Regular Article

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Development of Triazine-Based Benzylating Reagents Possessing *t*-Butyl Group on the Triazine Core: Thermally Controllable Reagents for the Initiation of Reaction

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Benzylating reagents, 4-(4,6-di-*t*-butyl-1,3,5-triazin-2-yl)-4-benzylmorpholinium triflate, and related derivatives have been developed. The reagents release benzyl triflate as a benzyl cation equivalent upon heating the solution to 40°C under neutral conditions. The *O*-benzylation of alcohols using a stoichiometric amount of these reagents afforded corresponding benzyl ethers in good to high yields. This was due to the presence of a bulky *t*-butyl group on the triazine ring of these reagents that prevents the consumption of benzyl triflate *via* a side reaction with a morpholinotriazine derivative.

Key words benzylating reagent; triazine; thermally controllable

The benzyl group is a protecting group for alcohols that is stable under strong acidic and basic conditions, and its deprotection is easily achieved *via* hydrogenolysis under mild conditions. Therefore, the benzyl group has been used for several organic synthetic reactions and various benzylation methods have been developed.[1,2] Recently, we have developed several alkylating reagents based on the characteristics of 1,3,5-triazine. 2,4,6-Tris(benzyloxy)-1,3,5-triazine (TriBOT),[3,4] 2,4,6-tris(*p*-methoxybenzyloxy)-1,3,5-triazine (TriBOT-PM),[5] 6-(benzyloxy)-1,3,5-triazine-2,4(1*H*,3*H*)-dione (MonoBOT),[6,7] 2,4,6-tris(*t*-butoxy)-1,3,5-triazine (TriAT-*t*Bu),[8] 2,4,6-tris(allyloxy)-1,3,5-triazine (TriAT-allyl),[9] and *N*,*N*′-dimethylated 6-(benzyloxy)-1,3,5-triazine-2,4(1*H*,3*H*)-dione (DMBOT)[10] achieve the alkylation of various alcohols in good to excellent yields in the presence of a catalytic amount of an acid. 4-(4,6-Diphenoxy-1,3,5-triazin-2-yl)-4-benzylmorpholinium triflate (DPT-BM (**X-a**))[11,12] converts acid- or alkali-labile alcohols into corresponding benzyl ethers because of the nearly neutral conditions (Fig. 1).

DPT-BM (**X-a**) has several advantages compared with other representative benzylating reagents, such as benzyl triflate (BnOTf)[13] and 2-benzyloxy-1-methylpyridinium triflate (Dudley reagent),[14–18] available under neutral conditions. DPT-BM (**X-a**) is a non-hygroscopic and stable solid, allowing its treatment under atmospheric conditions. This reagent has a good reactivity and can release BnOTf as a benzyl cation species at room temperature, when dissolved in solvents. However, one major drawback of DPT-BM (**X-a**) is a slightly remaining nucleophilic property of morpholinotriazine (**Y-a**), a co-product arising from DPT-BM (**X-a**), because it consumes BnOTf to form a regioisomeric *N*-benzyl triazinium salt (**Z-a**) as a byproduct, which no longer acts as a benzylating reagent under the reaction conditions. Although **Y-a** is less nucleophilic, the side reaction proceeds at the most nucleophilic site of the triazine nitrogen owing to the high electrophilicity of BnOTf. Thus, an excess of DPT-BM (**X-a**) (>2.0 equiv.) is required to obtain benzyl ethers in good yields. Despite its salt structure, the solubility of the DPT-BM (**X-a**) in dimethoxyethane (DME) is higher than that of the non-charged **Y-a** (the saturation solubility in DME: 0.12 mol/L). By exploiting this property, the production of byproduct **Z-a** can be reduced by performing the reaction in a high concentration of alcohols. Since a significant amount of **Y-a** is precipitated, the relative concentration of **Y-a** is decreased compared with that of the alcohols. This results in relatively increasing the benzylation rate of the alcohols compared to that of **Y-a**. However, such conditions are not suitable for alcohols exhibiting a low reactivity or a low solubility in DME. When the *O*-benzylation of a sterically hindered alcohol (**1a**) with DPT-BM (**X-a**) (1.2 equiv.) in the presence of MS5A, which was used as a dehydrating reagent to remove residual moisture, was performed at concentrations of 1.0 and 0.2 mol/L, the yields of **2a** were insufficient (86 and 76%), although a better result was obtained when a higher concentration of **1a** was used (Table 1, entries 1, 2).

