Highly Efficient Synthesis of Azos Catalyzed by Common Metal of Copper (0) through Oxidative Coupling Reactions

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Experimental Section

The synthesis of symmetric AAzos from corresponding aromatic amines: As aniline for example, to a reaction tube equipped with a magnetic stirrer, appropriate amount of red copper (10 mg), NH_4Br (98 mg, 1 mmol), pyridine (48 μL ,0.6 mmol), aniline (91 μL, 1 mmol), toluene (2 mL) was charged. Followed by, air in the tube was exchanged with 1 atm of dioxygen for 3 times in an ice bath. Finally, the tube was immersed in an oil bath thermostated at 100^{o}C under a dioxygen atmosphere. The azobenzene was allowed to proceed under stirring for 24 h. The resultant product mixture was taken to determine the composition by GC(VARIAN CP-3800 GC, HP-5 capillary column, FID detector) and GC-MS(VARIAN 450-GC & VARIAN 240-GC) equipped with a CP8944 capillary column (30 m × 0.25 mm) and an FID detector. The aromatic azo compounds were characterized by ^{1}H NMR and ^{13}C NMR.

The synthesis of asymmetric AAzos from corresponding aromatic amines: As aniline and p-toluidine for example, to a reaction tube equipped with a magnetic stirrer, appropriate amount of red copper (10 mg), NH_4Br (98 mg, 1 mmol), pyridine (48 μL ,0.6 mmol), aniline (45.5 μL, 0.5 mmol), p-toluidine (53.3 mg, 0.5 mmol), toluene (2 mL) was charged. Followed by, air in the tube was exchanged with 1 atm of dioxygen for 3 times in an ice bath. Finally, the tube was immersed in an oil bath thermostated at 100^{o}C under a dioxygen atmosphere. The product was allowed to proceed under stirring for 24 h. The resultant product mixture was taken to determine the composition by GC(VARIAN CP-3800 GC, HP-5 capillary column, FID detector) and GC-MS(VARIAN 450-GC & VARIAN 240-GC) equipped with a CP8944 capillary column (30 m × 0.25 mm) and an FID detector. The aromatic azo compounds were characterized by ^{1}H NMR and ^{13}C NMR.

The synthesis of Cu nanoparticles: $Cu(acac)_2$ (100 mg), 1,2-hexadecanediol (200mg), dioctyl ether (20 mL), oleic acid (1 mL), and oleylamine (1 mL) were mixed under stirring and bubbled by N_2 for 5 minutes. The mixture was then heated to 90^{o}C to dissolve $Cu(acac)_2$. The temperature was raised to about 265^{o}C in 20 minutes. The solution was kept at this temperature for 3 hour before it was cooled down to room temperature. Anhydrous ethanol was added and then the suspension was centrifuged to separate the nanoparticles. The nanoparticles were dispersed in 10 mL hexane and precipitated out by adding ethanol. The process was repeated one more time to purify the nanoparticles. The final product was dispersed in 10 mL of ethanol for further use.

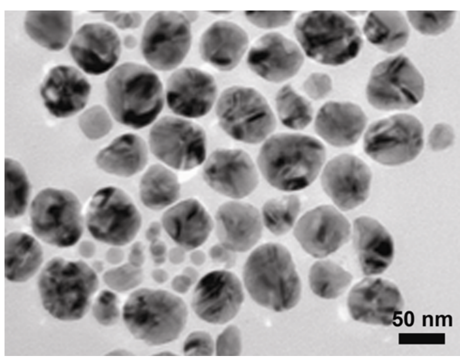


Fig. S1 TEM images of Cu NPs (A).

