625	-0.15153302
C	-2.28062305	-1.51318905	-0.01368984
C	-2.32157794	-0.26290779	0.19218386
C	-2.15676392	1.03691470	0.37485074
C	-1.09308419	1.72139594	0.41554731
O	0.06125844	-0.80800939	3.58452701
K	-0.43434543	1.45604183	3.44337708
K	-1.62447385	-2.26483316	2.93073212
K	2.22406404	-1.42013250	3.00220910

$Li_3O@C_{14}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.05875749	2.89022401	0.05252687
C	1.24337044	2.47549296	0.11509787
C	2.23623955	1.61965263	0.17459679
C	2.65919854	0.43871121	0.20822880
C	2.84482349	-0.86061508	0.30338299
C	2.06897556	-1.87572089	0.32425902
C	1.18718155	-2.82078361	0.38603835
C	-0.10545304	-2.80253759	0.31258099
C	-1.37375602	-2.65774491	0.28087465
C	-2.18319210	-1.62401951	0.18315304
C	-2.73840339	-0.49979973	0.14982714
C	-2.66885253	0.80978543	0.09711583
C	-2.14425564	1.95013713	0.04317445
C	-1.25250877	2.91195018	0.04418648
O	0.00379505	-0.06024498	2.59328672
Li	-0.58075456	1.34077468	1.84899927
Li	-1.00587528	-1.38358835	2.36000613
Li	1.65894338	-0.25986253	2.37774523

$Na_3O@C_{14}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.00476317	3.02424978	-0.17622310
C	1.19078517	2.61508430	-0.07776224
C	2.15545083	1.73269462	0.04593667
C	2.57077049	0.55279790	0.14031458
C	2.78275534	-0.74317772	0.24719651
C	2.01326635	-1.76371805	0.25782256
C	1.14811011	-2.72494455	0.32103982
C	-0.14579967	-2.68099622	0.24716724
C	-1.41188367	-2.52433455	0.22554144
C	-2.20392564	-1.47572340	0.11718474
C	-2.76408702	-0.35874084	0.02267033
C	-2.73044942	0.94992376	-0.09550036
C	-2.20166723	2.08661752	-0.18755012
C	-1.30219659	3.03733518	-0.21026475

O	0.16536359	-0.45669071	3.08587805
Na	2.20182303	-0.62589391	3.02416827
Na	-1.12016490	-2.04321348	3.01078754
Na	-0.43968741	0.98325444	1.72900470

$K_3O@C_{14}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.04604281	3.06588914	-0.34811009
C	1.22129966	2.64142090	-0.20290072
C	2.15602092	1.73269616	-0.03111845
C	2.53079214	0.54348131	0.10166980
C	2.69226436	-0.75791016	0.23720802
C	1.89237335	-1.75282367	0.24778640
C	0.99573418	-2.68783099	0.30304975
C	-0.29491572	-2.58282081	0.18250473
C	-1.54731793	-2.36577649	0.12445670
C	-2.33498315	-1.32076982	-0.03185874
C	-2.86115776	-0.19328171	-0.18192749
C	-2.80692200	1.11104559	-0.33366260
C	-2.19908154	2.21105158	-0.42379298
C	-1.25653232	3.11102698	-0.43450985
O	0.33540063	-0.79630869	3.37148066
K	-0.49177518	0.85994597	1.91111502
K	-0.99210819	-2.67818505	3.35215533
K	2.63773360	-0.79961822	3.42333226

$Li_3O@C_{16}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.61449358	3.23727814	0.13636764
C	-0.61826173	3.22399394	0.14985538
C	-1.85337627	2.69318591	0.17753475
C	-2.70888097	1.81145628	0.21130797
C	-3.21721395	0.56852487	0.24361612
C	-3.25292276	-0.66226764	0.24299639
C	-2.73385215	-1.90583811	0.26870111
C	-1.83744451	-2.74905233	0.31200279
C	-0.59386547	-3.25788682	0.32087229

