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Synthesis of Binaphthyl Derivatives through Radical Cation Formation

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Introduction

Aromatic radical cations are well-known species in electrochemical and gas phase reactions that produce biaryls through a coupling reaction with aromatic compounds, and the existence of aromatic radical cations in the liquid phase has recently been proposed on the basis of spectroscopic studies.[1] Metal salts such as $CuCl_2$,[2] $Tl(OCOCF_3)_3$,[3] and $Mn[CH(COCH_3)_2]_3$ [4] and Lewis acids such as $AlCl_3$,[5] $FeCl_3$,[6] and $SbCl_3$ [7] are reported to be effective oxidants for aromatic compounds that produce biaryls through the formation of aromatic radical cations. The latter reaction is known as the Scholl reaction.[8] While the formation of biaryls is regarded as conclusive evidence for the formation of aromatic radical cations as reaction intermediates in reaction mechanism studies, there have been few reports of a general synthetic application using the coupling reaction of aromatic radical cations to obtain biaryls, probably because of their high reactivity.

On the other hand, NO^+ is a remarkable, diverse reagent not only for nitrosation[9] and nitration[10] but also for oxidations that produce aromatic radical cations.[11] It is reported that $NOBF_4$ catalytically oxidizes naphthalene derivatives to give binaphthyl derivatives in the presence of O_2 under acidic conditions[12] where the formation of radical cations tends to be favorable.[13] These results prompted us to study the coupling reaction of aromatic radical cations to obtain biaryls under acidic conditions.

In this paper, we report the catalytic coupling reaction of naphthalene derivatives using $NaNO_2$ with CF_3SO_3H, the role of acids in this reaction, and the Scholl reaction using SbF_5.

Results and Discussion

Coupling Reaction of Naphthalene Derivatives. It is well-known that NO^+ is easily produced by treatment of $NaNO_2$ with Brønsted acids. In this study, we chose CF_3SO_3H as the Brønsted acid because a mixture of $NaNO_2$ and CF_3SO_3H in CH_3CN gave a homogeneous solution.[14] When 1-methylnaphthalene (5 mmol) was added to the solution of CH_3CN (50 mL) with $NaNO_2$ (0.5 mmol) and CF_3SO_3H (10 mmol) at 0 °C in air, the coupling reaction of 1-methylnaphthalene took place to produce 4,4′-dimethyl-1,1′-binaphthyl in 97% yield.[15] Similarly, other naphthalene derivatives gave the corresponding binaphthyl derivatives as shown in Table 1. Most substrates were converted to give binaphthyl derivatives in good yields with high regioselectivity; however, some substrates showed different tendencies. The low yield of naphthalene was clearly caused by its low reactivity because most of the naphthalene was recovered after the reaction. On the contrary, 1-naphthol was too reactive even at −78 °C for the formation of biaryls and resulted only in unidentifiable oily products. On the other hand, the deterioration of the yield and regioselectivity in the cases of 2-methyl-, 2-ethyl-, 2,3-dimethyl-, and 2,6-dimethylnaphthalene seems to be caused by steric hindrance because 2-methoxynaphthalene, which is more reactive but has a smaller steric hindrance at the 1-position than 2-methylnaphthalene, gave the coupling product in excellent regioselectivity although 2-methylnaphthalene showed low regioselectivity. Furthermore, 1,4- and 1,5-dimethylnaphthalene, which are more reactive than 1-methylnaphthalene, gave only trace amounts of coupling products resulting in the recovery of unreacted substrates. Therefore, steric hin-

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(15) In the absence of $NaNO_2$, no reaction was observed.