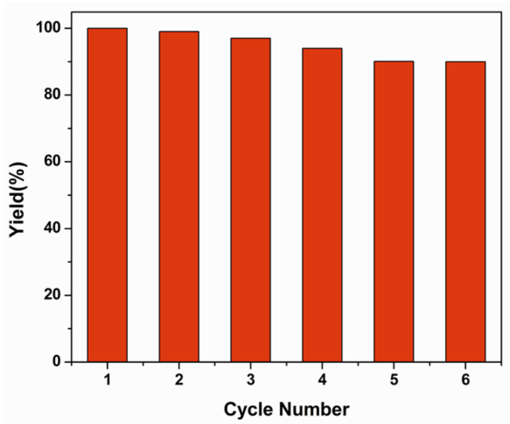
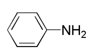
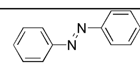
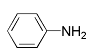
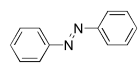
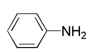
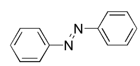


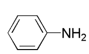
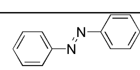
Fig. S2 The catalytic stability of red copper in the synthesis of azobenzene from the oxidation of aniline in different cycles.

Table S1. Azobenzene formation from aniline using red copper, Cu NPs and CuBr as catalysts. [a]

entry	catalyst	substrate	product	yield(%)[c]
1	red copper			100
2	Cu NPs			61
3[b]	CuBr			96

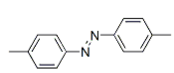
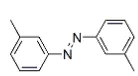
[a] The reaction was carried out with appropriate amount of catalysts, 1 mmol aniline compounds, 1 mmol NH_4Br, 0.6 mmol pyridine, and 2 mL toluene at 100°C for 24 h under 1 atm of oxygen. [b] The reaction was carried out with the presence of 3 mmol% catalyst, 1 mmol aniline compounds and 0.18 mmol pyridine in 4 mL toluene at 60°C for 20 h under 1 atm of oxygen. [c] GC yield.

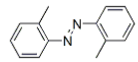
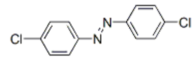
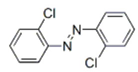
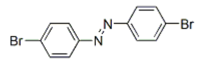
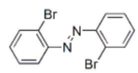
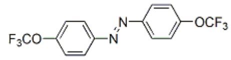
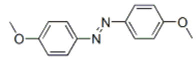
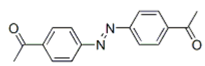
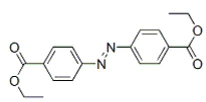
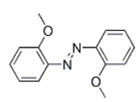
Table S2. Azobenzene formation from aniline without red copper catalysts. [a]

entry	substrate	product	yield(%)[b]
1			2

[a] The reaction was carried out with 1 mmol aniline compounds, 1 mmol NH_4Br, 0.6 mmol pyridine, and 2 mL toluene at 100°C for 24 h under 1 atm of oxygen. [b] GC yield.

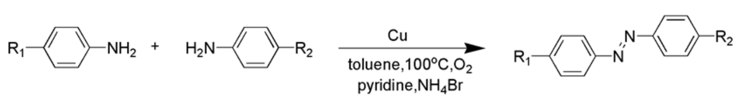
Table S3. Symmetric aromatic azos formation from different corresponding aniline compounds.[a]

entry	product	yield(%)[b]
1		>99 (99)
2		>99 (99)

3		99 (90)
4		>99 (91)
5		22 (21)
6		>99 (95)
7		36 (28)
8		>99 (99)
9		28 (20)
10		96 (94)
11		>99 (98)
12		69 (57)

[a] All reactions were carried out with 10mg of Cu catalyst, 1 mmol aniline compounds, 1 mmol NH_4Br, 0.6 mmol pyridine, and 2 mL toluene at 100°C for 24 h under 1 atm of oxygen. [b] GC yield. The values in parentheses are the yields of the isolated products.