C	0.63462780	-3.26697320	0.31186346
C	1.87917428	-2.75883868	0.28769637
C	2.76404730	-1.90184646	0.24995281
C	3.25998675	-0.64964622	0.23621767
C	3.21676751	0.58022364	0.23694035
C	2.69875331	1.81842070	0.20937152
C	1.85289566	2.71062849	0.16521290
O	0.02043541	0.03155507	2.20199931
Li	0.18131119	1.67364916	1.86252848
Li	-1.48159182	-0.66272270	1.88387334
Li	1.38075152	-0.91245796	1.89075197

$Na_3O@C_{16}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.61739963	3.18020333	-0.02108004
C	-0.61591152	3.25509008	-0.01013969
C	-1.82032036	2.66273975	0.01811987
C	-2.72358982	1.82582010	0.07597598
C	-3.17995123	0.56657621	0.09080902
C	-3.27993855	-0.66372361	0.09595591
C	-2.70731825	-1.87818631	0.09233230
C	-1.87317867	-2.78718415	0.14297103
C	-0.60549384	-3.22507552	0.15732178
C	0.62514935	-3.28936852	0.16895115
C	1.84003987	-2.72537181	0.13001873
C	2.77667146	-1.92117232	0.08874311
C	3.21788756	-0.65287346	0.05166515
C	3.25989434	0.57994223	0.06732871
C	2.68513272	1.79013483	0.04180938
C	1.87675347	2.72091699	0.01478864
O	0.02352626	0.04706959	2.92907177
Na	1.79415896	-0.87630068	2.46148106
Na	-1.65547941	-1.03576155	2.46612792
Na	-0.07502362	2.03069689	2.41955273

$K_3O@C_{16}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*

C	0.69918055	3.02661576	0.31931205
C	-0.53072437	3.04640433	0.33603799
C	-1.78521995	2.56344621	0.31932595
C	-2.68359013	1.72422614	0.27406847
C	-3.23206247	0.49737555	0.20967403
C	-3.27975565	-0.73123737	0.12542928
C	-2.79585348	-1.98680866	0.05926196
C	-1.93446290	-2.86777814	0.03246930
C	-0.70356385	-3.41007401	0.00320139
C	0.52642525	-3.43094200	0.00269505
C	1.78087854	-2.94787917	0.01888146
C	2.67939418	-2.10809191	0.04669413
C	3.22768241	-0.88056543	0.09904134
C	3.27500162	0.35030748	0.13828540
C	2.79077906	1.60572852	0.20525082
C	1.92983006	2.48449230	0.27809163
O	0.01285887	-0.30358443	2.33721194
K	-0.00264707	-0.18268072	-0.01141211
K	-1.98913592	-1.10453672	3.09442433
K	2.02556675	0.41722936	3.14494113

$Li_3O@C_{18}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.60176800	-3.68923034	0.13430803
C	-0.62992880	-3.84605227	0.21968158
C	-1.83214907	-3.27392270	0.31601379
C	-2.84423317	-2.56974677	0.36864050
C	-3.43788443	-1.35876342	0.41138118
C	-3.61953502	-0.14779537	0.44393313
C	-3.37066494	1.17704536	0.42650192
C	-2.75055205	2.22865963	0.37593895
C	-1.76891627	3.14694733	0.29146283
C	-0.61337941	3.55237672	0.18462623
C	0.72061548	3.66853252	0.08432527
C	1.81572525	3.08078830	0.03148769
C	2.88858309	2.31338793	-0.05855037
C	3.36316146	1.15111415	-0.08432344

C	3.65554380	-0.11840367	-0.13284067
C	3.44474912	-1.36658931	-0.14023137
C	2.76433956	-2.47588253	-0.07899707
C	1.87097651	-3.35976436	0.00801949
O	0.10647567	-0.06977107	1.51186705
Li	1.57279843	-0.81291362	1.14550244
Li	-1.29491234	-0.99090317	1.40964944
Li	0.04841889	1.59924442	1.32125522