We envisioned that increasing steric hindrances around the

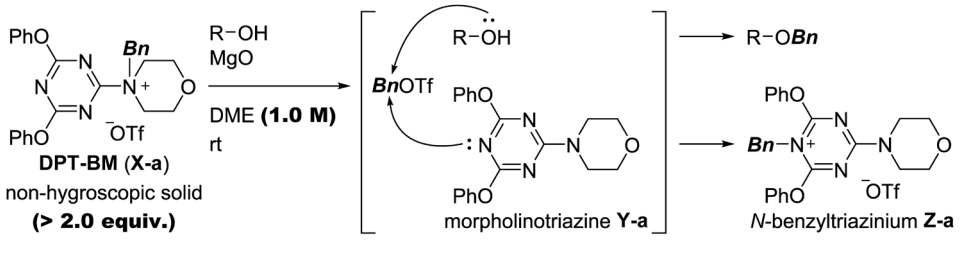

Fig. 1. The Benzylation with DPT-BM (**X-a**) Proceeds under Nearly Neutral Conditions

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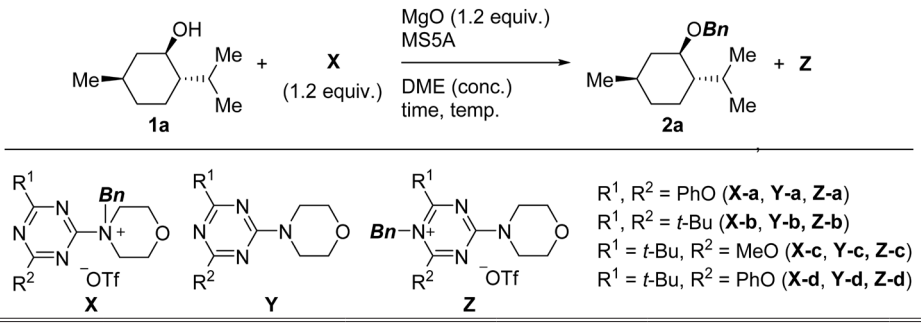
Table 1. *O*-Benzylation of Sterically Hindered Alcohol **1a** with Benzylating Reagent **X**



Entry	X	Conc. of **1a** [mol/L]	Time [h]	Temp. [°C]	Yield of **2a** [%][a)]	Yield of **Z** [%][a)]
1	**X-a**	1.0	2	rt	86	22 (**Z-a**)
2	**X-a**	0.2	2	rt	76	29 (**Z-a**)
3	**X-b**	0.2	56	rt	93	0 (**Z-b**)
4	**X-b**	0.2	4	40	93	0 (**Z-b**)
5	**X-c**	0.2	4	40	92	1 (**Z-c**)
6	**X-d**	0.2	4	40	93	0 (**Z-d**)

a) Yields were calculated by ^{1}H-NMR analysis using an internal standard.

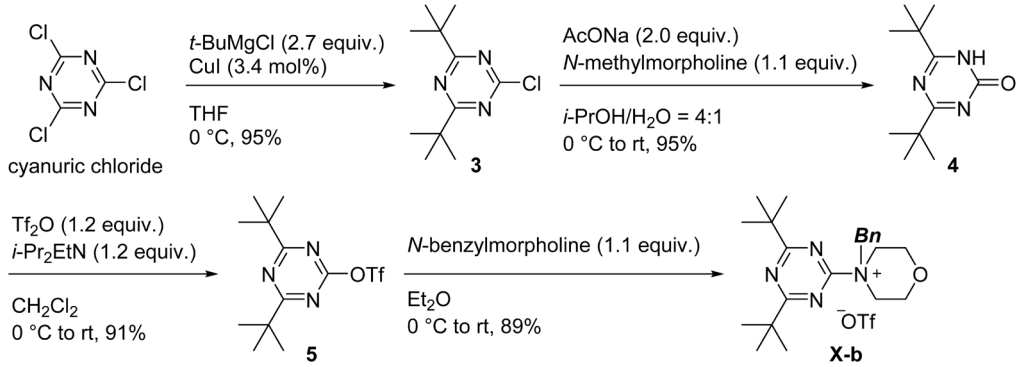


Chart 1. The Synthetic Scheme of the New Benzylating Reagent (**X-b**)

nitrogen atoms on the triazine ring might prevent the formation of *N*-benzyltriazinium compounds that correspond to **Z-a**. In this paper, we report the development of a new benzylating reagent (**X-b**) that was designed by replacing the two phenoxy groups of DPT-BM (**X-a**) with a sterically bulky *t*-butyl group.