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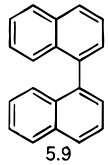
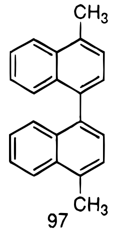
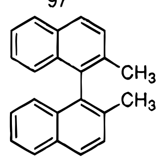
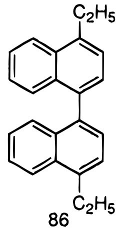
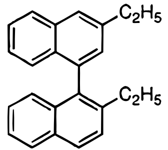
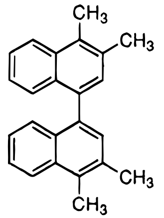
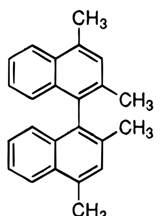
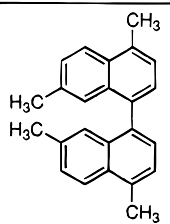
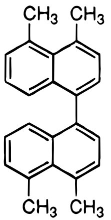
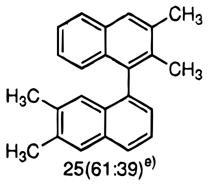
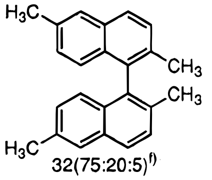
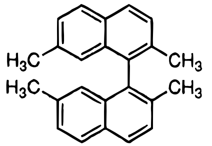
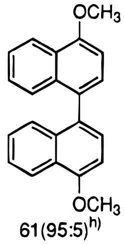
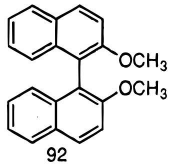
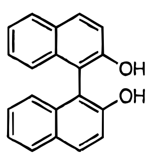
Table 1. Coupling Reaction of Naphthalene Derivatives[i]

substrate	temp. (°C)	solvent	product yield,%
naphthalene	0	CH_3CN	 5.9
1-methylnaphthalene	0	CH_3CN	 97
2-methylnaphthalene	0	CH_3CN	 71(50:38:12)[a]
1-ethylnaphthalene	0	CH_3CN	 86
2-ethylnaphthalene*	0	CH_3CN	 36(53:29:18)[b]
1,2-dimethylnaphthalene	-40	CH_3CN	 73(97:3)[c]
1,3-dimethylnaphthalene	-40	CH_3CN	 79
1,4-dimethylnaphthalene*	-40	CH_3CN	trace
1,5-dimethylnaphthalene*	-40	CH_3CN	trace
1,6-dimethylnaphthalene	-40	CH_3CN	 90(92:8)[d]
1,8-dimethylnaphthalene	-40	CH_3CN	 75
2,3-dimethylnaphthalene*	-40	CH_3CN	 25(61:39)[e]
2,6-dimethylnaphthalene*	-40	CH_3CN	 32(75:20:5)[f]
2,7-dimethylnaphthalene	-40	CH_3CN	 95(97:3)[g]
1-methoxynaphthalene	-78	$CH_3CN-CH_2Cl_2$	 61(95:5)[h]
2-methoxynaphthalene	-78	$CH_3CN-CH_2Cl_2$	 92
1-naphthol	-78	$CH_3CN-CH_2Cl_2$	0
2-naphthol	-78	$CH_3CN-CH_2Cl_2$	 68

* Unreacted substrate was recovered. [a] Isomer ratio of 2,2′-dimethyl-1,1′-binaphthyl:2,3′-dimethyl-1,1′-binaphthyl:other isomers. [b] Isomer ratio of 2,3′-diethyl-1,1′-binaphthyl:2,2′-diethyl-1,1′-binaphthyl:other isomers. [c] Isomer ratio of 3,3′,4,4′-tetramethyl-1,1′-binaphthyl:other isomers. [d] Isomer ratio of 4,4′,7,7′-tetramethyl-1,1′-binaphthyl:other isomers. [e] Isomer ratio of 2,3,6′,7′-tetramethyl-1,1′-binaphthyl:2,2′,3,3′-tetramethyl-1,1′-binaphthyl. [f] Isomer ratio of 2,2′,6,6′-tetramethyl-1,1′-binaphthyl:2,3′,6,7′-tetramethyl-1,1′-binaphthyl:other isomers. [g] Isomer ratio of 2,2′,7,7′-tetramethyl-1,1′-binaphthyl:other isomers. [h] Isomer ratio of 4,4′-dimethoxy-1,1′-binaphthyl:other isomers. [i] The reactions were carried out using 0.5 mmol of $NaNO_2$, 10 mmol of CF_3SO_3H, and 5 mmol of substrate in air for 1 h.

Table 2. Reaction Using Nitro-1-methylnaphthalene[a]

substrate composition 1-methylnaphthalene: nitro-1-methylnaphthalene (mmol)	products yield (%)				
	1	**2**	**3**	**4**	**5**
0:0.7	93	0	0	0	0
0.7:0.7	46	0	1.9	4.8	19
0.7:0	0	1.1	1.4	1.9	15

[a] The reactions were carried out using 1 mmol of $NaNO_2$, 10 mmol of CF_3SO_3H, and 50 mL of CH_3CN at 0 °C in air for 1 h. The yield was determined on the basis of the total amounts of substrates.

