Supporting Information

for

Hunting for organic molecules with artificial intelligence: Molecules optimized for desired excitation energies

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1. Materials

Compound **I** (2-methyl-oxazole) is commercially available and was purchased from J&W Pharmlab LLC (catalog No. 56R0594). Compound **II** (1,2-dimethy-1H-imidazol-5-ol), compound **III** (4-methyl-6-quinolinol), compound **IV** (5-methylnaphtalene-2-ol) and compound **VI** (1-(dimethylamino)-2,3-butanedione) were obtained from Tokyo Chemical Industry Co., Ltd. (TCI) upon custom synthesis. Compound **V** (N-(2-hydroxybenzyl)-N-methylnitrous amide) was obtained from HeBei Sundia Meditech Company, Ltd. upon custom synthesis. All chemical compounds obtained by custom synthesis satisfy reagent-grade purity (> 96 %), and were used as received.

2. Characterization

Compound II: ^{1}H-NMR (in $CDCl_3$) in ppm: 4.04 (q, 2H, CH_2), 3.05 (s, 3H, CH_3), 2.20 (t, 3H, CH_3). ^{13}C-NMR (in $CDCl_3$) in ppm: 181.2, 163.2, 58.3, 26.4, 15.9. LC-MS (m/z): calculated for [$C_5H_8N_2O$] = 112.06 m/z, found 113.3 m/z ($M+H^+$). Purity (GC): 98.9%. Note that ^{1}H- and ^{13}C-NMR spectra indicate that compound **II** mainly exist as keto-form in tautomerism.

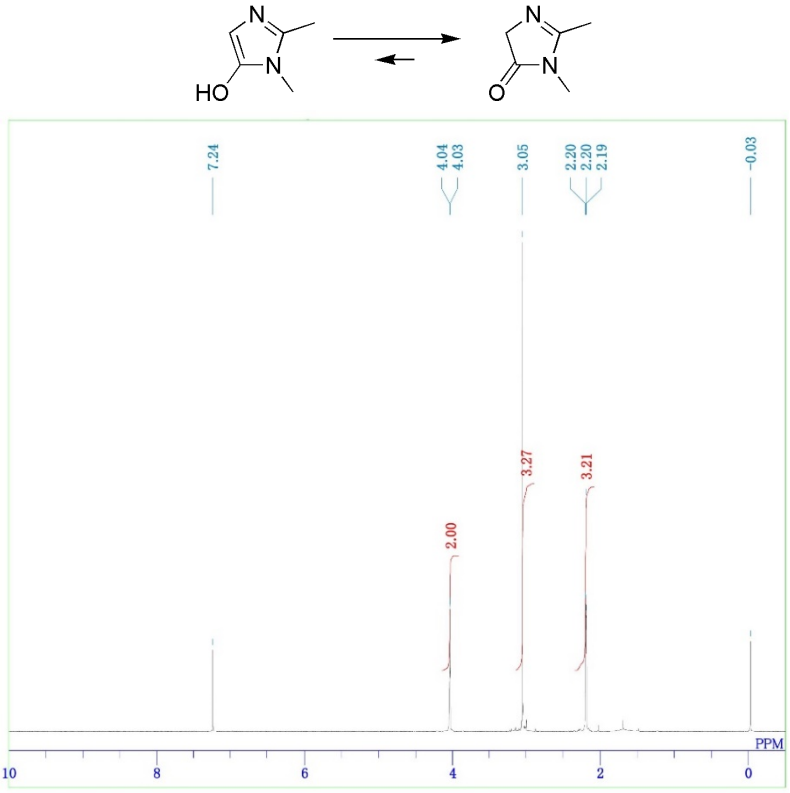


Figure S1. ^{1}H-NMR spectrum of compound **II** (as prepared). This chart is measured by TCI that performed custom synthesis and shown wih permission from TCI.

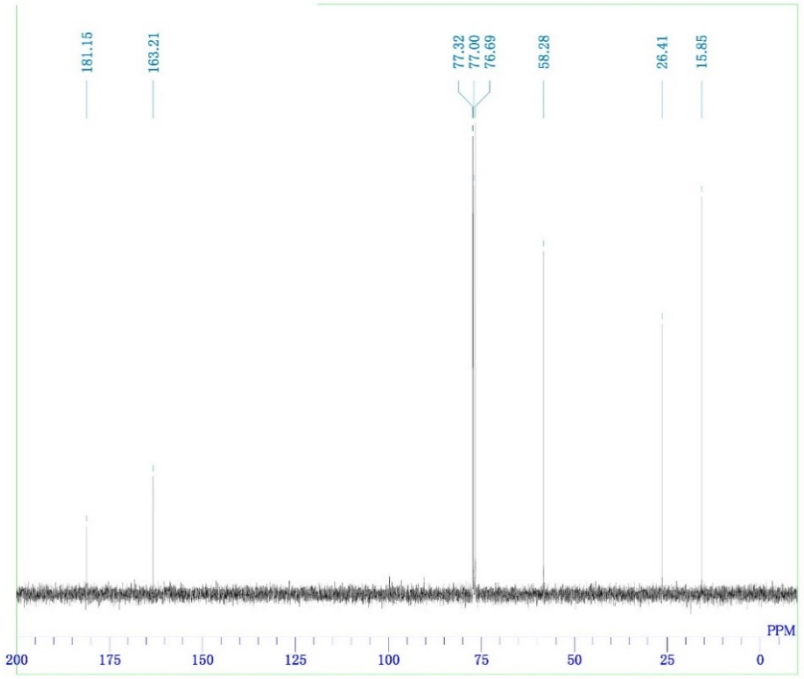


Figure S2. ^{13}C-NMR spectrum of compound **II** (as prepared). This chart is measured by TCI that performed custom synthesis and shown with permission from TCI.