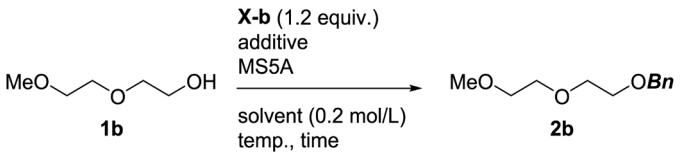
The benzylating reagent, **X-b**, was prepared according to the procedure shown in Chart 1. The copper-catalyzed cross-coupling of *t*-butyl magnesium chloride with cyanuric chloride afforded 2,4-di-*t*-butyl-6-chloro-1,3,5-triazine (**3**),[19] which was converted into 2,4-di-*t*-butyl-6-trifluoromethanesulfonyloxy-1,3,5-triazine (**5**) *via* 4,6-di-*t*-butyl-1,3,5-triazin-2(1*H*)-one (**4**) using our original method.[6,7,10,20] The treatment of **5** with *N*-benzylmorpholine in Et_2O afforded 4-(4,6-di-*t*-butyl-1,3,5-triazin-2-yl)-4-benzylmorpholinium triflate (**X-b**) as a precipitate in 89% yield.

To compare the benzylating ability of **X-b** with that of DPT-BM (**X-a**), the benzylation of **1a** was performed under the same condition shown in Table 1, entry 2. As a result, the reaction proceeded to afford **2a** in 93% yield, and the generation of the corresponding *N*-benzyltriazinium **Z-b** was not detected probably because the introduction of bulky *t*-butyl groups to the triazine ring prevented the **Z-b** formation (Table 1, entry 3). However, the completion of benzylation required a longer time than Table 1, entry 2. This would be because the liberation of the benzyl cation species, BnOTf, from **X-b** was slower than that from DPT-BM (**X-a**) due to the replacement of electron-withdrawing phenoxy groups with the electron-donating *t*-butyl groups, which would decrease the leaving ability of **Y-b**. Heating at 40°C shortened the reaction time to 4 h and improved the yield to 93% (entry 4).

To accelerate the *O*-benzylating reaction with **X-b**, we investigated the optimal conditions using primary alcohol, **1b**, as a model alcohol (Table 2). Under the same condition as in Table 1, entry 3, the reaction was slow and required 56 h for its completion (entry 1). Despite the addition of a catalytic amount of triflic acid to activate **X-b**, the reaction time could not be reduced (entry 2). When the reaction was conducted at 40°C, **1b** completely disappeared within 5 h, and product **2b** was afforded in 95% yield (entry 3). Moreover, the reaction at 60°C completed within 1 h (entry 4). Compared with entry 3, when the reaction was performed at a higher concentration, the yield of **2b** decreased and a longer reaction time was required (entry 5).[21] The yield of **2b** decreased to 80% in CH_2Cl_2 (entry 6), whereas high yields were maintained in ethereal solvents such as 1,4-dioxane and diglyme (entries 7, 8).

We examined the scope and limitations of various alcohols for the benzylation using **X-b** at optimal conditions mentioned

Table 2. The Screening of the Benzylation of Primary Alcohol **1b** Using **X-b**



Entry	Solvent	Additive	Temp. [°C]	Time [h]	Yield [%][a)]
1	DME	MgO (1.2 equiv.)	rt	56	91
2	DME	TfOH (1 mol%)	rt	54	81
3	DME	MgO (1.2 equiv.)	40	5	95
4	DME	MgO (1.2 equiv.)	60	1	97
5[b)]	DME	MgO (1.2 equiv.)	40	28	89
6	CH_2Cl_2	MgO (1.2 equiv.)	40	24	80
7	1,4-Dioxane	MgO (1.2 equiv.)	40	10	95
8	Diglyme	MgO (1.2 equiv.)	40	9.5	96

a) Yields were calculated by ^{1}H-NMR analysis using an internal standard. *b*) The reaction was performed in 1.0 mol/L DME solution of **1b**.