$Na_3O@C_{18}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.60291993	-3.67792284	-0.03165314
C	-0.62342965	-3.83513472	0.06596856
C	-1.80689304	-3.21173774	0.12931139
C	-2.79261442	-2.47708264	0.19609162
C	-3.34048953	-1.24557414	0.24618956
C	-3.53580229	-0.03857992	0.30414705
C	-3.30799635	1.28884200	0.29952390
C	-2.73624528	2.37054388	0.24650387
C	-1.81334781	3.34424963	0.15924450
C	-0.65147889	3.74416958	0.04328749
C	0.67172464	3.88384588	-0.06155788
C	1.74531224	3.23833992	-0.14633503
C	2.71371331	2.36494352	-0.21039527
C	3.23680612	1.21511410	-0.24764731
C	3.44427797	-0.06207913	-0.27734256
C	3.40410979	-1.33036192	-0.29113521
C	2.68129328	-2.41929542	-0.24323473
C	1.89671151	-3.39642785	-0.13151648
O	0.23325466	-0.42935419	2.26715270
Na	2.10307895	-1.25233257	2.24574413
Na	-1.51421494	-1.45283346	2.50379520
Na	0.08001133	1.21746784	1.03697027

$K_3O@C_{18}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	1.87241850	-3.11844435	0.00051734

C	0.64774657	-3.42160453	-0.00356869
C	-0.64784364	-3.42166204	0.00499304
C	-1.87239242	-3.11864754	0.02526909
C	-2.86198024	-2.26598618	0.02588179
C	-3.70629099	-1.33496574	0.04840758
C	-3.76164608	-0.00097860	0.01859342
C	-3.63297855	1.22779768	0.03307999
C	-2.82842619	2.30521802	0.00603170
C	-1.89488721	3.09739240	-0.00054697
C	-0.61079311	3.50566910	-0.01995162
C	0.61063497	3.50568260	-0.02811993
C	1.89488659	3.09744558	-0.02588134
C	2.82840263	2.30523593	-0.03175625
C	3.63332214	1.22787502	-0.01537915
C	3.76173930	-0.00091030	-0.03150092
C	3.70658016	-1.33487944	-0.00085842
C	2.86195444	-2.26582283	-0.01209365
O	0.01302976	-0.12041910	1.97571298
K	2.15532689	-0.31066542	2.78706672
K	-2.11806640	-0.31112853	2.81579249
K	-0.00262981	0.08606665	-0.34682696

$Li_3O@C_{20}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.62025723	3.98118557	0.36778921
C	-0.60443741	4.03555286	0.36006546
C	-1.86869668	3.55134009	0.35362910
C	-2.83785245	2.81160751	0.34989562
C	-3.61492252	1.70386089	0.34838370
C	-3.99200025	0.54691366	0.34951977
C	-4.01246319	-0.80662734	0.35379878
C	-3.64697965	-1.97153229	0.35943891
C	-2.89571269	-3.09686819	0.36814183
C	-1.84794645	-3.73617173	0.37705548
C	-0.59309646	-4.21070730	0.38676836
C	0.63406547	-4.09648331	0.39420466
C	1.91560253	-3.74083230	0.40113172

C	2.89224992	-2.97617904	0.40469956
C	3.69423277	-1.92208720	0.40636364
C	4.06581711	-0.73306232	0.40477615
C	4.00746574	0.59035126	0.40016780
C	3.68641950	1.78612369	0.39435592
C	2.84417688	2.81852841	0.38563839
C	1.91856440	3.63060237	0.37716528
O	0.02378150	-0.09996095	0.37698589
Li	-0.12298551	1.58581366	0.37055461
Li	-1.31535150	-1.13387550	0.37199755
Li	1.57773682	-0.78627987	0.38918993