Table S4. Asymmetric aromatic azos formation from different corresponding aniline compounds.[a]



entry	R_1	R_2	yield(%)[b]
1	H	CH_3	49 (30)
2	H	Cl	70 (59)
3	H	CF_3O	64 (60)

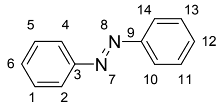
4	H	$CH_3C{=}O$	29 (29)
5	H	CH_3CH_2OOC	42 (40)
6	CH_3	Cl	53 (40)
7	CH_3	CF_3O	38 (31)
8	CH_3	$CH_3C{=}O$	11 (10)
9	CH_3	CH_3CH_2OOC	19 (16)
10	CF_3O	Cl	66 (63)
11	CF_3O	$CH_3C{=}O$	63 (62)
12	CF_3O	CH_3CH_2OOC	66 (60)
13	$CH_3C{=}O$	CH_3CH_2OOC	57 (50)
14	$CH_3C{=}O$	Cl	37 (33)
15	Cl	CH_3CH_2OOC	46 (44)
16	Br	H	65 (57)
17	Br	CH_3	40 (29)
18	Br	Cl	57 (30)
19	Br	CF_3O	72 (65)
20	Br	$CH_3C{=}O$	37 (37)
21	Br	CH_3CH_2OOC	58 (56)

[a] All reactions were carried out with 10mg of Cu catalyst, 0.5 mmol amino aromatic compounds containing R_1 group, 0.5 mmol amino aromatic compounds containing R_2 group, 1 mmol NH_4Br, 0.6 mmol pyridine, and 2 mL toluene at 100^{o}C for 24 h under 1 atm of oxygen. [b] GC yield. The values in parentheses are the yields of the isolated products.

NMR data of the AAzos:

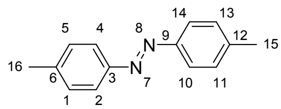
Full spectroscopic data for Aromatic Azo products are provided as follows.[1-5]

Symmetric AAzos:

0. 

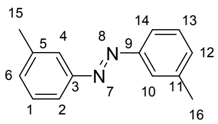
^{1}H NMR(400 MHz, $CDCl_3$):δ=7.95-7.93(d, 4H, H_2, H_4, H_{10}, H_{14}), 7.54-7.47(m, 6H, H_1, H_5, H_6, H_{11}, H_{12}, H_{13}).

^{13}C NMR(100 MHz, $CDCl_3$): δ=123.07, 129.31, 131.22, 152.85.

1. 

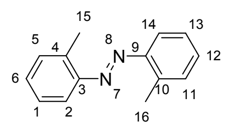
^{1}H NMR(400 MHz, $CDCl_3$):δ=7.83-7.81(d,4H, H_2, H_4, H_{10}, H_{14}), 7.32-7.30(d,4H, H_1, H_5, H_{11}, H_{13}),2.44(s,6H, H_{15}, H_{16}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 21.71, 122.92, 129.91, 141.43, 150.99.

2. 

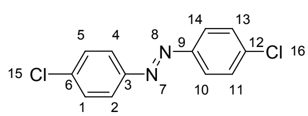
^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.73 (s, 4H, H_2, H_4, H_{10}, H_{14}), 7.43-7.39 (m, 2H, H_1, H_{13}), 7.31-7.26 (d, 2H, H_6, H_{12}), 2.47 (s, 6H, H_{15}, H_{16}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 21.61, 120.70, 123.04, 129.11, 131.91, 139.19, 152.97.

3. 

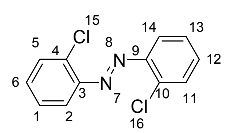
^{1}H NMR (400 MHz,$CDCl_3$): δ = 7.62-7.60(d, 2H, H_2, H_{14}),7.33-7.30(m, 4H, H_1, H_6, H_{12}, H_{13}),7.24-7.22(m, 2H, H_5, H_{11}),2.72(s, 6H, H_{15}, H_{16}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 17.88, 116.07, 126.60, 130.92, 138.26, 151.31.

4. 

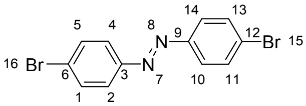
^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.87-7.85 (d, 4H, H_2, H_4, H_{10}, H_{14}), 7.49-7.47 (d, 4H, H_1, H_5, H_{11}, H_{13}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 124.41, 129.63, 137.45, 150.92.