in the entries 3, 4 in Table 2. The results are summarized in Table 3. For comparison, the yields using DPT-BM (**X-a**) or Dudley reagent that were reported in the literature are also listed. The benzylation of simple primary alcohols, **1b** and **1c**, proceeded in high yields (entries 1, 2). Bulky secondary and tertiary alcohols, **1a** and **1d**–**f**, were converted into their corresponding benzyl ethers in >80% yields which are comparable to those using DPT-BM (**X-a**) (entries 3, 5, 7, 9). When the reaction temperature was increased to 60°C, the reactions completed within 1 h without a decrease of the yields (entries 4, 6, 8, 10). Base-labile alcohols, such as **1g** and **1h**, possessing a chloroalkyl or acetoxy group, provided benzyl ethers, **2g** and **2h**, in high yields (entries 11, 12). *β*-Hydroxyester **1i** and 2-(trimethylsilyl)ethanol **1j**, which are prone to decompose under acidic or basic conditions, were converted into products **2i** and **2j**, respectively, in good yields (entries 13, 14). No racemization was observed during the benzylation of **1i**. The *O*-benzylation of cholesterol **1k** also afforded the corresponding ether **2k** in good yields (entries 15, 16). Although tertiary alcohols, such as **1l** and **1m**, and their ethers (**2l** and **2m**) are known to decompose under acidic or heating conditions, **2l** and **2m** were obtained without decomposition using approximately stoichiometric amount of **X-b** (entries 17–20). In particular, better yields were observed at 40°C, which is milder than the temperature used for Dudley reagent.

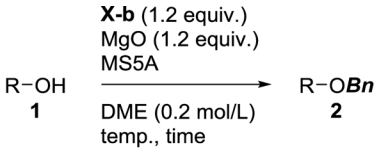
The formation of *N*-benzyltriazinium **Z-b** from **X-b** was not observed in all the reactions listed in Table 3. Therefore, we synthesized the benzylating reagents possessing a *t*-butyl and an alkoxy group on the triazine ring (**X-c** and **X-d**) to study the effect of *t*-butyl group on the formation of the corresponding *N*-benzyltriazinium (**Z-c** and **Z-d**). The benzylation of **1a** using **X-c** or **X-d** resulted in the formation of **2a** in higher yields compared with that obtained using DPT-BM (**X-a**), and it did not produce *N*-benzyltriazinium **Z-c** or **Z-d** (Table 1, entries 5, 6).

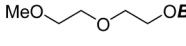
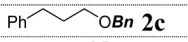
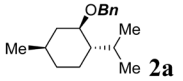
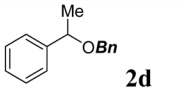
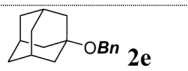
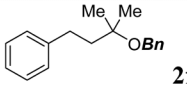
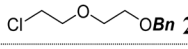
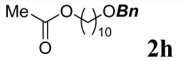
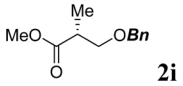
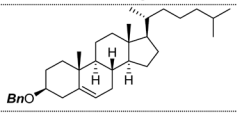
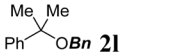
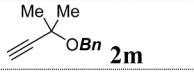
To compare the relative nucleophilicities of the morpholinotriazines **Y**,[22)] which are co-products produced from **X** and could undergo *N*-benzylation to form *N*-benzyltriazinium **Z**, crossover experiments were performed. A mixture of the equimolar amount of morpholinotriazines, **Y-a**, **Y-c**, and **Y-d**, was treated with the same amount of **X-b**, which is a source of benzyl cation species. As a result, *N*-benzyltriazinium, **Z-a**, **Z-b**, **Z-c**, and **Z-d**, were produced in a ratio of 94 : 0 : 4 : 2, respectively (Chart 2[A]). A similar reaction was conducted in the absence of **Y-a** because the relative nucleophilicities of **Y-b**, **Y-c**, and **Y-d** were ambiguous owing to the low yields of **Z-b**, **Z-c**, and **Z-d**, which result from the high nucleophilicity of **Y-a**. As a result, **Z-b**, **Z-c**, and **Z-d** were formed in a ratio of 0 : 77 : 23, respectively (Chart 2[B]). To conduct the reaction in the presence of one equivalent of **Y-b**, other benzylating reagents were used instead of **X-b**. The reaction employing DPT-BM (**X-a**) in the presence of **Y-b**, **Y-c**, and **Y-d** also produced a similar ratio of products as in reaction [A] (Chart 2[C]). When **X-d** was employed as the benzylating reagent in the presence of **Y-b** and **Y-c**, only a slight increase in **Z-c** was observed, and **Z-b** was not produced (Chart 2[D]). These experimental results clearly indicate that **Y-b** that possesses two *t*-butyl groups on the triazine ring does not react with a benzyl cation species at all.