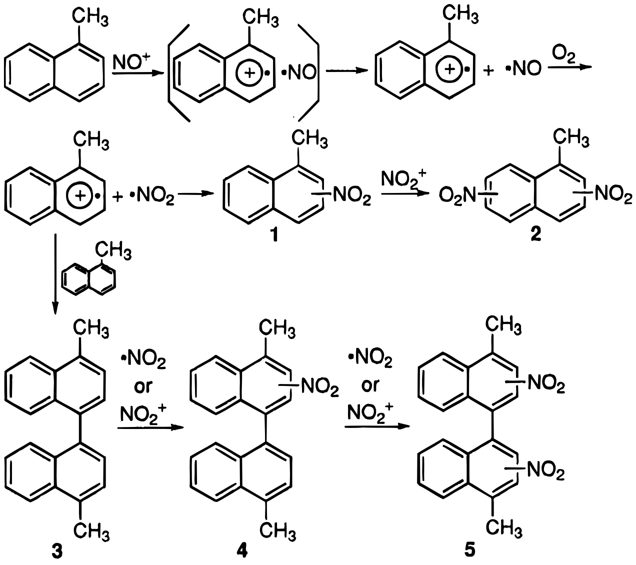
Table 3. Coupling Reaction of 1-Methylnaphthalene[a]

entry	oxidant	CF_3SO_3H (mmol)	atmosphere	products yield (%)				
				1	**2**	**3**	**4**	**5**
1	$NaNO_2$	10	air	0	0	97	1.5	0
2	$NaNO_2$	10	N_2	0	0	15	0	0
3	$NOBF_4$	10	air	0	0	90	0	0
4	$NOBF_4$	0	air	5	0	0.8	0	0
5	NO_2BF_4	10	air	0.7	0	86	0	0
6	NO_2SbF_6	10	air	2.3	0	55	0	0

[a] The reactions were carried out using 0.5 mmol of an oxidant, 5 mmol of 1-methylnaphthalene, and 50 mL of CH_3CN at 0 °C for 1 h.

drance is obviously an important factor for controlling the yield and regioselectivity during this reaction. A similar result is reported for the iodination reaction of aromatic compounds using ICl.[16]

Formation Path of Products. In the reaction of 1-methylnaphthalene with $NaNO_2$ in the presence of CF_3SO_3H, five products, nitro-1-methylnaphthalene (**1**), dinitro-1-methylnaphthalene (**2**), 4,4′-dimethyl-1,1′-binaphthyl (**3**), nitro-4,4′-dimethyl-1,1′-binaphthyl (**4**), and dinitro-4,4′-dimethyl-1,1′-binaphthyl (**5**) were formed, and the product composition was dependent on the combination of reagents. When the amount of $NaNO_2$ was small, the product **3** was produced mainly in the presence of CF_3SO_3H. With an increasing amount of $NaNO_2$, the nitration proceeded to give **1**, **2**, **4**, and **5**. On the other hand, an increase in the CF_3SO_3H amount caused the formation of **3**, **4**, and **5**. However, no nitroso products were obtained, which shows that NO^+ acts only as an oxidant for 1-methylnaphthalene. The identified isomers of **1** were 2-nitro-, 3-nitro-, 4-nitro-, 5-nitro, and 8-nitro-1-methylnaphthalene. However, in the case of **4**, only one isomer, 2-nitro-4,4′-dimethyl-1,1′-binaphthyl, was identified because of the difficulty in separation. The main isomers of **1** and **4** were 4-nitro-1-methylnaphthalene (about 60% in ratio) and 2-nitro-4,4′-dimethyl-1,1′-binaphthyl (about 80% in ratio), respectively. Isomers of **2** and **5** were not identified because of their difficulty in separation. Products **1** and **3** were clearly formed through the coupling reaction of the 1-methylnaphthalene radical cation with NO_2[17] or 1-methylnaphthalene,[10] respectively. On the other hand, there are two plausible reaction paths for **2**, **4**, and **5**. For **2**, the plausible reaction paths are the nitration of **1** with NO_2^+ and the coupling reaction of the **1** radical cation with NO_2. For **4** and **5**, the paths include the nitration (the electrophilic substitution by NO_2^+ and the coupling reaction by NO_2) of **3** or **4** and the coupling reaction of the 1-methylnaphthalene radical cation or the **1** radical cation with **1**, respectively. In order to investigate the formation path of the products, the reactions were carried out using a mixture of 1-methylnaphthalene and **1**, and the results are summarized in Table 2. When the reaction was conducted using only **1**, no reaction occurred. Therefore, **2** was produced by the nitration of **1** with NO_2^+ because the **1** radical cation was not formed by NO^+. Next, the reaction using equimolar amounts of 1-methylnaphthalene and **1** was carried out to give the same amount of **1** as the former reaction using only **1**, but the reaction using only 1-methylnaphthalene did not give **1** at all. These results show that **1** is inert under these conditions, and **4** and **5** were formed through the nitration of **3** with NO_2^+ or NO_2. Therefore, the formation path of the products can be described as follows.