5. 

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.78-7.76 (d, 2H, H_2, H_{14}), 7.58-7.56 (d, 2H, H_5, H_{11}), 7.44-7.40 (m, 2H, H_6, H_{12}), 7.38-7.34 (m, 2H, H_1, H_{13}).

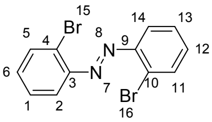
^{13}C NMR (100 MHz, $CDCl_3$): δ = 118.29, 127.61, 130.94, 132.47.



6.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.81-7.78 (d, 4H, H_2, H_4, H_{10}, H_{14}), 7.66-7.64 (d, 4H, H_1, H_5, H_{11}, H_{13}).

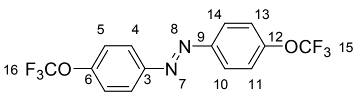
^{13}C NMR (100 MHz, $CDCl_3$): δ = 124.62, 132.60.



7.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.77-7.76 (d, 4H, H_2, H_5, H_{11}, H_{14}), 7.43-7.39 (m, 2H, H_1, H_{13}), 7.36-7.33 (m, 2H, H_6, H_{12}).

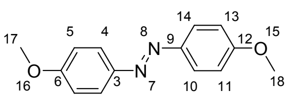
^{13}C NMR (100 MHz, $CDCl_3$): δ = 118.63, 128.33, 132.71, 134.02.



8.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.97-7.95 (d, 4H, H_2, H_4, H_{10}, H_{14}), 7.37-7.35 (d, 4H, H_1, H_5, H_{10}, H_{14}).

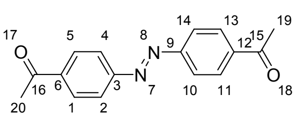
^{13}C NMR (100 MHz, $CDCl_3$): δ = 113.93, 121.29, 121.73, 124.55, 124.84.



9.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.89-7.87 (d, 4H, H_2, H_4, H_{10}, H_{14}), 7.01-6.99 (d, 4H, H_1, H_5, H_{11}, H_{13}), 3.89 (s, 6H, H_{17}, H_{18}).

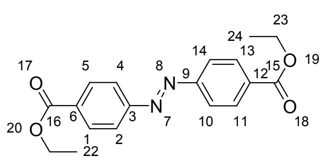
^{13}C NMR (100 MHz, $CDCl_3$): δ = 55.78, 114.37, 124.55, 147.26, 161.81.



10.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.13-8.11 (d, 4H, H_1, H_5, H_{11}, H_{13}), 8.02-7.99 (d, 4H, H_2, H_4, H_{10}, H_{14}), 2.67(s, 6H, H_{19}, H_{20}).

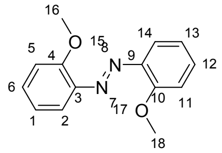
^{13}C NMR (100 MHz, $CDCl_3$): δ = 27.13, 123.42, 129.63, 140.12, 154.50, 197.69.



11.

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.22-8.20 (d, 4H, H_1, H_5, H_{11}, H_{13}), 7.99-7.97 (d, 4H, H_2, H_4, H_{10}, H_{14}), 4.45-4.40 (m,4H, H_{21}, H_{23}), 1.45-1.41 (p, 6H, H_{22}, H_{24}).

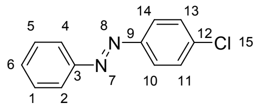
^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.53, 61.56, 123.07, 130.81, 132.92, 155.01, 166.11.

12. 

^{1}H NMR (400 MHz, $CDCl_3$): δ=7.65-7.61 (m, 2H, H_2, H_{14}), 7.42-7.39 (m, 2H, H_6, H_{12}), 7.09-7.06 (m, 2H, H_1, H_{13}), 7.03-6.98 (m, 2H, H_5, H_{11}), 4.02 (s, 6H, H_{16}, H_{18}).