$Na_3O@C_{20}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.60386058	4.02098149	0.04403545
C	-0.62678211	3.99970835	0.05180697
C	-1.88729167	3.54305353	0.07829816
C	-2.84759467	2.77688684	0.10244765
C	-3.62286919	1.68537738	0.10622779
C	-4.02775674	0.52581805	0.08642708
C	-4.05144762	-0.81436778	0.06262134
C	-3.68443053	-1.98894473	0.04677926
C	-2.87118438	-3.05643994	0.06126968
C	-1.84132485	-3.72760529	0.09294609
C	-0.55913300	-4.11373041	0.10348640
C	0.66889589	-4.11330807	0.09735348
C	1.94818245	-3.71645816	0.07706493
C	2.96426621	-3.02365616	0.04559322
C	3.74728734	-1.93339672	0.04268487
C	4.07874547	-0.74874136	0.06642002
C	4.03530023	0.59035576	0.08893553
C	3.62714575	1.74849300	0.10041312
C	2.84320359	2.83406835	0.09085793
C	1.87432808	3.59019751	0.06315836
O	0.02162695	-0.10508590	2.10818208
Na	1.70986886	-1.22020041	1.78982659
Na	-1.78709588	-1.01427165	1.79706928

Na	0.14215183	1.91200673	1.77333646

$K_3O@C_{20}$ (charge = 0, spin multiplicity = 2)			
atom	*x*	*y*	*z*
C	0.59480940	4.00296693	0.22727765
C	-0.63344509	3.95615230	0.23594317
C	-1.88961408	3.49095786	0.25442622
C	-2.85582324	2.73138376	0.27015658
C	-3.63026015	1.63858582	0.29498824
C	-4.01237274	0.46946253	0.30921038
C	-4.01883450	-0.87035847	0.35464787
C	-3.60663841	-2.02733298	0.42045470
C	-2.80366632	-3.09726818	0.48260150
C	-1.78818261	-3.78649976	0.51903072
C	-0.50884730	-4.17814723	0.51854244
C	0.71836146	-4.17542256	0.48125267
C	1.98264550	-3.73860417	0.41749468
C	2.97267846	-3.01209446	0.34883054
C	3.71749619	-1.89842342	0.30108963
C	4.05525942	-0.71572667	0.28633450
C	4.02337819	0.62333022	0.26236148
C	3.64721848	1.79339050	0.24823050
C	2.86641532	2.88186255	0.23209862
C	1.87599117	3.60976542	0.22546835
O	-0.02299803	-0.31577018	2.10421636
K	0.28252167	0.72768675	0.04216345
K	-2.19139649	-0.28060438	2.88600009
K	1.81610592	-1.47002054	2.87869106

Table S2. Root-mean-square-deviations of cyclocarbon units (RMSD, in Å), molecular planarity parameters of cyclocarbon units (MPP, in Å), spans of deviation from plane of cyclocarbon units (SDP, in Å), average bond lengths of superalkali units ($r_{M\text{-}O}^{ave}$, in Å), distances between O atom and ring plane (d, in Å), and angles between the alkali-metal plane and the ring plane (θ, in°) in $M_3O@C_{2n}$