Catalytic Cycle of NO^+. In order to investigate the catalytic cycle of NO^+, the reactions of 1-methylnaphthalene were carried out using $NaNO_2$ with CF_3SO_3H under different conditions and using $NOBF_4$, NO_2BF_4, or NO_2SbF_6 instead of $NaNO_2$. The results are tabulated in Table 3. The coupling reaction of 1-methylnaphthalene using $NaNO_2$ with CF_3SO_3H at 0 °C in air gave **3** and **4** in 97% and 1.5% yields, respectively (entry 1). However, when the reaction using the same amounts of materials was carried out under a N_2 atmosphere (entry 2), the yield of **3** decreased from 97% to 15%, and **4** was not formed. Therefore, the oxidation of NO with O_2 to form NO_2 after the electron transfer of NO^+ with 1-methylnaphthalene is one step of the cycle. Next, the reactions using $NOBF_4$ were conducted in the presence or absence of CF_3SO_3H. In the presence of CF_3SO_3H (entry 3), **3** was obtained in excellent yield similar to the reaction using $NaNO_2$ (entry 1). However, in the absence of CF_3SO_3H, only a small amount of 1-methylnaphthalene was converted to **1** as the main product and **3**, and unreacted substrate was recovered (entry 4). These results show that CF_3SO_3H protonates NO_2, N_2O_4, and $NO^+NO_3^-$ exactly to reproduce NO^+ with the formation

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(17) Eberson, L.; Radner, F. *Acta Chem. Scand.* **1986**, *40*, 71. When the reaction of 1-methylnaphthalene (5 mmol) with $NOBF_4$ (5 mmol) was carried out in the absence of CF_3SO_3H at −45 °C in CH_3CN (25 mL) with CH_2Cl_2 (25 mL), five isomers of nitro-1-methylnaphthalene, 2-nitro-, 3-nitro-, 4-nitro-, 5-nitro-, and 8-nitro-1-methylnaphthalene, were identified, and the isomer distribution was 4:1:84:8:3. This isomer distribution shows fairly good agreement with the results of the coupling nitration of 1-methylnaphthalene radical cation salt with NO_2 at −70 °C, 8:0:88:1:3 (see ref 15). Therefore, we concluded that the nitration is a radical reaction.

of an equal amount of NO_2^+.[18] Recently, it has been proposed that NO_2^+ has the ability to function not only as an electrophile but also as an oxidant, which oxidizes NO to form NO^+.[19] In order to study whether NO_2^+ acts as the oxidant of NO to form NO^+ or as an electrophile to form **1**, the reaction was conducted using NO_2BF_4 and NO_2SbF_6, which is known to always contain a small amount of NO^+ as an impurity, under the same conditions in the presence of CF_3SO_3H (entries 5 and 6). Similar to the reaction using $NOBF_4$ (entry 3), **3** was formed mainly, but the formation of **1** was observed.[20] These results clearly show that NO_2^+ acted as the electrophile under the conditions when the NO concentration was low, and the formation of **1** was not observed when $NaNO_2$ and $NOBF_4$ were used in the presence of CF_3SO_3H. Therefore, it seems that NO_2^+ acts as the oxidant of NO to produce NO^+ rather than as an electrophile to form **1** in the $NaNO_2-CF_3SO_3H$ system. These results suggest the following catalytic cycle of NO^+.[21]