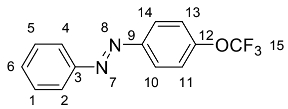
^{13}C NMR (100 MHz, $CDCl_3$): δ =56.01, 112.22, 117.21, 120.49, 131.88, 142.63, 156.51.

Asymmetric AAzos:

2. 

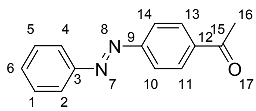
^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.92-7.86 (m, 4H, H_2, H_4, H_{10}, H_{14}), 7.52-7.48 (m, 5H, H_1, H_5, H_6, H_{11}, H_{13}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 123.12, 124.33, 129.34, 129.54, 131.49.

3. 

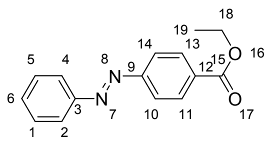
^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.97-7.95 (d, 2H, H_2, H_4), 7.92-7.90 (d, 2H, H_{10}, H_{14}), 7.54-7.48 (m, 3H, H_1, H_5, H_6), 7.36-7.34 (d, 2H, H_{11}, H_{13}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 31.12, 121.48, 123.14, 124.54, 129.34, 131.57.

4. 

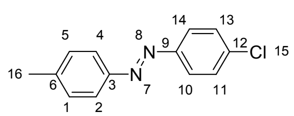
^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.11-8.09 (d, 2H, H_{11}, H_{13}), 7.98-7.94 (m, 4H, H_2, H_4, H_{10}, H_{14}), 7.54-7.52 (m, 3H, H_1, H_5, H_6), 2.66 (s, 3H, H_{16}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 31.14, 123.08, 123.36, 129.40, 129.58, 131.99.

5. 

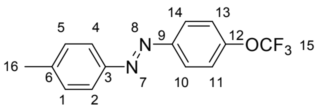
^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.20-8.18 (d, 2H, H_{11}, H_{13}), 7.96-7.94 (d, 4H, H_{10}, H_{14}, H_2, H_4), 7.55-7.51 (m, 3H, H_1, H_5, H_6), 4.44-4.39 (m, 2H, H_{18}), 1.44-1.41 (p, 3H, H_{19}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.53, 31.12, 61.46, 122.79, 123.33, 129.37, 130.76, 131.87.

6. 

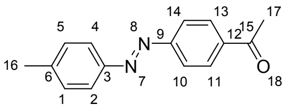
^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.86-7.84 (d, 2H, H_{10}, H_{14}), 7.83-7.81 (d, 2H, H_2, H_4), 7.49-7.46 (d, 2H, H_{11}, H_{13}), 7.32-7.30 (d, 2H, H_1, H_5), 2.44 (s, 3H, H_{16}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 123.14, 124.21, 129.50, 135.53.

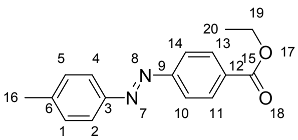
7. 

^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.94-7.92 (d, 2H, H_{10}, H_{14}), 7.83-7.81 (d, 2H, H_2, H_4), 7.35-7.30 (p, 4H, H_{11}, H_{13}, H_1, H_5), 2.43 (s, 3H, H_{16}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 21.72, 29.90, 31.12, 121.47, 123.16, 124.37, 130.00.

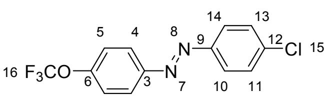
8. 

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.11-8.09 (d, 2H, H_{11}, H_{13}), 7.96-7.94 (d, 2H, H_{10}, H_{14}), 7.87-7.85 (d, 2H, H_2, H_4), 7.34-7.32 (d, 2H, H_1, H_5), 2.66 (s, 3H, H_{16}), 2.45 (s, 3H, H_{17}).

9. 