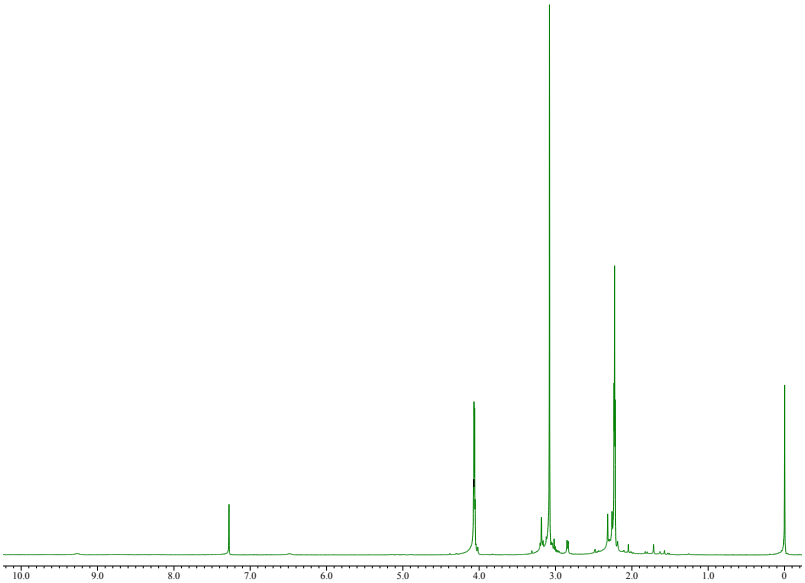


Figure S3. ^{1}H-NMR spectrum of compound **II** measured in two weeks after the reagent bottle was opened in air for taking out sample. After the bottle was opened, the bottle was securely sealed and stored at −20 °C for two weeks. Compared with ^{1}H-NMR spectra of as-prepared compound, signals from some impurities are observed beside main signals around 2.0−3.5 ppm. This chart is measured by authors using AL300 BX NMR spectrometer (JEOL, Tokyo, Japan).

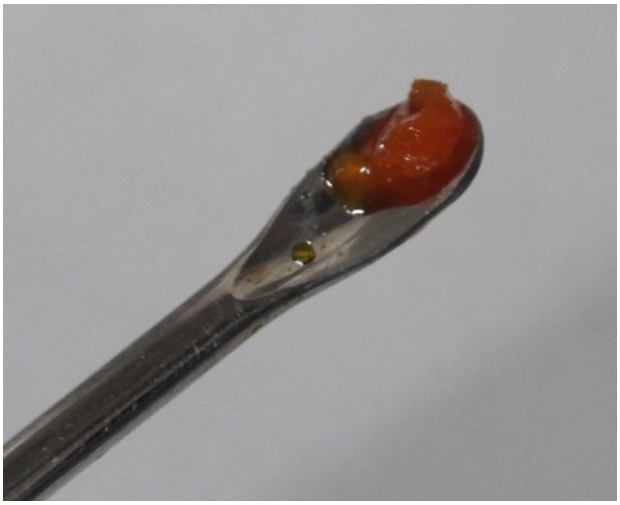


Figure S4. Photograph of compound **II**.

Compound III: ^{1}H-NMR (in DMSO-d_6) in ppm: 10.04 (s, 1H, OH), 8.56 (s, 1H, ArH), 7.89 (d, 1H, ArH), 7.34-7.25 (m, 3H, ArH), 2.60 (s, 3H, CH_3). ^{13}C-NMR (in DMSO-d_6) in ppm: 156.4, 147.7, 143.6, 142.7, 132.0, 130.1, 122.8, 122.3, 106.0, 19.2. LC-MS (m/z): calculated for [$C_{10}H_9NO$] = 159.07 m/z, found 160.2 m/z ($M+H^+$). Purity (GC): 96.6%.

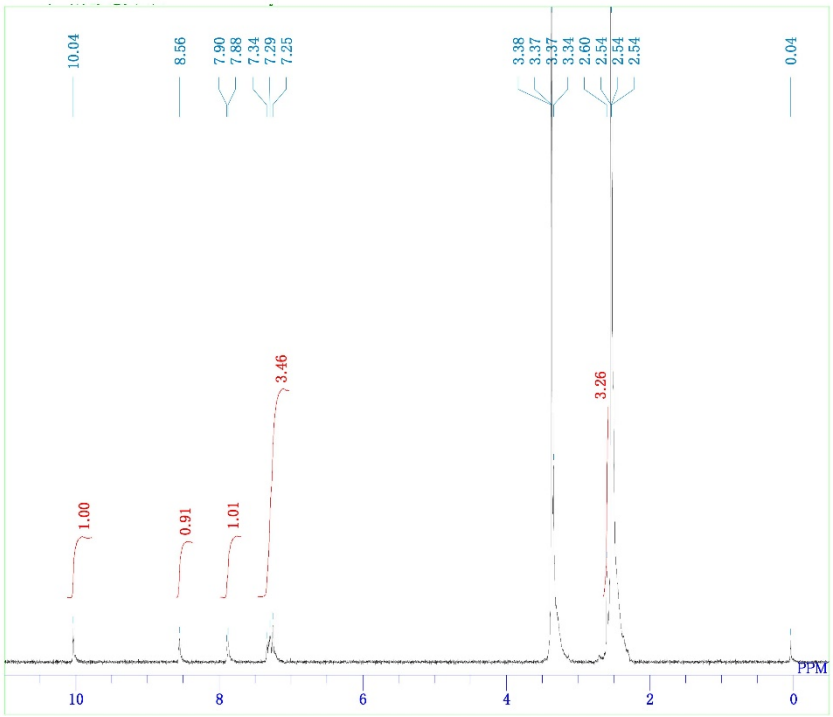


Figure S5. ^{1}H-NMR spectrum of compound **III**. This chart is measured by TCI that performed custom synthesis and shown wih permission from TCI.

