BASE-CATALYZED REARRANGEMENT OF α-BENZOTRIAZOLYL ALKOXIDE ANIONS: SYNTHESIS OF ONE-CARBON HOMOLOGATED α-SUBSTITUTED ALKYL KETONES*

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Abstract — Deprotonated benzotriazole derivatives (**1a-c**) reacted with aldehydes to give the α-oxyanion substituted intermediates (**2**), which upon further treatment with butyllithium, without separation, resulted in the formation of one-carbon homologated α-substituted alkyl ketones (**6a-f**). Similar treatment of benzotriazole derivatives (**1d-f**) generated the benzotriazole ring-opening and rearranged products (**7a-f**). Plausible mechanisms for these reactions have been proposed.

Carbon insertion for the one-carbon homologation of aldehydes and ketones is of great importance in organic synthesis.[1] Extensive methodology developed by others for this purpose was summarized in our recent publications, which also described our novel and efficient insertion routes to one-carbon homologated α-alkoxy-, α-alkenyl-, α-aryl- and α-phenylthioalkyl ketones by Lewis acid ($ZnBr_2$) induced pinacol-type rearrangements of benzotriazole-stabilized anions (**2**).[2] Many of the approaches reported earlier utilized base-induced carbenoid-type insertions: the α-oxyanion substituted intermediate from the reaction of a conjugated base of a dihetero-substituted system (RCHXY) and an aldehyde or a ketone was further treated with a strong base to remove one of the hetero-substituents (X or Y), following rearrangement of a hydrogen atom (in the case of an aldehyde) or an alkyl (aryl) group (for ketones) to give the insertion products. The ease of the reactions and the nature of final products are strongly influenced by the properties of the X and Y groups. This concept has been utilized for both the carbon chain extension and ring enlargement, using as inserting reagents: dihaloalkanes,[3] phenyl thioacetals,[4]

chloroalkyl sulfoxides[5] and methoxy sulfones.[6] In this manner, α-phenylthio, α-chloro, α-alkyl and α-methoxy groups have been introduced.

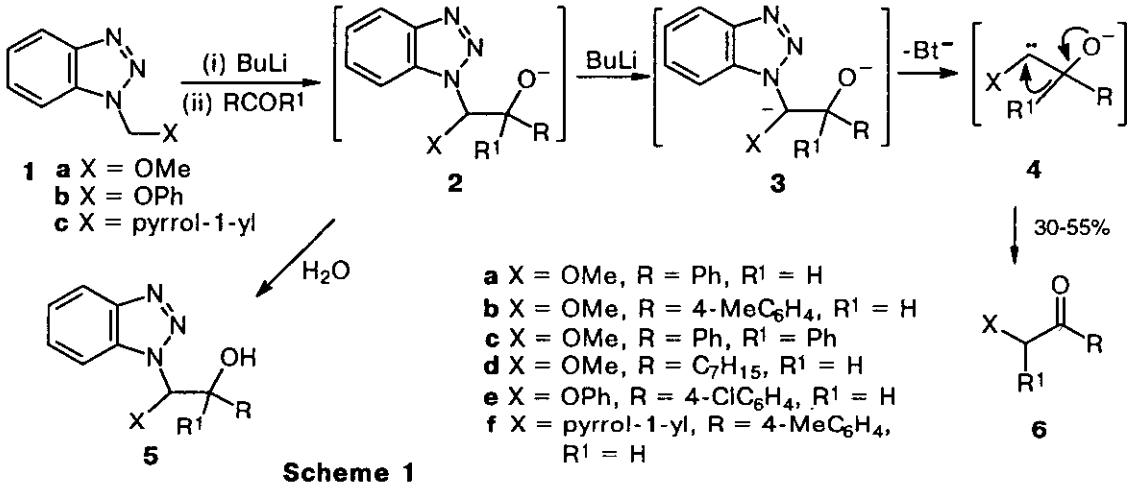
The present work was undertaken to determine whether the benzotriazole intermediates, which we earlier successfully rearranged by Lewis acid catalysis to afford the insertion products,[2] would undergo base-catalysed rearrangement. We now report that in the cases of **1a-c**, the expected carbenoid-type insertions do indeed occur to produce the α-alkoxyalkyl, α-(pyrrol-1-yl)alkyl ketones (**6**). In these reactions, as in all the earlier work,[2] the benzotriazole group is removed in the final products as a leaving group. However, in an interesting variation the intermediates (**3d-f**), upon further treatment with BuLi, undergo benzotriazole *ring-opening and rearrangement* to give α-aryl-α-(phenylamino)alkyl (**7a,b**) and α-(heteroaryl)-α-(phenylamino)alkyl ketones (**7c-f**); compounds of type (**7c-f**) were hitherto unknown.