As we previously reported,[11)] these results again indicate that the formation of **Z** proceeds intermolecularly between BnOTf and **Y**, and not *via* an intramolecular rearrangement. As expected, the introduction of a bulky *t*-butyl group into the triazine core caused a decrease in the rate of the *N*-benzylation of **Y**; therefore, the *O*-benzylation of alcohols produced high yields by using only 1.2 equiv. of **X-b**, **X-c**, or **X-d**.

We reported that DPT-BM (**X-a**) dissolved in $CDCl_3$ disappeared according to first-order kinetics, and the release of the benzyl cation species from DPT-BM (**X-a**) may occur *via* a nucleophilic attack to the *N*-benzyl group by the triflate counter anion[11)] (S_N2 reaction, Fig. 2). Thus, we conducted a kinetic study of the degradation of **X-b**, **X-c**, and **X-d** because the release rate of BnOTf from **X** should be affected by substituents on the triazine ring. As a result, all the reagents degraded according to first-order kinetics. The order of the first-order reaction rate constants, *k* of **X**, were determined to be DPT-BM (**X-a**)>**X-d**>**X-c**>**X-b** (Table 4). This result can be accounted for by the electronic effect of the substituents on the triazine ring. Based on the Hammett's substituent constant, σ_m,[23)] the electron-withdrawing ability of the phenoxy group is larger than that of the methoxy group. In contrast, the *t*-butyl group has an electron-donating ability. The observed first-order rate constants of **X** are assumed to correlate with the leaving ability of **Y** altered by the substituents on the triazine. It has been reported that the Hammett equation is applicable to triazine derivatives using the sum of σ_m of two substituents on the tri-

Table 3. Benzylation of Several Alcohols Using **X-b**, and the Comparison of Benzylating Ability with Other Benzylating Reagents