$$NaNO_2 + 2CF_3SO_3H \rightarrow CF_3SO_3NO + CF_3SO_3Na + H_2O$$

$$2{\cdot}NO + O_2 \rightarrow 2{\cdot}NO_2$$

$$2{\cdot}NO_2 \rightleftharpoons N_2O_4 \rightleftharpoons NO^+NO_3^-$$

$$NO^+NO_3^- + 2CF_3SO_3H \rightarrow CF_3SO_3NO + CF_3SO_3NO_2 + H_2O$$

$${\cdot}NO + CF_3SO_3NO_2 \rightarrow CF_3SO_3NO + {\cdot}NO_2$$

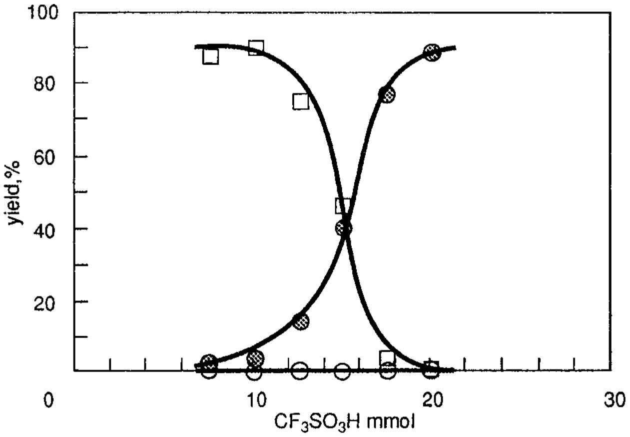

Figure 1. Coupling reaction of 1-methylnaphthalene using 5 mmol of $NaNO_2$. The reactions were carried out using 5 mmol of 1-methylnaphthalene at 0 °C in air for 1 h. The symbols □, ○, and ● represent the yields of **1**, **3**, and **4**, respectively, and **2** and **5** were not formed under these conditions.

Role of Acid. In the $NaNO_2-CF_3SO_3H$ system, the nitration of 1-methylnaphthalene was almost completely inhibited when $NaNO_2$ was used in a catalytic amount. Therefore, equimolar amounts of $NaNO_2$ and 1-methylnaphthalene were used with various amounts of CF_3SO_3H to investigate the influence of CF_3SO_3H on the nitration. The results are represented in Figure 1. The yield of **1** decreased with an increasing amount of CF_3SO_3H. On the contrary, the yield of **4** increased with an increase in CF_3SO_3H. This result reveals that CF_3SO_3H has the ability to inhibit the nitration of 1-methylnaphthalene with NO_2.[22] The ability of CF_3SO_3H to inhibit the nitration seems to stem from the ability to protonate NO_2 (N_2O_4 and $NO^+NO_3^-$) before the coupling reaction of NO_2 with the 1-methylnaphthalene radical cation.[21] In order to clarify this deduction, the reactions of 1-methylnaphthalene were conducted using several acids such as FSO_3H, CF_3SO_3H, and H_2SO_4 under the same conditions, and the influence of acid strength on the ability to inhibit the nitration was studied. The order of acid strength of these acids is well-known as $FSO_3H > CF_3SO_3H > H_2SO_4$. The results are shown in Table 4. The yield order of coupling products (the sum of **3**, **4**, and **5**) was consistent with the acid strength order. This result evidently shows that acids have two roles in this reaction, which are the production of NO^+ from $NaNO_2$ and the protonation of the formed NO_2 (N_2O_4 and $NO^+NO_3^-$) to

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[18] (a) Boughriet, A.; Wartel, M. *J. Chem. Soc., Chem. Commun.* **1989**, 809. (b) Bosch, E.; Rathore, R.; Kochi, J. K. *J. Org. Chem.* **1994**, *59*, 2529. (c) Bosch, E.; Kochi, J. K. *J. Org. Chem.* **1994**, *59*, 3314. (d) Bosch, E.; Kochi, J. K. *J. Org. Chem.* **1995**, *60*, 3172. It is reported that N_2O_4 which is in the equilibrium with NO_2 dissociates to $NO^+NO_3^-$ in a solvent (see ref 16). In the presence of a strong acid such as CF_3SO_3H, NO_3^- is evidently converted to NO_2^+ and H_2O.