RESULTS AND DISCUSSION

1-(Alkoxymethyl)benzotriazoles (**1a,b**)[7-9] and *N*-(benzotriazol-1-ylmethyl)pyrrole (**1c**)[10] were prepared according to our previously reported procedures. Treatment of compound (**1a**) with one equivalent of BuLi at -78 °C in THF for a few minutes followed by reaction with benzaldehyde and subsequent treatment with a second equivalent of BuLi at -78 °C rising to 20 °C over 10 h gave 2-methoxyacetophenone (**6a**) in 50% isolated yield. The structure of **6a** was characterized by ^{1}H and ^{13}C NMR spectroscopy, CHN analysis and compared with known data. Compounds (**6b-f**) were similarly prepared in 30-55% yields. The transformations are believed to proceed *via* the dianions (**3**) and carbenoid intermediates (**4**) as shown in Scheme 1. In our previous work,[11] deprotonated **1** on reaction with aldehydes followed by quenching of **2** with water at -78 °C gave the corresponding hydroxy derivatives (**5**). Further treatment of **2** with BuLi removes the methine proton forming the dianions (**3**), in which the benzotriazole group under the influence of the alkoxide anion behaves as a leaving group losing the benzotriazole anion, leading to carbenoids (**4**). Subsequent rearrangement produces the ketones (**6**). In comparison with our previously reported Lewis acid induced rearrangement,[2] the present approach gives low to medium yields, probably due to complex side reactions caused by the reactive dianions and the carbene intermediates in the system. The low yield for **6c** can be explained by the relatively low migration ability of the aryl group compared with hydrogen. This is consistent with the previous observation.[2]

Synthesis of α-alkoxy- and α-phenoxyketones of type (**6a-e**) was previously accomplished by (i) reaction of α-diazoketones[12,13] or α-haloketones[14,15] with an alcohol or phenol,[16] (ii) α-alkoxylation of ketones with hypervalent iodine reagent[17] or lead tetraacetate,[18] (iii) palladium-catalyzed coupling of acyl chlorides

with organotins of type $MeOCH_2SnR_3$[19] or with organomanganous reagents,[20] and (iv) reaction of an aldehyde with triethylphosphonium reagents (prepared from a selenoester and triethylphosphine).[21] Approaches closely related to the present method are the Lewis acid mediated rearrangement of β-hydroxy-α-methoxysulfones[6] and our recently reported $ZnCl_2$-promoted benzotriazole-mediated one-carbon homologations.[2] Preparations of aroylmethylpyrroles of type (**6f**) have rarely been reported. Artico *et al.*[22] synthesized aroylmethylpyrrole compounds in varying yields (24-78%) by reaction of a phenacylamine hydrochloride with 2,5-dimethoxytetrahydrofuran or 2,5-dioxohexane in refluxing DMF. We have previously prepared *N*-(4-methylbenzoylmethyl)pyrrole by reduction of *N*-[(4-methylbenzoyl)(benzotriazol-1-yl)methyl]pyrrole[10] or by treatment of *N*-[2-(4-methylphenyl)-2-hydroxy-1-(benzotriazol-1-yl)ethyl]pyrrole with phenylmagnesium bromide.[23]