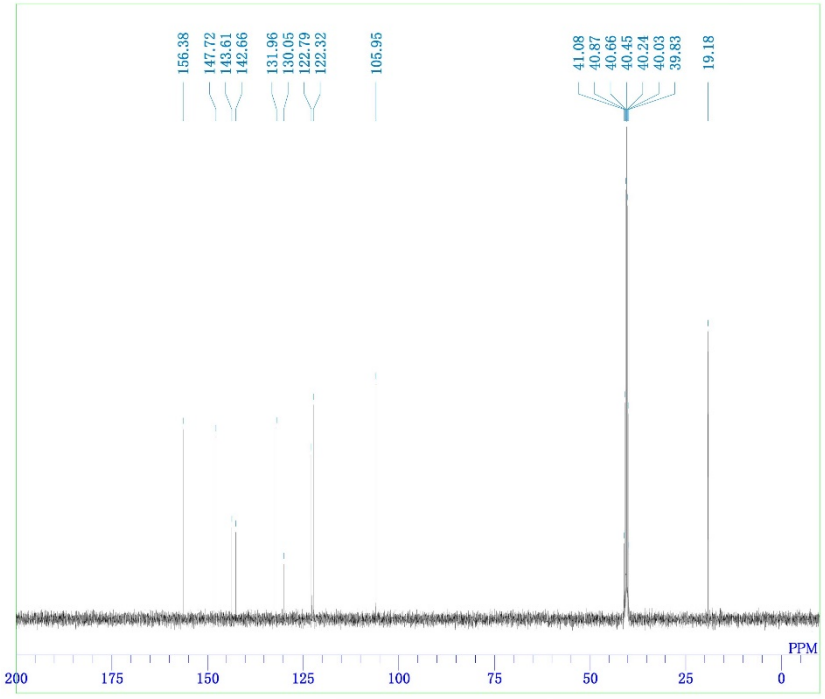


Figure S6. ^{13}C-NMR spectrum of compound **III**. This chart is measured by TCI that performed custom synthesis and shown wih permission from TCI.

Figure S7. Photograph of compound **III**.

Compound IV: ^{1}H-NMR (in $CDCl_3$) in ppm: 7.90 (d, 1H, ArH), 7.53 (d, 1H, ArH), 7.31 (t, 1H, ArH), 7.16-7.10 (m, 3H, ArH), 4.87 (br, 1H, OH), 2.64 (s, 3H, CH_3). ^{13}C-NMR (in $CDCl_3$) in ppm: 153.0, 134.8, 134.3, 128.1, 126.4, 126.2, 124.9, 124.5, 117.3, 110.2, 19.4. LC-MS (m/z): calculated for [$C_{11}H_{10}O$] = 158.07 m/z, found 159.0 m/z ($M+H^+$). Purity (LC): 99.5%.

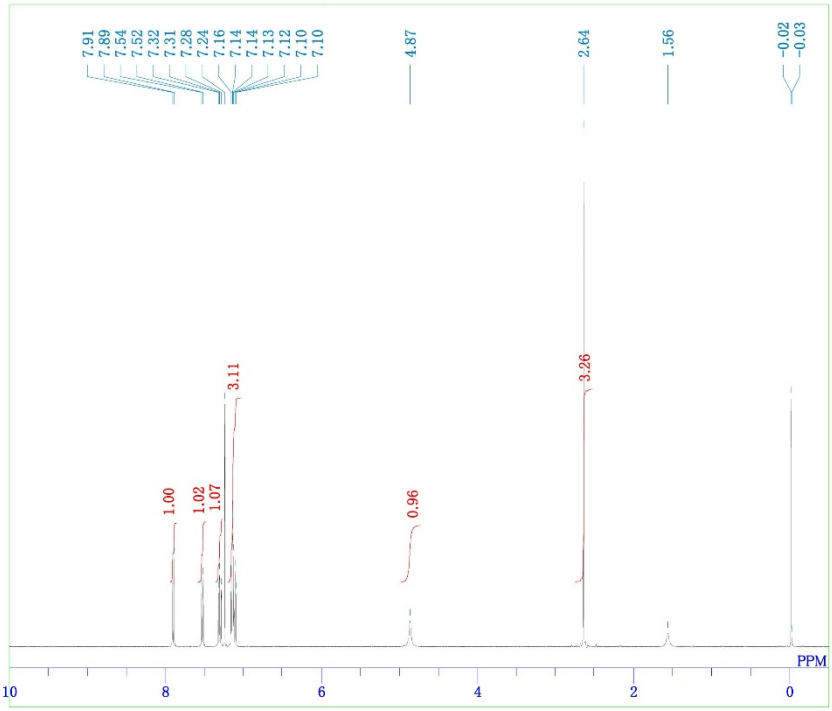


Figure S8. ^{1}H-NMR spectrum of compound **IV**. This chart is measured by TCI that performed custom synthesis and shown wih permission from TCI.

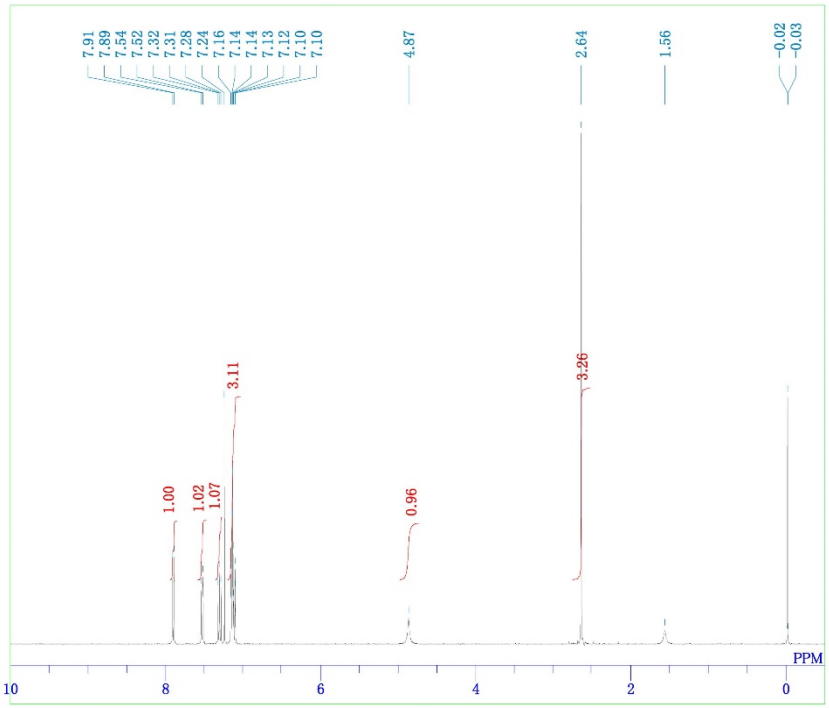


Figure S9. ^{13}C-NMR spectrum of compound **IV**. This chart is measured by TCI that performed custom synthesis and shown with permission from TCI.