[19] (a) Morrison, J. D.; Stanney, K.; Tedder, J. M. *J. Chem. Soc., Perkin Trans. 2* **1981**, 967. (b) Clemens, A. H.; Ridd, J. H.; Sandall, J. P. B. *J. Chem. Soc., Perkin Trans. 2* **1984**, 1659. (c) Clemens, A. H.; Ridd, J. H.; Sandall, J. P. B. *J. Chem. Soc., Perkin Trans. 2*, **1984**, 1667. (d) Clemens, A. H.; Helsby, P.; Ridd, J. H.; Al-Omran, F.; Sandall, J. P. B. *J. Chem. Soc., Perkin Trans. 2* **1985**, 1217. (e) Lee, K. Y.; Kuchyčan, D. J.; Kochi, J. K. *Inorg. Chem.* **1990**, *29*, 4196. (f) Lee, K. Y.; Amatore, C.; Kochi, J. K. *J. Phys. Chem.* **1991**, *95*, 1285. (g) Eberson, L.; Gonzalez-Luque, R.; Lorentzon, J.; Merchan, M.; Roos, B. O. *J. Am. Chem. Soc.* **1993**, *115*, 2898.

[20] (a) Eberson, L.; Radner, F. *Acta Chem. Scand.* **1984**, *38*, 861. (b) Barnett, J. W.; Moodie, R. B.; Schofield, K.; Taylor, P. G.; Weston, J. B. *J. Chem. Soc., Perkin Trans. 2* **1979**, 747. (c) Roberts, R. M. G.; Sadri, A. R. *Tetrahedron* **1983**, *39*, 137. (d) Tanaka, M.; Fujiwara, M.; Ando, H. *J. Org. Chem.* **1995**, *60*, 2106. (e) Eberson, L.; Hartshorn, M. P.; Radner, F. *Acta Chem. Scand.* **1994**, *48*, 937. The results are very interesting because both yields of **1** (entries 5 and 6) were significantly less than with $NOBF_4$ (entry 4). To explain these results, there are two plausible interpretations, the oxidation of 1-methylnaphthalene by NO_2^+ to produce radical cations and the inhibition of NO_2^+ attack to 1-methylnaphthalene by the protonation. The former has been studied widely, but is still controversial (see refs 12, 15, and 18a). The latter is derived from the fact that the protonation inhibits electrophilic substitutions (see ref 18b–d) and means that NO_2^+ attack may be inhibited more strongly than NO^+ attack to 1-methylnaphthalene by the protonation; namely, NO^+ reaction proceeds as the main reaction because NO_2^+ and NO^+ reactions seem to proceed through inner- and outer-sphere reactions, respectively (see refs 12, 15, and 18a,e). We are now studying this problem using superacids.

[21] The catalytic cycle in ref 13 is significantly different from our proposal and is shown as follows.

$$2ArH + 2NO^+ \rightarrow ArAr + 2NO + 2H^+$$

$$2NO + 2H^+ + 0.5O_2 \rightarrow 2NO^+ + H_2O$$

$$NO + NO_2 + 2H^+ \rightarrow 2NO^+ + H_2O$$

Concerning the catalytic cycle of NO^+, it is necessary to explain the inhibition of the nitration by superacids, namely, the role of superacids as NO_2 scavengers. However, in ref 13, the role of acids is only to produce NO^+, and there is no description about the role of acids in inhibiting the nitration. Therefore, we proposed that superacids can act as NO_2 scavengers by the protonation of N_2O_4 and $NO^+NO_3^-$, not NO_2. The important concept is that O_2 oxidizes two NO to give two NO_2; therefore, the NO_2 produced by O_2 may exist as N_2O_4 or $NO^+NO_3^-$ first, and then, N_2O_4 and $NO^+NO_3^-$ are converted to NO^+ and NO_2^+ by superacids before the coupling reaction of the radical cation with NO_2 occurs to give nitro compounds.