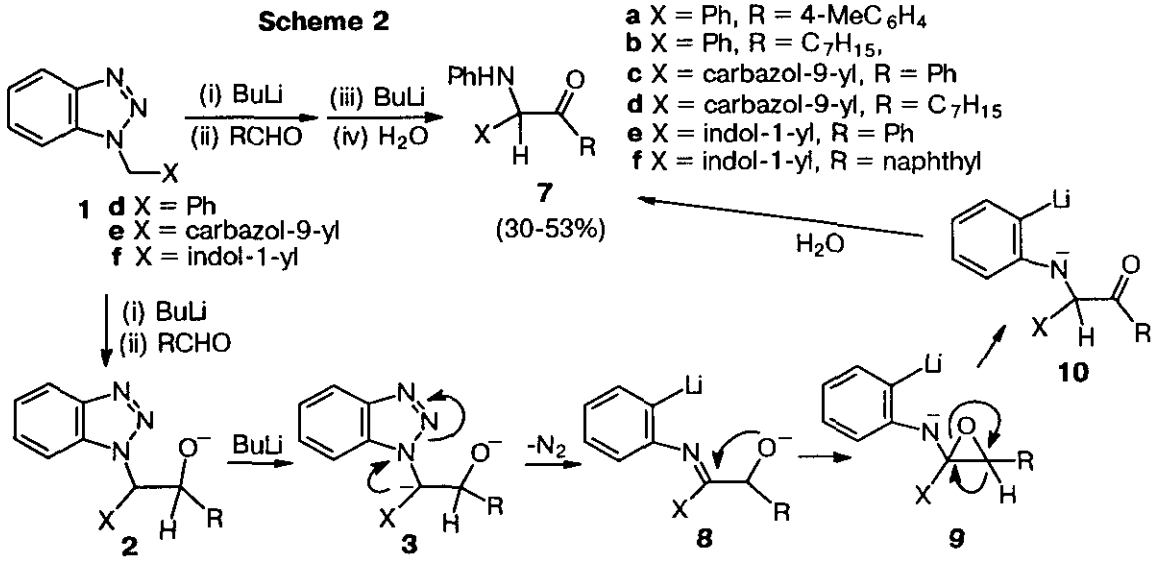


Scheme 1

When 1-benzylbenzotriazole (**1d**), 9-(benzotriazol-1-ylmethyl)carbazole (**1e**)[10] and 1-(benzotriazol-1-ylmethyl)indole (**1f**)[10] were used as the insertion reagents to carry out similar reactions, benzotriazole ring-opening and rearranged products (**7a-f**) were produced in 30-53% yields. Thus, treatment of compound (**1d**) with BuLi at -78 °C in THF for a few minutes followed by reaction with 4-methylbenzaldehyde and subsequent treatment with a second equivalent of BuLi at -78 °C rising to 20 °C over 10 h gave 1-(4-methylphenyl)-2-phenyl-2-phenylaminoethanone (**7a**) in 40% isolated yield. The structure of **7a** was elucidated by ^1H and ^{13}C NMR spectroscopy and by CHN analysis. The ^1H NMR spectrum clearly showed the presence of 19 protons and a heteroatom substituted methine proton. ^{13}C NMR also indicated the presence of a carbonyl signal at 198 ppm and the disappearance of the benzotriazole group.

Compounds (**7b-f**) were similarly prepared in 30-53% yields by this procedure and their structures were fully characterized spectroscopically and by elemental analysis.

Scheme 2



The formation of the ketones (**7a-f**) can be rationalized by intermediates (**3, 8, 9** and **10**) as shown in Scheme 2. Unlike the previous cases of **1a-c** (where X = MeO, PhO and pyrrolyl), further treatment of **2** with BuLi removes the methine proton forming the dianions (**3**), which undergo benzotriazole ring-scission and nitrogen loss to *o*-lithiated aniline imines (**8**). Subsequent intramolecular attack and the migration of hydrogen *via* intermediates (**9** and **10**) afford the final α-phenylamino-α-heteroaryl (aryl)-substituted ketones (**7**). Although the benzotriazole ring is stable to most acidic and basic conditions, we previously observed similar benzotriazole ring opening in several benzotriazol-1-yl alkyl carbanions[24] where the benzotriazole does not act in its usual role as a leaving group. α-Aryl-α-(phenylamino)alkyl ketones of type (**7a,b**) have previously been prepared by (i) reaction of α-chloro ketones[25] with aniline, (ii) condensation of 1,2-dicarbonyl compounds with aniline followed by reduction,[26] (iii) the imino-Wittig rearrangement,[27] and (iv) reaction of alkanoyl anions with an imine.[28,29] Compounds of type (**7c-f**) have not been previously reported.