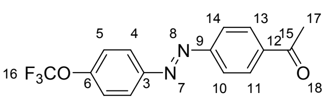
^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.20-8.18 (d, 2H, H_{11}, H_{13}), 7.94-7.92 (d, 2H, H_{10}, H_{14}), 7.87-7.85 (d, 2H, H_2, H_4), 7.34-7.32 (d, 2H, H_1, H_5), 4.44-4.39 (m, 2H, H_{19}), 2.45 (s, 3H, H_{16}), 1.44-1.41 (m, 3H, H_{20}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 14.56, 61.46, 122.69, 123.37, 130.06, 130.75.

10. 

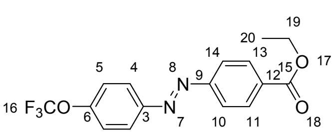
^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.97-7.95 (d, 2H, H_{10}, H_{14}), 7.88-7.86 (d, 2H, H_2, H_4), 7.50-7.48 (d, 2H, H_{11}, H_{13}), 7.37-7.35 (d, 2H, H_1, H_5).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 121.51, 124.39, 124.43, 124.65, 124.70, 129.61.

11. 

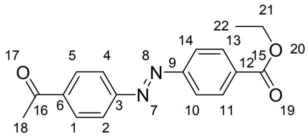
^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.12-8.10 (d, 2H, H_{11}, H_{13}), 8.01-7.96 (m, 4H, H_{10}, H_{14}, H_2, H_4), 7.38-7.36 (d, 2H, H_1, H_5), 2.66 (s, 3H, H_{17}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 31.11, 121.48, 123.19, 124.91, 129.58.

12. 

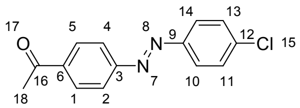
^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.21-8.18 (d, 2H, H_{11}, H_{13}), 8.00-7.98 (d, 2H, H_{10}, H_{14}), 7.95-7.93 (d, 2H, H_2, H_4), 7.38-7.36 (d, 2H, H_1, H_5), 4.44-4.39 (m, 2H, H_{19}), 1.44-1.40 (p, 3H, H_{20}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 31.12, 121.48, 122.89, 124.87, 130.79.

13. 

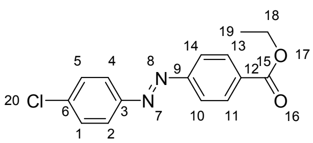
^{1}H NMR (400 MHz, CDCl$_3$): δ = 8.21-8.19 (d, 2H, H$_{11}$, H$_{13}$), 8.13-8.10 (d, 2H, H$_1$, H$_5$), 8.01-7.96 (p, 4H, H$_{10}$, H$_{14}$, H$_2$, H$_4$), 4.44-4.39 (m, 2H, H$_{21}$), 2.66 (s, 3H, H$_{18}$), 1.44-1.40 (p, 3H, H$_{22}$).

^{13}C NMR (100 MHz, CDCl$_3$): δ = 31.11, 61.56, 109.99, 123.08, 129.59, 130.81.

14. 

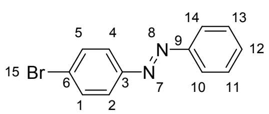
^{1}H NMR (400 MHz, CDCl$_3$): δ = 8.11-8.09 (d, 2H, H$_1$, H$_5$), 7.97-7.95 (d, 2H, H$_2$, H$_4$), 7.91-7.89 (d, 2H, H$_{10}$, H$_{14}$),7.51-7.49 (d, 2H, H$_{11}$, H$_{13}$), 2.66 (s, 3H, H$_{18}$).

^{13}C NMR (100 MHz, CDCl$_3$): δ = 31.14, 109.99, 123.16, 124.62, 129.59, 129.68.

15. 