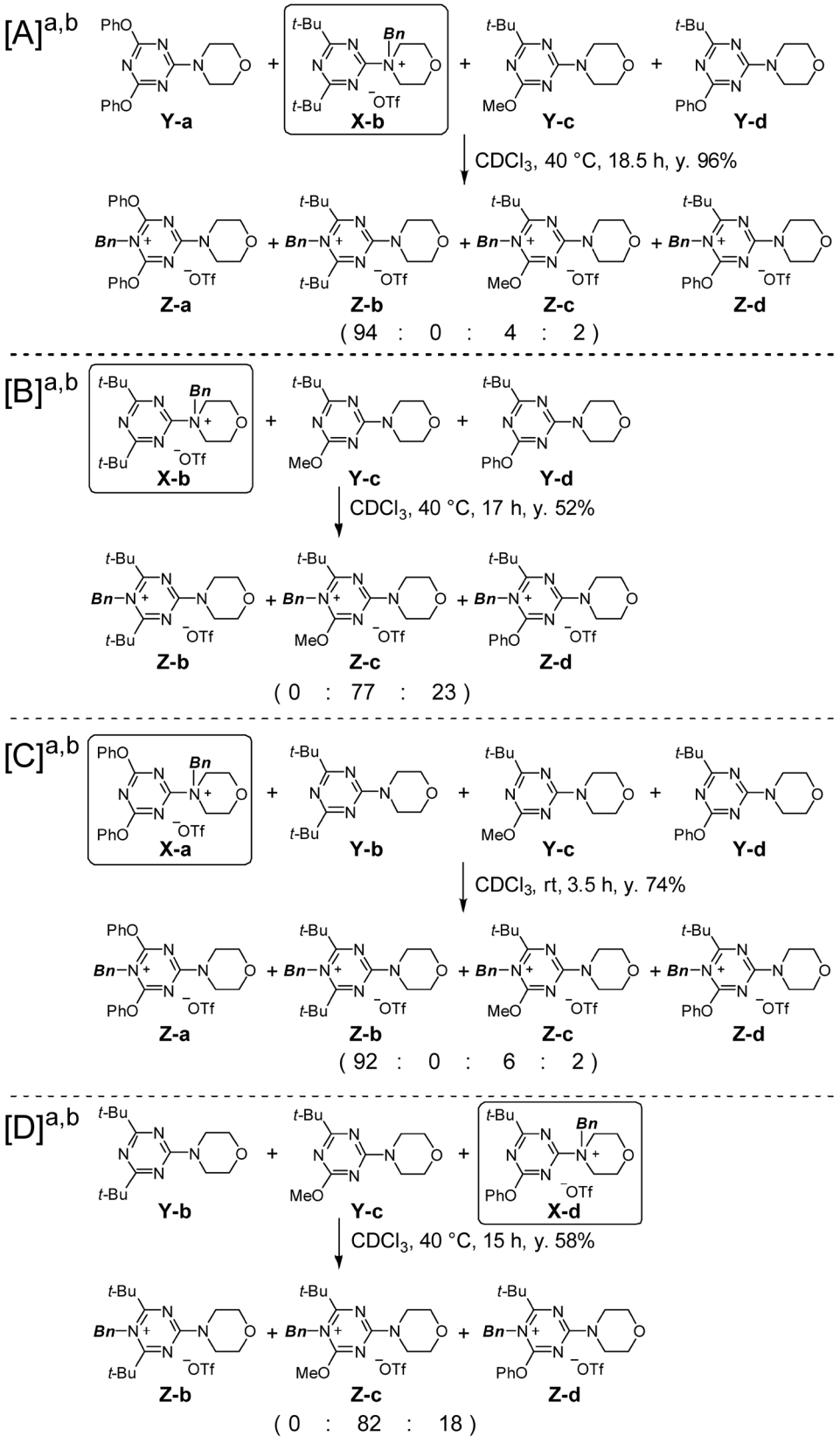


entry	product	temp. [°C]	time [h]	yield [%][a]	yield using other reagents [%]
1	 **2b**	40	5	92	97[b]
2	 **2c**	40	5	95[c]	95[b,d]
3	 **2a**	40	4	94	86[b]
4	**2a**	60	0.5	91	86[b]
5	 **2d**	40	9	85	61[e] (85[b,d])
6	**2d**	60	1	81	61[e] (85[b,d])
7	 **2e**	40	5.5	82	84[f]
8	**2e**	60	1	86	84[f]
9	 **2f**	40	7.5	81	78[g] (91[d,h])
10	**2f**	60	1	72	78[g] (91[d,h])
11	 **2g**	40	6	94	97[b]
12	 **2h**	40	5	89	90[b]
13	 **2i**	40	5	85	82[b]
14	**2j**	40	5	72	86[b]
15	 **2k**	40	17.5	82	n.d.[i,j]
16	**2k**	60	1	87	n.d.[i,j]
17	 **2l**	40	3.5	65	44[i]
18	**2l**	60	40 min	39	44[i]
19	 **2m**	40	5	73	59[k]
20	**2m**	60	40 min	50	59[k]

a: Isolated yields. b: **X-a** (2.0 equiv.), MgO (2.0 equiv.), DME (1.0 mol/L), r.t., 2 h.[11] c: The yield was determined by ^{1}H-NMR analysis because a small amount of **Y-b** could not be separated from **2c**. d: The yield was determined by ^{1}H-NMR analysis because a small amount of Bn_2O could not be separated from **2**. e: **X-a** (2.0 equiv.), MgO (2.0 equiv.), DME (1.0 mol/L), r.t., 4 h.[11] f: **X-a** (3.0 equiv.), MgO (3.0 equiv.), DME (1.0 mol/L), r.t., 19 h.[11] g: **X-a** (3.0 equiv.), MgO (3.0 equiv.), DME (1.0 mol/L), r.t., 7 h.[11] h: **X-a** (3.0 equiv.), MgO (3.0 equiv.), DME (1.0 mol/L), r.t., 2 h.[11] i: Dudley reagent (2.0 equiv.), MgO (2.0 equiv.), $PhCF_3$ (0.1–0.5 mol/L), 83°C, 24 h.[14] j: The authors described that the yield was not determined because a small amount of Bn_2O could not be separated from **2k**.[15] k: Dudley reagent (2.0 equiv.), $PhCF_3$, 120°C (MW), 20 min.[17]

azine ring.[24–26] In the case of these benzylating reagents, the Hammett plot for the degradation rate also provides a good linear relation (Fig. 3). The positive ρ value (3.471) shows that electron-withdrawing groups stabilize the transition state by delocalization of the electron density increasing on the morpholine nitrogen. These results indicate that the reactivity of **X** can be modulated by the inclusion of electron-donating or electron-withdrawing substituents on the triazine ring using the σ_m as an indicator.