In conclusion, novel base-catalyzed rearrangement of α-benzotriazolylalkoxide anions for the synthesis of one-carbon homologated α-substituted alkyl ketones has been described. The present approach employs mild reaction conditions and simple operation. Preparation of **6a-f** is of mechanistic interest and can be synthetically useful for the acid-sensitive systems. Formation of **7a-f** is of both mechanistic and synthetic interest.

EXPERIMENTAL

General Comments. Melting points were determined on a hot stage apparatus without correction. ^1H (300 MHz) and ^{13}C NMR (75 MHz) spectra were recorded on a Gemini-300 spectrometer in $CDCl_3$ with TMS or $CDCl_3$, respectively, as the internal reference. Elemental analyses were performed using a Carlo Erba 1106 elemental analyzer. High resolution mass spectra were measured on an AEI-30 mass spectrometer. Column chromatography was carried out on MCB silica gel (230-400 mesh). Tetrahydrofuran (THF) was freshly distilled from sodium-benzophenone. Lithiation reactions were carried out under the protection of dry nitrogen.

General Procedure for the Synthesis of Compounds (6a-f) and (7a-f). To a solution of benzotriazole derivative **1a-f** (10 mmol) in THF (80 mL) at -78 °C was added BuLi (2 *M*, 5 mL, 10 mmol). The solution was stirred for 15 min and the appropriate aldehyde or ketone (10 mmol) was added. After 30 min, one more equivalent of BuLi (10 mmol) was added at this temperature. The solution was allowed to warm to rt gradually and was stirred for 10 h . Water (50 mL) was added and the mixture was extracted with ether (2 x 100 mL), dried with $MgSO_4$. Evaporation of the solvents gave a residue, which was chromatographed on silica gel to give the final product.

2-Methoxy-1-phenylethanone (6a): Obtained as a yellowish oil; yield 50% (Lit.,[12] bp 124-126 °C/19 mmHg); ^1H NMR δ 3.51 (s, 3 H), 4.71 (s, 2 H), 7.47 (dt, 2 H, J = 7.2, 1.1 Hz), 7.58 (t, 1 H, J = 7.7 Hz), 7.93 (d, 2 H, J = 8.4 Hz); ^{13}C NMR δ 59.0, 74.9, 127.4, 128.4, 133.2, 134.5, 195.8. Anal. Calcd for $C_9H_{10}O_2$: C, 71.97; H, 6.72. Found: C, 72.13; H, 7.00.

2-Methoxy-1-(4-methylphenyl)ethanone (6b): Obtained as a yellow oil; yield 52% (Lit.,[30] bp 83-88 °C/ 0.15 mmHg); ^1H NMR δ 2.41 (s, 3 H), 3.50 (s, 3 H), 4.69 (s, 2 H), 7.27 (d, 2 H, J = 8.0 Hz), 7.83 (dd, 2 H, J = 8.2 and 1.3 Hz); ^{13}C NMR δ 21.4, 59.0, 74.9, 127.6, 129.1, 132.1, 144.1, 195.4.

2-Methoxy-2-phenylacetophenone (6c): Obtained as a yellow oil; yield 35% (Lit.,[26] oil); ^1H NMR δ 3.44 (s, 3 H), 5.51 (s, 1 H), 7.27-7.39 (m, 5 H), 7.45-7.48 (m, 3 H), 7.98 (d, 2 H, J = 7.2 Hz); ^{13}C NMR δ 57.3, 86.5, 127.5, 128.4, 128.5, 128.7, 133.1, 134.9, 136.0, 197.0.

1-Methoxy-2-nonanone (6d): Obtained as a colorless oil; yield 30% (Lit.,[31]); ^1H NMR δ 0.81 (t, 3 H, J = 7.02 Hz), 1.15-1.26 (m, 8 H), 1.48-1.58 (m, 2 H), 2.36 (t, 2 H, J =7.3 Hz), 3.35 (s, 3 H), 3.94 (s, 2 H); ^{13}C NMR δ 14.0, 22.5, 23.3, 28.9, 29.1, 31.6, 38.8, 59.2, 77.6, 208.7.