Figure S10. Photograph of compound **IV**.

Compound V: ^{1}H-NMR (in DMSO-d_6) in ppm: 9.79 (s, 1H, OH), 7.17 (t, 1H, ArH), 7.09 (d, 1H, ArH), 6.87 (d, 1H, ArH), 6.80 (t, 1H, ArH), 5.26 (s, 2H, benzyl-CH_2), 2.89 (s, 3H, CH_3). ^{13}C-NMR (in DMSO-d_6) in ppm: 156.6, 129.9, 129.4, 121.1, 119.2, 115.4, 51.7, 31.1. LC-MS (m/z): calculated for [$C_8H_{10}N_2O_2$] = 166.07 m/z, found 167.0 m/z ($M+H^+$). Purity (LC): 99.2%.

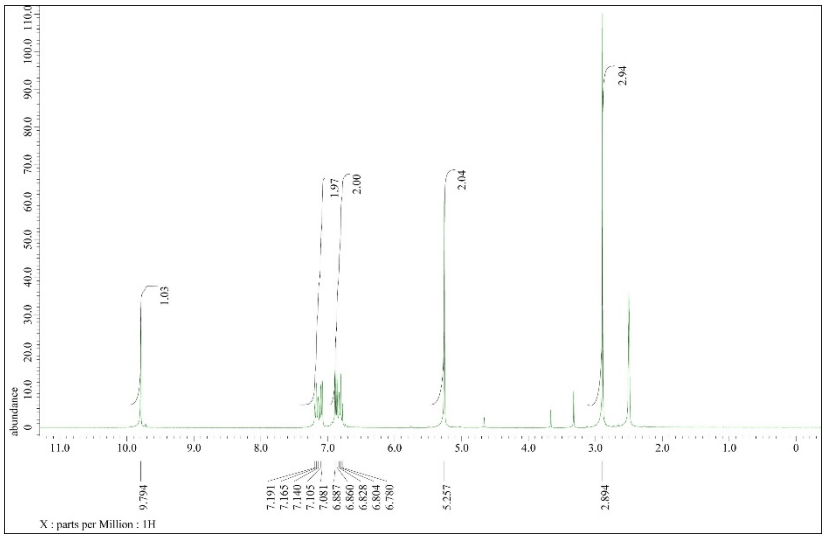


Figure S11. ^{1}H-NMR spectrum of compound **V**. This chart is measured by authors using AL300 BX NMR spectrometer (JEOL, Tokyo, Japan).

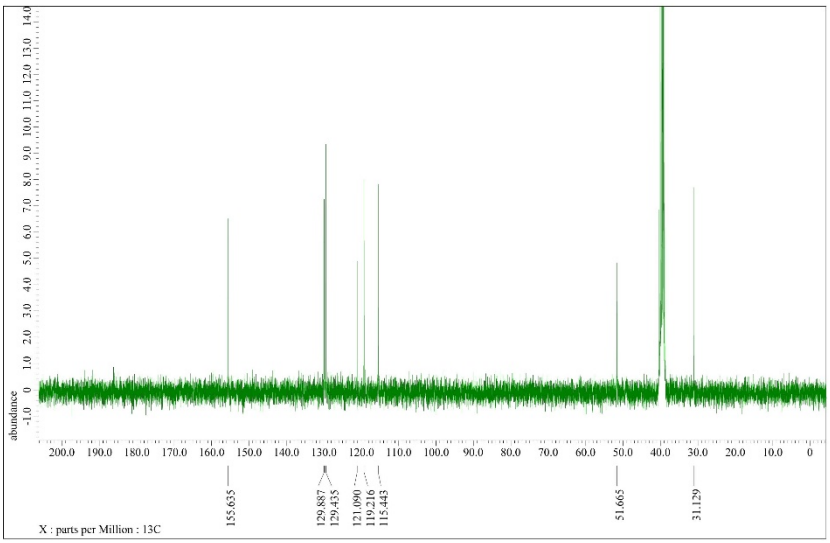
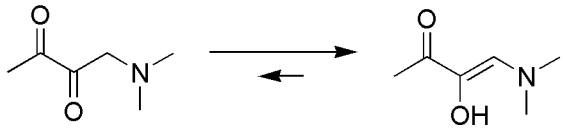


Figure S12. ^{13}C-NMR spectrum of compound **V**. This chart is measured by authors using AL300 BX NMR spectrometer (JEOL, Tokyo, Japan).

Figure S13. Photograph of compound **V**.

Compound VI: ^{1}H-NMR (in $CDCl_3$) in ppm: 6.31 (s, 1H, C=CH), 6.2-6.0 (br, 0.5H, OH), 3.10 (s, 6H, $N(CH_3)_2$), 2.14 (s, 3H, CH_3). ^{13}C-NMR (in $CDCl_3$) in ppm: 187.9, 132.6, 130.0, 42.3, 21.2. LC-MS (m/z): calculated for [$C_6H_{11}NO_2$] = 129.08 m/z, found 130.4 m/z ($M+H^+$). Purity (GC): 96.9%. Note that ^{1}H- and ^{13}C-NMR spectra indicate that compound **VI** mainly exist as enol-form in tautomerism.