The low reactivity of **X-b** is an advantage from an experimental point of view. The benzylation using DPT-BM (**X-a**) must be initiated by adding the reagent in the solid state to



a: These crossover experiments were performed in the presence of an equimolar amount of morpholinotriazines and a benzylating reagent. b: The product ratio was determined by ^{1}H-NMR analysis.

Chart 2. The Crossover Experiments of **Y** in the Presence of Benzyl Cation Species

the alcohol solution under anhydrous conditions because DPT-BM (**X-a**) rapidly decomposes when dissolved in a solvent. In contrast, the benzylating reaction with **X-b** can be conducted using a pre-prepared solution of **X-b**, which can be added to a solution of reactants because **X-b** has a sufficient stability to allow it to be dissolved in a solvent at room temperature.

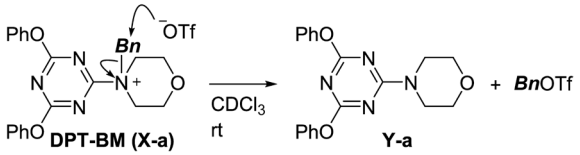


Fig. 2. The Generation of Benzyl Cation Species from DPT-BM (**X-a**)

Table 4. First-Order Reaction Rate Constant k of the Benzylating Reagents **X**

X → ($CDCl_3$ (21.2 mmol/L), 20–23 °C) → **Y** + **Z** + ***Bn***OTf

X	$k\times10^{-3}$ (/min)
X-a	35.5
X-b	0.1
X-c	2.5
X-d	5.0

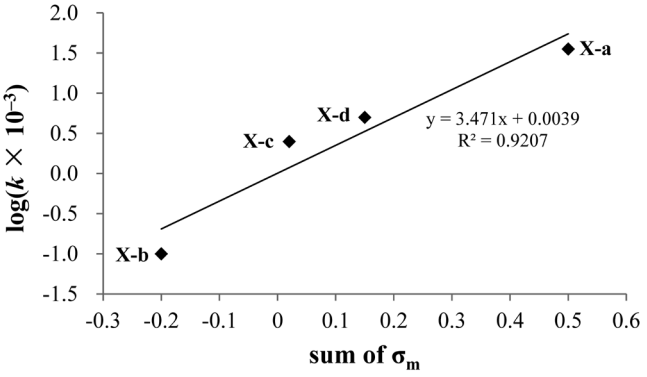


Fig. 3. Hammett Plot for Degradation of the Benzylating Reagents **X**[a)]

a) The $\log(k\times10^{-3})$, which is based on k in Table 4, *vs.* the sum of σ_m are shown in this plot. See the supporting information for details.

Therefore, the initiation and rate of benzylating reaction can be controlled by heating the reactant solution prepared at room temperature to an appropriate temperature.

In conclusion, we successfully developed a new benzylating reagent, **X-b**, by replacing the phenoxy groups on DPT-BM (**X-a**) with bulky *t*-butyl groups. The incorporation of two *t*-butyl groups into the triazine ring of the benzylating reagent inhibited the side reaction to form *N*-benzyltriazinium **Z-b** because the morpholinotriazine **Y-b** cannot undergo reaction with BnOTf owing to the steric hindrance of the *t*-butyl groups. Hence, the *O*-benzylation of alcohols with a stoichiometric amount of **X-b** (1.2 equiv.) afforded benzyl ethers in good to high yields. A solution of **X-b** is stable, allowing its treatment at room temperature; however, the benzylation proceeds at 40°C. As a result of the kinetic study, the electronic effect of the substituents installed on the triazine ring affects the reactivity of the benzylating reagents. This result indicates that the leaving abilities of the morpholinotriazines correlate with the reactivity of the corresponding benzylating reagent and can be controlled by the substituents on the triazine core.

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Conflict of Interest The authors declare no conflict of interest.

Supplementary Material The online version of this article contains supplementary materials (detailed experimental procedure, physical data, and NMR spectra of isolated products).

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