2-Phenoxy-1-(4-chlorophenyl)ethanone (6e): yellowish solid, yield 36%; mp 81-82 °C (hexane/ethyl acetate = 50:1 as the eluent); ^1H NMR δ 5.19 (s, 2 H), 6.92 (d, 2 H, J = 8.6 Hz), 6.98 (t, 1 H, J = 7.4 Hz), 7.28 (t, 2 H, J = 7.5 Hz), 7.45 (d, 2 H, J = 8.7 Hz), 7.94 (d, 2 H, J = 8.6 Hz); ^{13}C NMR δ 70.9, 114.7,

121.7, 129.1, 129.6, 140.3, 157.8, 193.7. Anal. Calcd for $C_{14}H_{11}O_2Cl$: C, 68.28; H, 4.51. Found: C, 68.21; H, 4.51.

***N*-(4-Methylbenzoylmethyl)pyrrole (6f):** yellowish solid, yield 32%; mp 145-146 °C (lit.,[10] mp 144-146 °C) (hexane/ethyl acetate = 40:1 as the eluent); ^{1}H NMR δ 2.41 (s, 3 H), 5.24 (s, 2 H), 6.23 (t, 2 H, J = 2.1 Hz), 6.64 (t, 2 H, J = 2.1 Hz), 7.27 (d, 2 H, J = 8.0 Hz), 7.83 (d, 2 H, J = 8.2 Hz); ^{13}C NMR δ 21.6, 55.1, 108.8, 121.8, 128.0, 129.5, 132.1, 144.8, 192.9. Anal. Calcd for $C_{13}H_{13}NO$: C, 78.35; H, 6.58; N, 7.03. Found: C, 78.31; H, 6.64; N, 6.77.

1-(4-Methylphenyl)-2-phenyl-2-phenylaminoethanone (7a): colorless solid, yield 40%; mp 137-138 °C (hexane/ethyl acetate = 8:1 as the eluent); ^{1}H NMR δ 2.34 (s, 3 H), 5.42 (d, 1 H, J = 6.6 Hz), 6.00 (d, 1 H, J = 6.6 Hz), 6.64-6.68 (m, 3 H), 7.08-7.28 (m, 7 H), 7.44 (d, 2 H, J = 7.2 Hz), 7.90 (d, 2 H, J = 8.2 Hz); ^{13}C NMR δ 21.6, 62.5, 113.5, 117.7, 127.9, 128.0, 128.9, 129.2, 129.3, 132.4, 138.0, 144.4, 146.2, 196.5. Anal. Calcd for $C_{21}H_{19}NO$: C, 83.68; H, 6.36; N, 4.65. Found: C, 83.72; H, 6.49; N, 4.59.

1-Phenyl-1-phenylamino-2-nonanone (7b): yellowish solid, yield 30%; mp 71-72 °C (hexane/ethyl acetate = 10:1 as the eluent); ^{1}H NMR δ 0.84 (t, 3 H, J = 7.1 Hz), 1.09-1.26 (m, 8 H), 1.41-1.52 (m, 2 H), 2.38-2.44 (m, 2 H), 4.98 (s, 1 H), 5.47 (br s, 1 H), 6.52-6.56 (m, 2 H), 6.61-6.66 (m, 1 H), 7.04-7.11 (m, 2 H), 7.23-7.38 (m, 3 H), 7.41-7.45 (m, 2 H); ^{13}C NMR δ 14.0, 22.5, 23.8, 28.8, 31.5, 39.1, 67.6, 113.3, 117.5, 127.8, 128.2, 129.1, 138.1, 146.0, 206.4. Anal. Calcd for $C_{21}H_{27}NO$: C, 81.50; H, 8.80; N, 4.53. Found: C, 81.68; H, 9.02; N, 4.48.

1-Phenyl-2-(carbazol-9-yl)-2-phenylaminoethanone (7c): yellow solid, yield 53%; mp 175-176 °C (hexane/ethyl acetate = 6:1 as the eluent); ^{1}H NMR δ 6.43 (d, 1 H, J = 5.2 Hz), 7.01 (t, 1 H, J = 7.3 Hz), 7.08 (d, 2 H, J = 7.7 Hz), 7.39 (t, 2 H, J = 7.4 Hz), 7.40-7.56 (m, 5 H), 7.64 (t, 1 H, J = 7.4 Hz), 7.81 (t, 2 H, J = 7.2 Hz), 8.04 (d, 2 H, J = 8.3 Hz), 8.18 (d, 2 H, J = 7.3 Hz), 8.28 (d, 2 H, J = 7.8 Hz); ^{13}C NMR δ 66.4, 110.0, 114.4, 119.1, 119.9, 120.5, 124.0, 126.1, 128.2, 128.5, 129.3, 133.9, 138.8, 144.7, 193.2. Anal. Calcd for $C_{26}H_{20}N_2O$: C, 82.94; H, 5.36; N, 7.45. Found: C, 82.82; H, 5.38; N, 7.37.