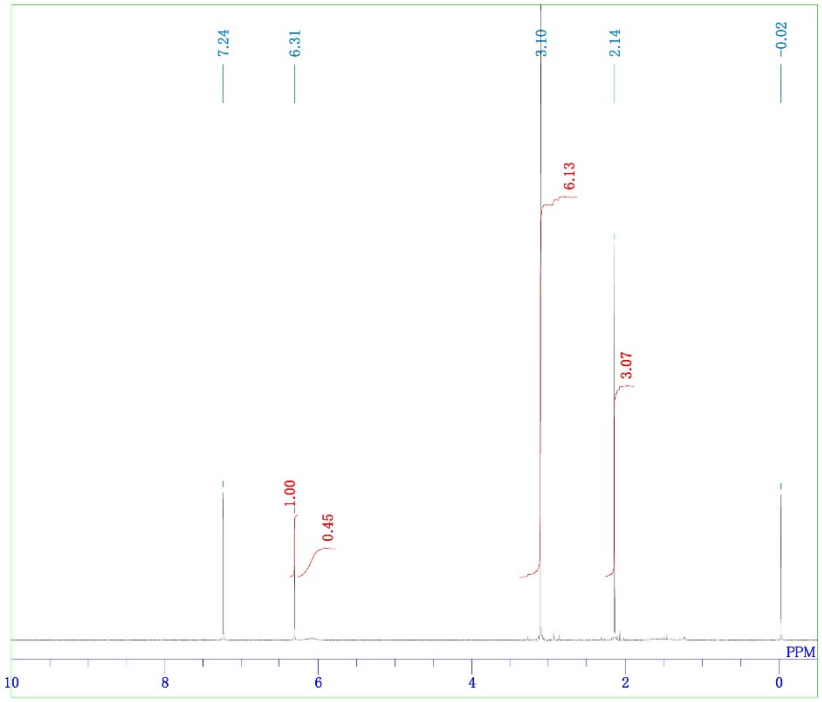


Figure S14. ^{1}H-NMR spectrum of compound **VI**. This chart is measured by TCI that performed custom synthesis and shown with permission from TCI.

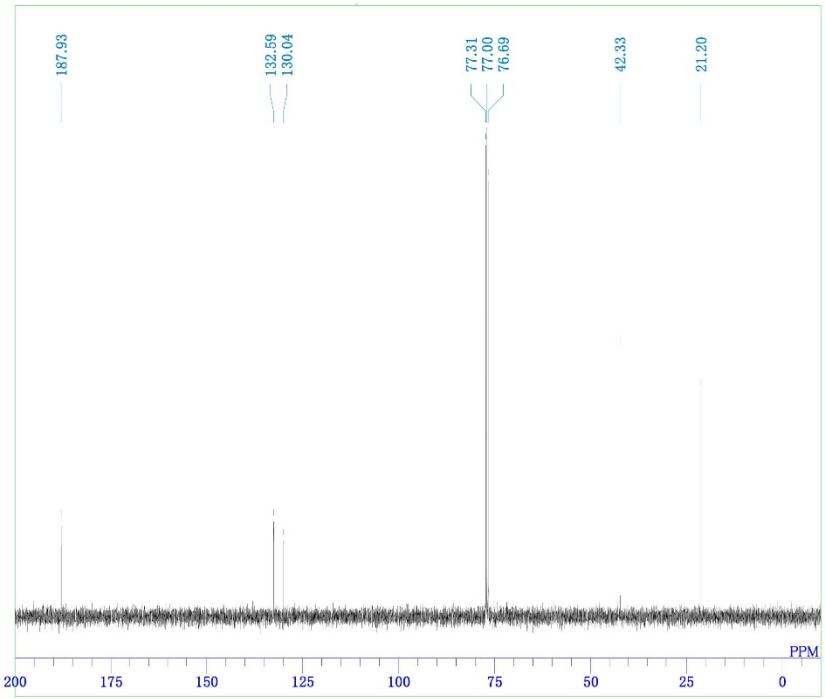


Figure S15. ^{13}C-NMR spectrum of compound **VI**. This chart is measured by TCI that performed custom synthesis and shown with permission from TCI.

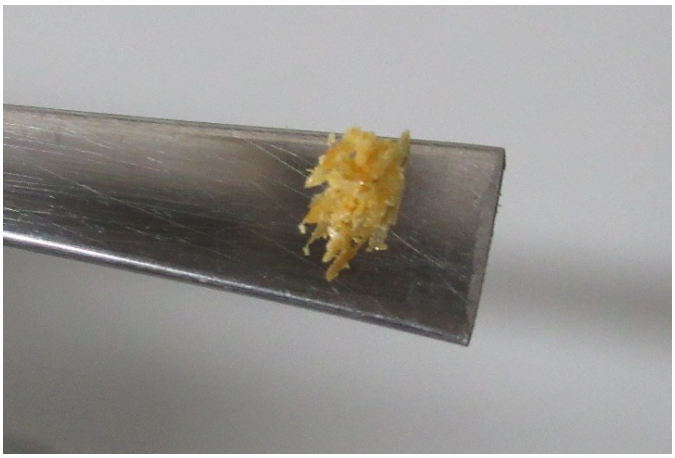


Figure S16. Photograph of compound **VI**.

Table S1. Energies of keto/enol-form of **II**

	Keto	Enol
Energy / E_h	-377.99028	-377.96296
Relative energy / kJ mol^{-1}	0.0	71.72

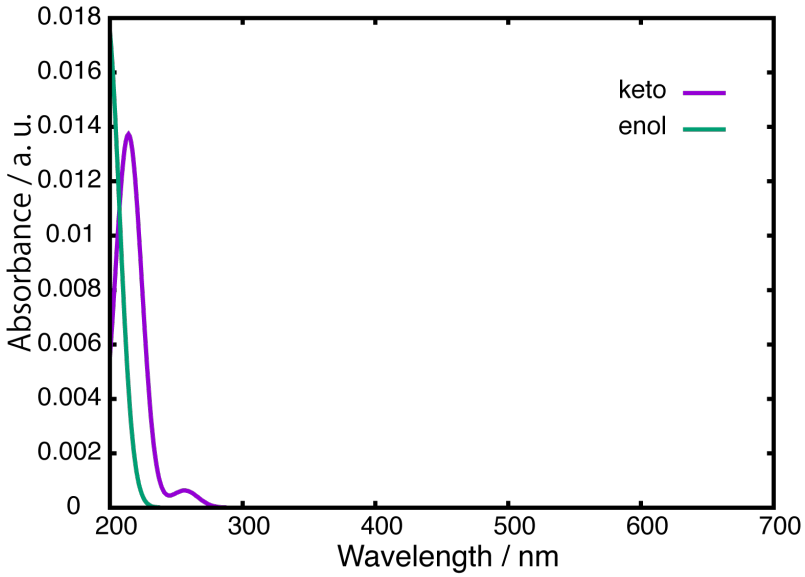


Figure S17. Computational UV-vis spectra for keto/enol-forms of **II**.