	RMSD	MPP	SDP	$r_{M\text{-}O}^{ave}$	d	θ
$Li_3O@C_{10}$	0.150	0.021	0.057	1.69	2.904	2.1
$Na_3O@C_{10}$	0.149	0.017	0.047	2.06	3.377	1.1
$K_3O@C_{10}$	0.152	0.014	0.046	2.32	3.599	0.1
$Li_3O@C_{12}$	0.030	0.018	0.049	1.69	2.642	0.1
$Na_3O@C_{12}$	0.030	0.013	0.036	2.05	3.222	0.0
$K_3O@C_{12}$	0.032	0.018	0.043	2.32	3.482	0.0
$Li_3O@C_{14}$	0.142	0.016	0.065	1.68	2.403	9.5
$Na_3O@C_{14}$	0.157	0.017	0.050	2.05	3.087	22.3
$K_3O@C_{14}$	0.149	0.018	0.054	2.32	3.599	20.0
$Li_3O@C_{16}$	0.027	0.014	0.045	1.68	1.968	0.8
$Na_3O@C_{16}$	0.045	0.019	0.066	2.05	2.855	0.5
$K_3O@C_{16}$	0.022	0.014	0.038	2.31	2.186	90.0
$Li_3O@C_{18}$	0.074	0.015	0.051	1.68	1.360	1.2
$Na_3O@C_{18}$	0.152	0.018	0.057	2.05	2.315	27.8
$K_3O@C_{18}$	0.146	0.010	0.032	2.31	1.980	82.6
$Li_3O@C_{20}$	0.046	0.000	0.001	1.69	0.000	0.0
$Na_3O@C_{20}$	0.046	0.021	0.061	2.05	2.033	0.4
$K_3O@C_{20}$	0.046	0.031	0.103	2.31	1.782	57.7

Table S3. Vertical ionization potentials (E_{VIP}, in eV), NPA charges on the superalkalis (Q_M, in |e|), dipole moments (μ_0, in D), frontier molecular orbital energies ($E_{\alpha HOMO}/E_{\beta HOMO}$ and $E_{\alpha LUMO}/E_{\beta LUMO}$, in eV), and αHOMO-αLUMO/βHOMO-βLUMO gaps ($\Delta E_{\alpha gap}/\Delta E_{\beta gap}$, in eV) in $M_3O@C_{2n}$

	E_{VIP}	Q_M	μ_0	$E_{\alpha HOMO}/E_{\beta HOMO}$	$E_{\alpha LUMO}/E_{\beta LUMO}$	$\Delta E_{\alpha gap}/\Delta E_{\beta gap}$
$Li_3O@C_{10}$	7.21	0.977	3.44	-7.17/-8.07	-0.80/-1.07	6.37/7.00
$Na_3O@C_{10}$	6.80	0.985	5.01	-6.45/-6.45	-0.35/-0.73	6.10/5.72
$K_3O@C_{10}$	6.35	0.995	7.97	-5.90/-5.90	0.04/-0.27	5.93/5.62
$Li_3O@C_{12}$	6.96	0.983	3.28	-6.88/-7.49	-1.22/-2.17	5.66/5.32
$Na_3O@C_{12}$	6.49	0.991	5.06	-6.43/-6.48	-0.84/-1.77	5.59/4.71
$K_3O@C_{12}$	5.98	1.000	8.03	-5.89/-5.91	-0.46/-1.35	5.44/4.56
$Li_3O@C_{14}$	6.86	0.978	2.77	-6.79/-8.13	-1.87/-1.87	4.92/6.25
$Na_3O@C_{14}$	6.56	0.984	5.70	-6.48/-6.49	-1.49/-1.58	5.00/4.90
$K_3O@C_{14}$	6.19	0.991	8.58	-5.93/-5.94	-1.15/-1.30	4.78/4.64
$Li_3O@C_{16}$	6.37	0.962	2.22	-6.32/-7.72	-2.26/-2.26	4.06/5.46
$Na_3O@C_{16}$	6.12	0.985	4.23	-6.09/-6.53	-1.96/-2.02	4.13/4.51
$K_3O@C_{16}$	5.66	0.974	5.83	-5.65/-5.67	-1.71/-1.64	3.94/4.03
$Li_3O@C_{18}$	6.45	0.966	1.95	-6.54/-8.08	-2.17/-2.17	4.37/5.91
$Na_3O@C_{18}$	6.26	0.986	4.43	-6.34/-6.58	-1.94/-1.93	4.40/4.65
$K_3O@C_{18}$	5.92	0.984	5.41	-5.92/-5.92	-1.60/-1.66	4.32/4.26
$Li_3O@C_{20}$	6.28	0.969	1.56	-6.38/-7.83	-2.63/-2.56	3.75/5.27
$Na_3O@C_{20}$	5.96	0.981	3.09	-5.96/-6.60	-2.39/-2.34	3.57/4.26
$K_3O@C_{20}$	5.56	0.985	5.12	-5.57/-5.97	-2.04/-1.98	3.53/3.98