1-(Carbazol-9-yl)-1-phenylaminononanone (7d): colorless solid, yield 40%; mp 89-90 °C (hexane/ethyl acetate = 6:1 as the eluent); ^{1}H NMR δ 0.77 (t, 3 H, J = 7.0 Hz), 0.85-1.18 (m, 8 H), 1.30-1.48 (m, 2 H), 1.90-2.02 (m, 1 H), 2.12-2.25 (m, 1 H), 5.81 (d, 1 H, J = 3.4 Hz), 6.29 (d, 1 H, J = 3.4 Hz), 6.62-6.69 (m, 3 H), 7.03 (t, 2 H, J = 7.4 Hz), 7.26 (t, 2 H, J = 7.5 Hz), 7.45 (t, 2 H, J = 7.2 Hz), 7.58 (d, 2 H, J = 8.2 Hz), 8.08 (d, 2 H, J = 7.5 Hz); ^{13}C NMR δ 13.9, 22.4, 23.4, 28.6, 31.3, 37.7, 69.9, 109.7, 113.9, 119.0, 120.2, 120.6, 124.0, 126.2, 129.3, 139.2, 144.9, 203.7. Anal. Calcd for $C_{27}H_{30}N_2O$: C, 81.36; H, 7.59; N, 7.03. Found: C, 81.70; H, 7.51; N, 7.03.

1-Phenyl-2-(indol-1-yl)-2-phenylaminoethanone (7e): yellowish solid, yield 46%; mp 118-119 °C (hexane/ethyl acetate = 6:1 as the eluent); ^{1}H NMR δ 5.78 (d, 1 H, J = 6.6 Hz), 6.48 (d, 1 H, J = 3.4 Hz), 6.71-6.77 (m, 3 H), 6.98 (d, 1 H, J = 6.8 Hz), 7.05-7.18 (m, 3 H), 7.21-7.33 (m, 4 H), 7.40-7.48 (m, 1 H), 7.52 (d, 1 H, J = 7.9 Hz), 7.59 (d, 1 H, J = 8.3 Hz), 7.91 (d, 2 H, J = 7.2 Hz); ^{13}C NMR δ 66.6, 104.0, 109.7, 114.2, 119.4, 120.2, 121.4, 122.3, 125.3, 128.6, 128.8, 129.4, 133.7, 134.1, 135.0, 144.3, 192.0. Anal. Calcd for $C_{22}H_{18}N_2O$: C, 80.95; H, 5.56; N, 8.59. Found: C, 81.31; H, 5.61; N, 8.58.

1-Naphthyl-2-(indol-1-yl)-2-phenylaminoethanone (7f): yellowish solid, yield 31%; mp 110-112 °C (hexane/ethyl acetate = 6:1 as the eluent); ^{1}H NMR δ 5.99 (d, 1 H, J = 5.7 Hz), 6.45 (d, 1 H, J = 3.3 Hz), 6.77-6.83 (m, 3 H), 7.03-7.08 (m, 2 H), 7.14-7.27 (m, 3 H), 7.31-7.39 (m, 2 H), 7.48-7.55 (m, 4 H), 7.74 (d, 1 H, J = 7.2 Hz), 7.79-7.82 (m, 1 H), 7.90 (d, 1 H, J = 8.4 Hz), 8.24 (d, 1 H, J = 8.4 Hz); ^{13}C NMR δ 68.6, 104.0, 109.6, 114.0, 119.4, 120.1, 121.3, 122.2, 124.0, 124.8, 124.9, 126.7, 126.9, 128.1, 128.5, 129.5, 133.4, 133.7, 144.3, 195.4. Anal. Calcd for $C_{26}H_{20}N_2O$: C, 82.94; H, 5.36; N, 7.45. Found: C, 82.96; H, 5.37; N, 7.44.

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dedicated in honor of professor Koji Nakanishi's 75th birthday.

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