Table S2. Energies of syn/anti-conformers in keto/enol-forms of **VI**

	syn	anti
keto		
Energy / E_h	-437.99645	-438.01127
Relative energy / kJ mol^{-1}	62.62	23.71
enol		
Energy / E_h	-438.02030	-438.01697
Relative energy / kJ mol^{-1}	0.0	8.74

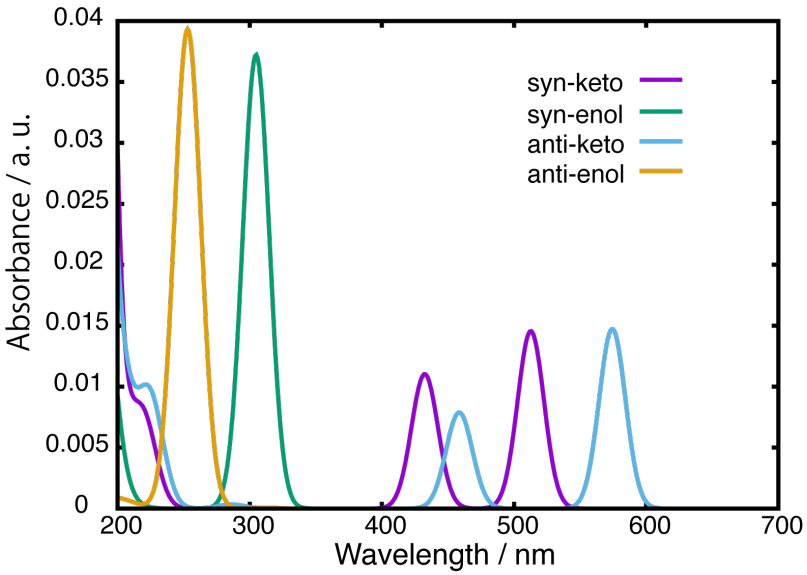


Figure S18. Computational UV-vis spectra keto/enol-forms of **VI**

3. Dependence of solvent and concentration

Compound I

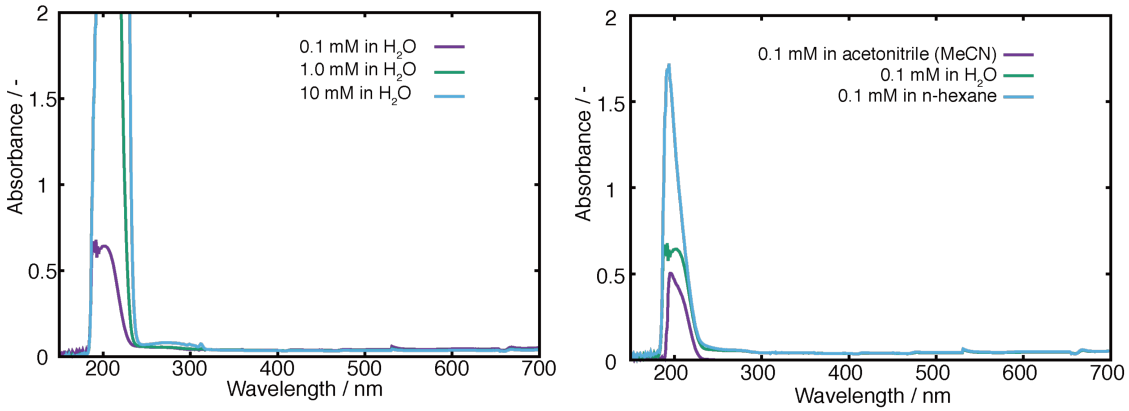


Figure S19. Experimental UV-vis absorption spectra of **I** with various concentration and solvent.