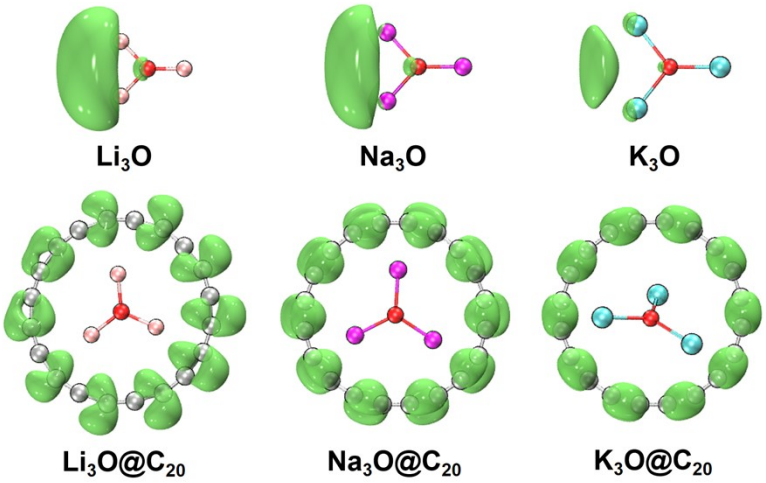

Figure S1. Excess electron isosurfaces (isovalue = 0.001 a.u.) of M_3O (top panel) and $M_3O@C_{20}$ (bottom panel).

Table S4. Interaction energies (ΔE_{int}, in kcal·mol^{-1}) and its components (ΔE_{els}, ΔE_{xrep}, ΔE_{orb}, and ΔE_{disp}, in kcal·mol^{-1}) between M_3O^+ and C_{20}^- in $M_3O@C_{20}$ calculated by the sobEDAw method

	ΔE_{int}	ΔE_{els}	ΔE_{xrep}	ΔE_{orb}	ΔE_{disp}
$Li_3O@C_{10}$	-115.4	-106.3	27.5	-20.7	-16.0
$Na_3O@C_{10}$	-102.2	-99.4	25.1	-11.8	-16.2
$K_3O@C_{10}$	-89.3	-87.5	22.7	-8.3	-16.3
$Li_3O@C_{12}$	-116.5	-102.7	26.4	-22.8	-17.4
$Na_3O@C_{12}$	-102.7	-96.3	23.4	-12.5	-17.3
$K_3O@C_{12}$	-90.2	-85.7	22.5	-9.1	-17.9
$Li_3O@C_{14}$	-111.3	-92.5	22.5	-23.5	-17.9
$Na_3O@C_{14}$	-98.7	-89.4	23.4	-13.3	-19.5
$K_3O@C_{14}$	-88.7	-81.9	23.5	-9.7	-20.6
$Li_3O@C_{16}$	-116.5	-92.4	20.7	-26.5	-18.4
$Na_3O@C_{16}$	-101.5	-87.8	18.9	-14.2	-18.3
$K_3O@C_{16}$	-92.5	-81.3	26.9	-10.9	-27.2
$Li_3O@C_{18}$	-111.8	-84.4	19.4	-28.1	-18.8
$Na_3O@C_{18}$	-95.7	-79.7	18.0	-14.0	-20.0
$K_3O@C_{18}$	-88.5	-72.3	16.9	-8.4	-24.8
$Li_3O@C_{20}$	-109.5	-79.9	14.0	-26.0	-17.6
$Na_3O@C_{20}$	-99.2	-80.6	16.5	-16.0	-19.2
$K_3O@C_{20}$	-87.2	-72.1	16.6	-8.9	-22.7