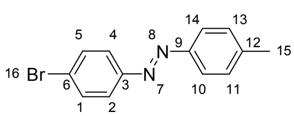
^{1}H NMR (400 MHz, CDCl$_3$): δ = 8.20-8.18 (d, 2H, H$_{11}$, H$_{13}$), 7.94-7.89 (m, 4H, H$_2$, H$_4$, H$_{10}$, H$_{14}$), 7.51-7.49 (d, 2H, H$_1$, H$_5$), 4.44-4.38 (m, 2H, H$_{18}$), 1.44-1.40 (p, 3H, H$_{19}$).

^{13}C NMR (100 MHz, CDCl$_3$): δ = 31.13, 61.51, 109.98, 122.87, 124.59, 129.65, 130.78.

16. 

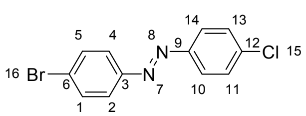
^{1}H NMR (400 MHz, CDCl$_3$): δ = 7.92-7.89 (m, 2H, H$_{10}$, H$_{14}$), 7.81-7.79 (d, 2H, H$_2$, H$_4$), 7.65-7.63 (d, 2H, H$_1$, H$_5$), 7.53-7.48 (m, 3H, H$_{11}$, H$_{12}$, H$_{13}$).

^{13}C NMR (100 MHz, CDCl$_3$): δ = 31.15, 109.99, 123.14, 124.55, 129.35, 132.52.

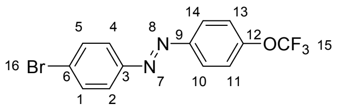
17. 

^{1}H NMR (400 MHz, CDCl$_3$): δ = 7.82-7.76 (m, 4H, H$_2$, H$_4$, H$_{10}$, H$_{14}$), 7.64-7.61 (d, 2H, H$_{11}$, H$_{13}$), 7.31-7.29 (d, 2H, H$_1$, H$_5$), 2.43 (s, 3H, H$_{15}$).

^{13}C NMR (100 MHz, CDCl$_3$): δ = 31.15, 123.15, 124.43, 130.01, 132.46.

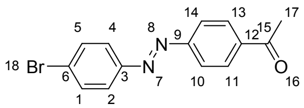
18. 

^{1}H NMR (400 MHz, CDCl$_3$): δ = 7.87-7.85 (d, 2H, H$_{10}$, H$_{14}$), 7.80-7.77 (d, 2H, H$_2$, H$_4$), 7.65-7.63 (d, 2H, H$_1$, H$_5$), 7.49-7.47 (d, 2H, H$_{11}$, H$_{13}$).

19. 

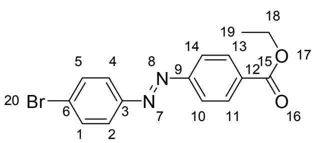
^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.96-7.94 (d, 2H, H_2, H_4), 7.80-7.78 (d, 2H, H_{10}, H_{14}), 7.66-7.64 (d, 2H, H_1, H_5), 7.36-7.34 (d, 2H, H_{11}, H_{13}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 121.51, 124.67, 132.61.

20. 

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.11-8.09 (d, 2H, H_{11}, H_{13}), 7.97-7.95 (d, 2H, H_{10}, H_{14}), 7.84-7.82 (d, 2H, H_2, H_4), 7.68-7.66 (d, 2H, H_1, H_5), 2.66 (s, 3H, H_{17}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 31.12, 109.98, 123.17, 124.80, 129.58, 132.66, 158.51, 200.07.

21. 

^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.19-8.16 (d, 2H, H_{11}, H_{13}), 7.94-7.91 (d, 2H, H_{10}, H_{14}), 7.82-7.80 (d, 2H, H_2, H_4), 7.66-7.64 (d, 2H, H_1, H_5), 4.43-4.37 (m, 2H, H_{18}), 1.43-1.39 (p, 3H, H_{19}).

^{13}C NMR (100 MHz, $CDCl_3$): δ = 31.10, 110.08, 122.87, 124.77, 130.78, 132.63.

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