Compound II

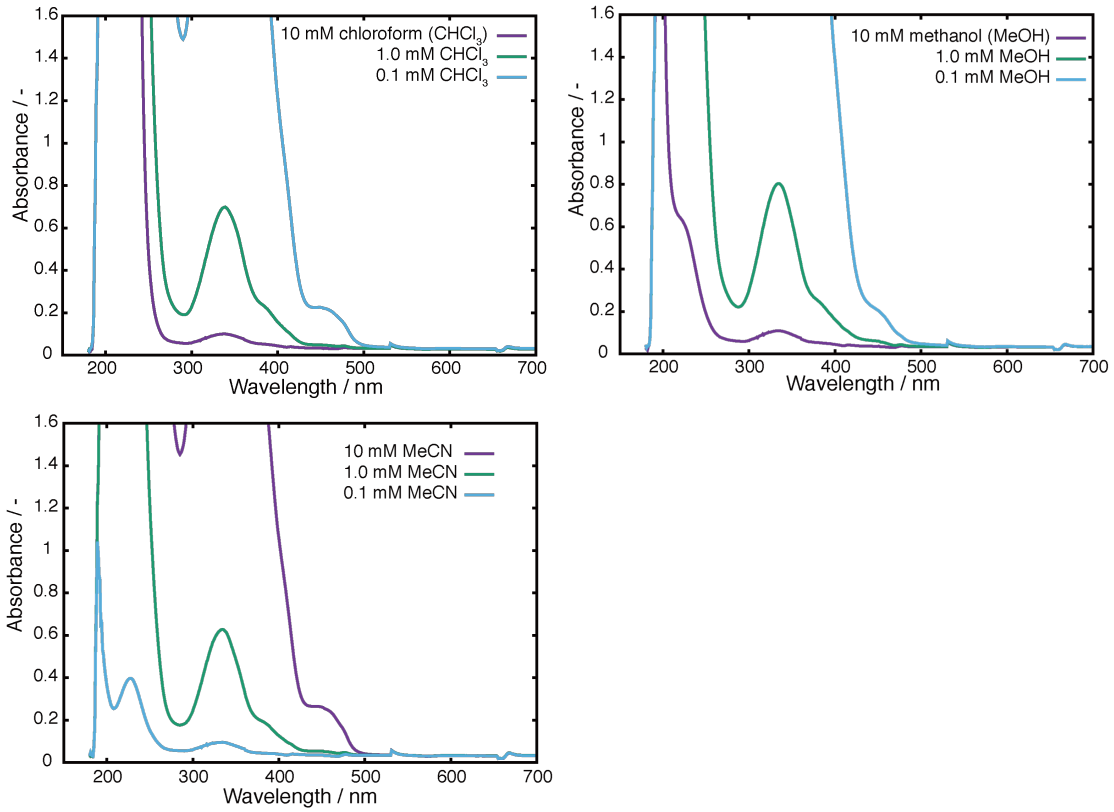


Figure S20. Experimental UV-vis absorption spectra of **II** with various concentration and solvent.

Compound III

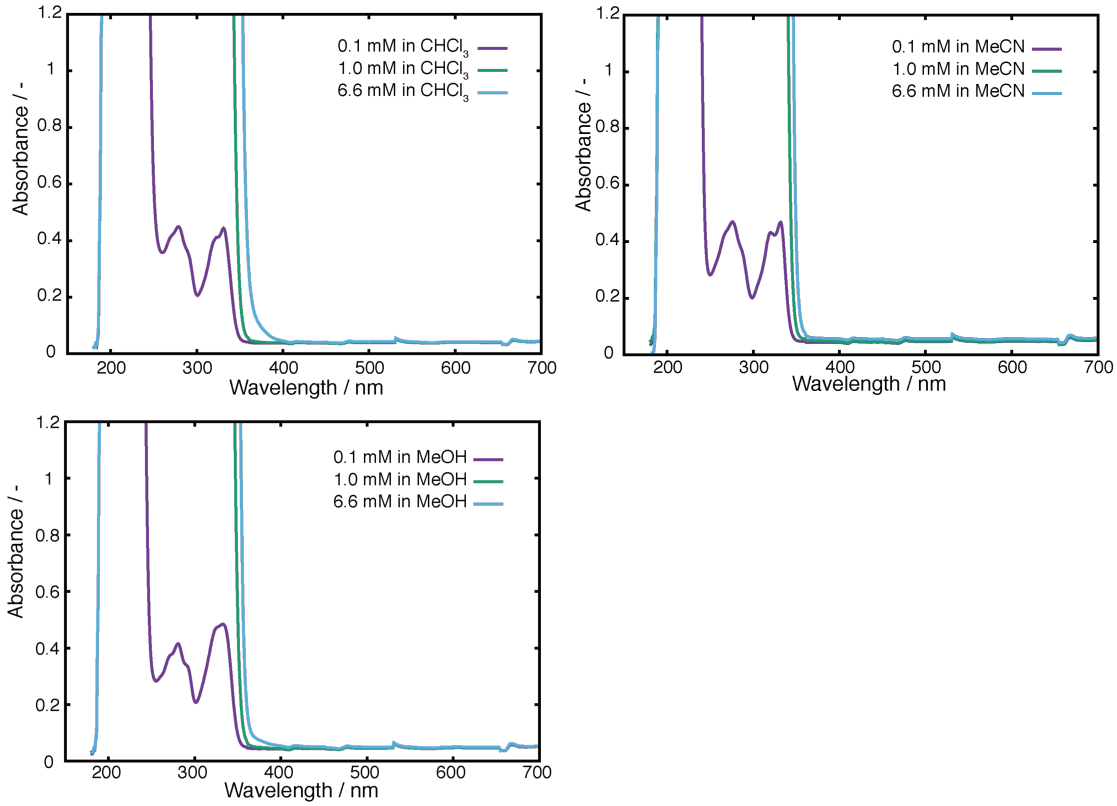


Figure S21. Experimental UV-vis absorption spectra of **III** with various concentration and solvent.

Compound IV

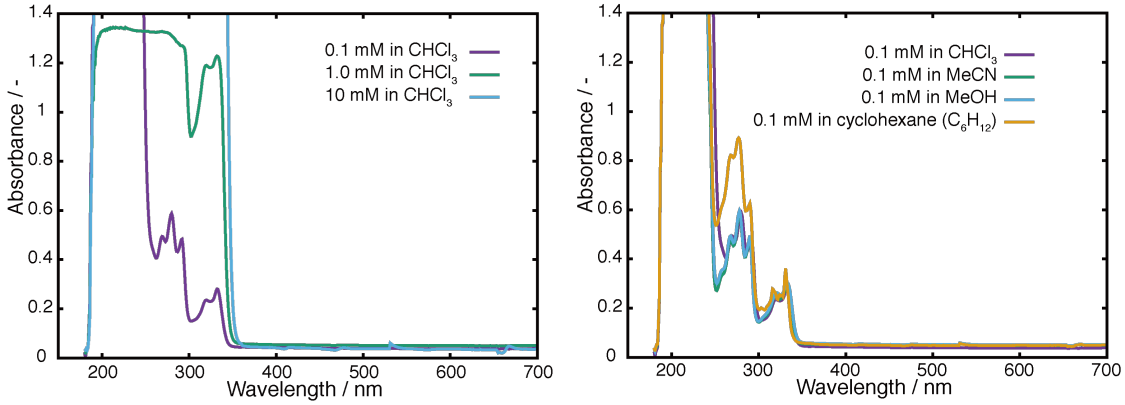


Figure S22. Experimental UV-vis absorption spectra of **IV** with various concentration and solvent.