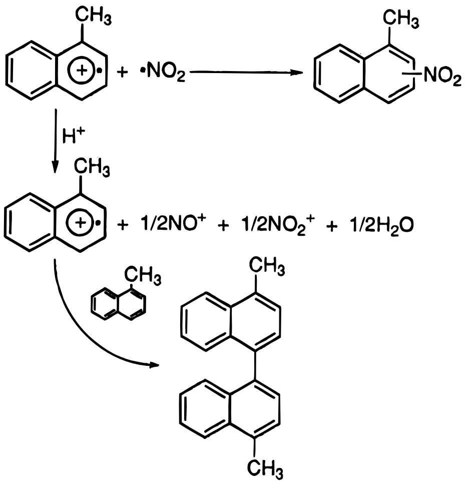
[22] The catalytic cycle of NO^+ shows that the concentration of NO_2 in the reaction systems is dependent not only on the superacid concentration (or the acid strength of superacids) but also on the reaction time. According to the catalytic cycle, the concentration of NO_2 in the presence of superacids is low at the initial stage of the reaction and increases with the progress of the reaction by the formation of NO_2 from NO_2^+. Therefore, it is difficult to inhibit the nitration of produced binaphthyl derivatives by the addition of superacids because radical cations of binaphthyl derivatives are a secondary product.

Table 4. Influence of Acid Strength[a]

	products yield (%)				
acid	**1**	**2**	**3**	**4**	**5**
FSO_3H	23	0	2.1	64	0
CF_3SO_3H	48	0	0	39	0
H_2SO_4	54	8.3	1.1	1.6	0

[a] The reactions were carried out using 5 mmol of $NaNO_2$, 15 mmol of acid, 5 mmol of 1-methylnaphthalene, and 50 mL of CH_3CN at 0 °C in air for 1 h.

inhibit the nitration and to produce NO^+ and NO_2^+. Strong acids give the formation of biaryls an advantage over the formation of nitro compounds.



Conclusions

It was found that the coupling reaction of naphthalene derivatives using $NaNO_2$ with CF_3SO_3H in CH_3CN catalytically gave the corresponding 1,1′-binaphthyl derivatives in air. The reaction is strongly influenced by steric hindrance; therefore, high regioselectivity was observed in some substrates. The acids have two roles in the reaction, which are the production of NO^+ and the inhibition of the nitration by the protonation of NO_2 (N_2O_4 and $NO^+NO_3^-$). Therefore, strong acids are necessary to more preferentially produce binaphthyl derivatives than nitro compounds.

Experimental Section

All reagents were of the highest available purity and were used without further purification. Dry CH_3CN was used when $NOBF_4$, NO_2BF_4, and NO_2SbF_6 were used. The product yields were determined with GC by the internal standard method (experimental error = ±5%). Nitrobenzene was used as an internal standard, and the separated products were used for the yield determination. The product structures were identified by NMR (^{1}H-, ^{13}C-NMR, COSY, NOESY, CHSHF, and COLOC) and mass analysis after isolation by recrystallization or GPC.

Coupling Reaction Procedures Using $NaNO_2$, $NOBF_4$, NO_2BF_4, and NO_2SbF_6. The required amounts of oxidant and acid were placed in a three-necked flask (300 mL) containing 50 mL of solvent (40 mL for solid substrates such as naphthalene, 1,4-, 1,5-, 1,8-, 2,3-, 2,6-, and 2,7-dimethylnaphthalene, 2-methoxynaphthalene, and 2-naphthol), and a substrate was added to the mixture with vigorous stirring in air under temperature control. When the substrate was a solid, the solution of the substrate dissolved in a suitable solvent was added to a mixture (10 mL of CH_3CN for naphthalene, and 1,4-, 1,8-, and 2,7-dimethylnaphthalene, 20 mL of CH_3CN for 2,3-dimethylnaphthalene, a mixture of CH_3CN (5 mL) and CH_2Cl_2 (5 mL) for 2-methoxynaphthalene and 2-naphthol, and a mixture of CH_3CN (10 mL) and CH_2Cl_2 (10 mL) for 1,5- and 2,6-dimethylnaphthalene). After 1 h, the reaction mixture was poured into ice water and extracted with $CHCl_3$. The product yields and structures were determined according to the general procedures.

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Supporting Information Available: Discussion of the influence of solvents on the coupling reaction and the Scholl reaction using SbF_5 (6 pages). This material is contained in libraries on microfiche, immediately follows this article in the microfilm version of the journal, and can be ordered from the ACS; see any current masthead page for ordering information.

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