Table S5. Isotropic polarizabilities (α_0, in a.u.) and its Cartesian components (α_{xx}, α_{yy}, and α_{zz}, in a.u.) of the $M_3O@C_{2n}$

	α_0	α_{xx}	α_{yy}	α_{zz}
$Li_3O@C_{10}$	149	182	169	97
$Na_3O@C_{10}$	171	203	191	118
$K_3O@C_{10}$	187	218	208	134
$Li_3O@C_{12}$	181	218	218	107
$Na_3O@C_{12}$	202	238	238	129
$K_3O@C_{12}$	219	253	251	152
$Li_3O@C_{14}$	234	281	305	115
$Na_3O@C_{14}$	253	298	323	139
$K_3O@C_{14}$	271	317	337	159
$Li_3O@C_{16}$	279	357	358	121
$Na_3O@C_{16}$	298	374	374	146
$K_3O@C_{16}$	309	390	372	163
$Li_3O@C_{18}$	345	424	484	128
$Na_3O@C_{18}$	361	422	507	153
$K_3O@C_{18}$	374	534	421	167
$Li_3O@C_{20}$	417	537	577	136
$Na_3O@C_{20}$	455	602	603	159
$K_3O@C_{20}$	464	613	602	178

Table S6. First hyperpolarizabilities (β_0, in a.u.) and its Cartesian components (β_x, β_y, and β_z, in a.u.) of the $M_3O@C_{2n}$

	β_0	β_x	β_y	β_z
$Li_3O@C_{10}$	647	329	99	-549
$Na_3O@C_{10}$	1283	262	65	-1254
$K_3O@C_{10}$	1911	201	104	1898
$Li_3O@C_{12}$	425	-11	14	-425
$Na_3O@C_{12}$	1054	-5	12	-1054
$K_3O@C_{12}$	293	6	41	290
$Li_3O@C_{14}$	1215	470	-1083	-288
$Na_3O@C_{14}$	1325	156	-343	-1270
$K_3O@C_{14}$	1451	67	-287	1421
$Li_3O@C_{16}$	116	61	68	-71
$Na_3O@C_{16}$	1070	-12	-10	-1070
$K_3O@C_{16}$	1784	-8	94	1782
$Li_3O@C_{18}$	1897	1840	-310	-345
$Na_3O@C_{18}$	3385	1536	-2819	-1073
$K_3O@C_{18}$	1530	-4	-710	-1355
$Li_3O@C_{20}$	10542	9811	-3856	75
$Na_3O@C_{20}$	202	20	-87	181
$K_3O@C_{20}$	1211	108	485	-1105

Table S7. First hyperpolarizabilities (β_0, in a.u.) and its Cartesian components (β_x, β_y, and β_z, in a.u.) of the $M_3O@C_{24}$

	β_0	β_x	β_y	β_z
$Li_3O@C_{24}$	19093	1731	19014	7
$Na_3O@C_{24}$	28414	-15195	24009	-236
$K_3O@C_{24}$	27217	-17659	20706	-424

Table S8. Response properties (α_0 and β_0, in a.u.) of $M_3O@C_{20}$ calculated at different theoretical levels

	α_0		β_0	
	CAM-B3LYP	BHandHLYP	CAM-B3LYP	BHandHLYP
$Li_3O@C_{20}$	417	409	10357	10850
$Na_3O@C_{20}$	443	448	285	513
$K_3O@C_{20}$	455	459	998	862

Table S9. Isotropic polarizabilities (α_0, in a.u.) and first hyperpolarizabilities (β_0, in a.u.) of the $Li_3O@C_{20}$ in zero-frequency limit ($\lambda = \infty$ nm) and under frequency-dependent fields (λ = 1907, 1460, 1340, 1180, and 1064 nm)

λ	α_0	β_0
∞ nm	417	10542
1907 nm	419	28449
1460 nm	419	305597
1340 nm	427	34582
1180 nm	438	48098
1064 nm	450	55937