Compound V

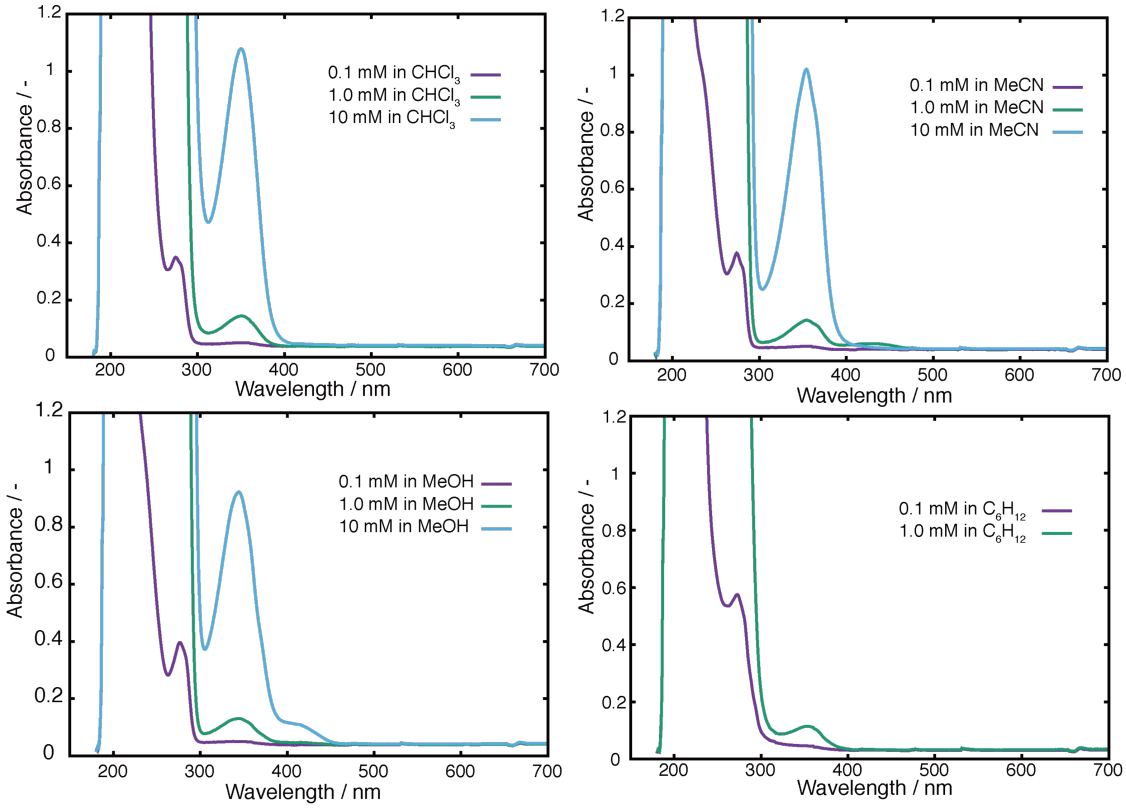


Figure S23. Experimental UV-vis absorption spectra of **V** with various concentration and solvent.

Compound VI

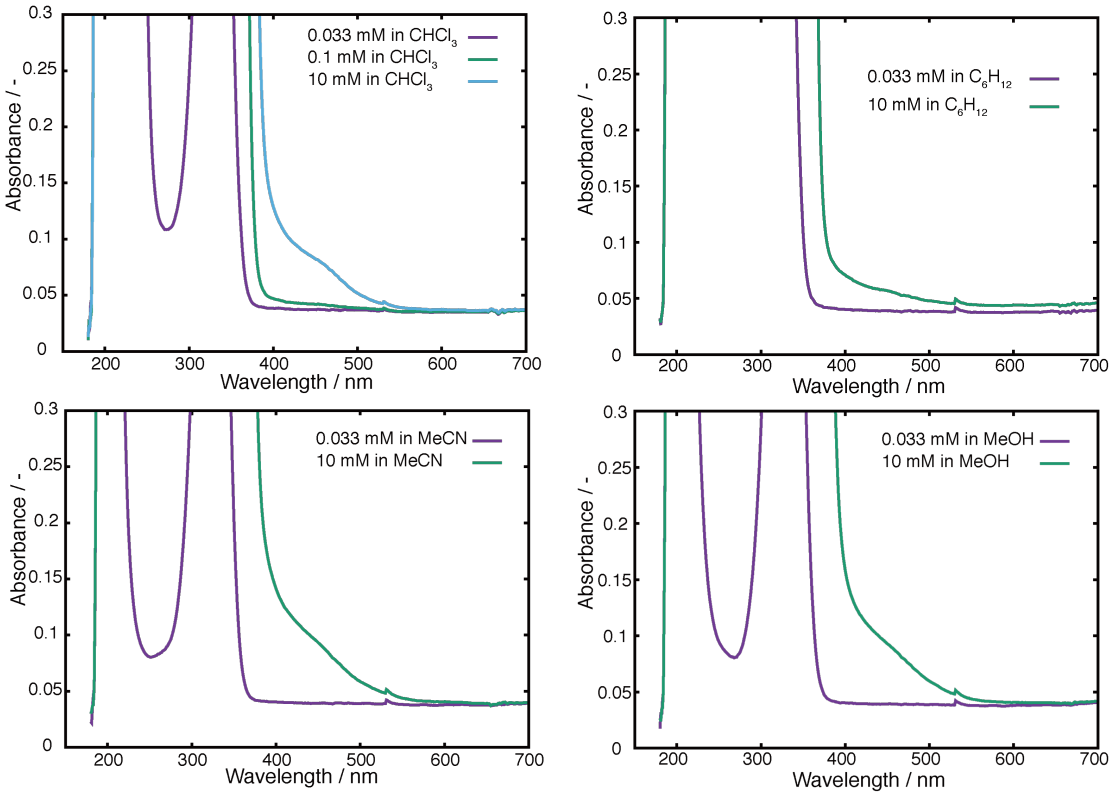


Figure S24. Experimental UV-vis absorption spectra of **VI** with various concentration and solvent.