

Note

Chemoselective reduction of α , β -unsaturated carbonyl compounds by sodium hydrogen telluride: Part I

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Sodium Hydrogen Telluride is used for the selective reduction of C=C bonds in various chalcones. The reduction proceeds smoothly in good yields.

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Sodium hydrogen telluride has earlier been shown by Shanmugam and co-workers of our department to serve as a convenient reagent for the hydrogenation of carbon-carbon double bonds^{1,2}, for the debromination of vicinal dibromides³, for the deacylation of aryl acetates and benzoates⁴, for deblocking allyl carboxylates and allyl phenyl ethers⁵ and for the partial deallylation of 3,3-diallyl-1,2,3,4-tetrahydroquinolin-2,4-diones⁶. In continuation of our work on sodium hydrogen telluride, we wish to report herein on the further utility of sodium hydrogen telluride as a mild and effective reagent for the selective reduction of C=C double bond in chalcones. We have now reduced a few α , β -unsaturated carbonyl compounds **1a** - **8a** which also include the newer chalcones viz. **5a** - **8a** (Table I). All these compounds have been prepared in our laboratory for the further synthesis of heterocycles. The compounds have been reduced by NaHTe prepared *in situ*⁷ from tellurium powder and sodium borohydride in ethanol on refluxing for 4-5 hr under nitrogen cover. The reduction proceeded smoothly in each case with the product yields varying from 71-95% and without the formation of any side products. This method has an advantage that it is operationally smooth, the conditions are mild and tellurium metal can be recovered almost quantitatively and reused.

Experimental Section

Melting points were determined on a Boetius micro-

heating table/in open capillaries and are uncorrected. IR spectra (KBr) were recorded on a Perkin-Elmer 597 infrared spectrophotometer and NMR spectra on a Hitachi R-600 spectrometer/AMX-400 NMR spectrometer using TMS as an internal reference. Elemental analyses were performed by Cairo-Elmer 1106 and Perkin-Elmer model 1240 CHN analyser.

Compounds **1a** and **2a** were obtained by reacting benzaldehyde with acetophenone⁸ and acetone⁹ respectively and **3a** by reacting furfural with acetone¹⁰ by following standard procedures. Coumarin **4a** was prepared from salicylaldehyde, sodium acetate and acetic anhydride¹¹.

Preparation of 5a - 8a. Acetophenone / acetone (3 mmole) was taken in a 50 mL round bottomed flask and 30 mL of methanol was added and stirred. To this 2 mL of 10% sodium hydroxide was added dropwise for a period of 10 min and the solution was kept stirring for 20 min at room temperature. To this 3-formyl-2-quinolone¹² (3 mmole) was added and the reaction mixture was stirred for 4 hr. The mixture was poured into ice water. The solid obtained was filtered, washed with water, dried and recrystallised from methanol.

5a: Yield 50%, m.p. 224°C; IR(KBr): 3300 (-NH), 1660 (-CO-CH=CH-), 1640 (-CONH-)cm⁻¹; ¹H NMR (CDCl₃): δ 7.5-8.31 (m, 12H, ethylenic, phenyl and aromatic protons), 10.21 (s, 1H, NH); Anal. Found: C, 78.39; H, 4.84; N, 5.01. Calcd for C₁₈H₁₃NO₂: C, 78.53; H, 4.76; N, 5.09%.

6a: Yield 80%, m.p. 221°C; IR(KBr): 3400 (NH), 1680 (-CO-CH=CH-), 1648 (CONH)cm⁻¹; Anal. Found: C, 78.43; H, 5.31; N, 4.79. Calcd for C₁₉H₁₅NO₂: C, 78.87; H, 5.23; N, 4.84%.

7a: Yield 66%, m.p. 226°C; IR(KBr): 3315 (NH), 1662 (-CO-CH=CH-), 1645 (CONH)cm⁻¹; Anal. Found: C, 78.74; H, 5.25; N, 4.78. Calcd for C₁₉H₁₅NO₂: C, 78.87; H, 5.23; N, 4.84%.

8a: Yield 74%, m.p. 264°C; IR(KBr): 3320 (NH), 1660 (-CO-CH=CH-), 1645 (CO-NH)cm⁻¹; Anal. Found: C, 73.88; H, 5.59; N, 6.05. Calcd for C₁₄H₁₃NO₂: C, 73.99; H, 5.77; N, 6.16%.

General procedure for the reduction 1a - 8a using NaHTe. To a solution of NaHTe prepared *in situ*⁷ from tellurium powder 1.30 g (0.01 mole) and sodium borohydride 0.9 g (0.03 mole) in ethanol (50 mL) was added the substrate a (0.01 mole). The mixture was

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Table I—Reduction using sodium hydrogen telluride

Sl. No.	Substrate a	Product ^c b	Reaction time (hr)	Yield (%)	m.p. or b.p./mm (°C)	Lit.m.p. or b.p./mm Mol. formula ^d
1			5	95	72	72-73 ¹³
2			4	85	116/13 mm	115/13 mm ¹⁴
3			5	85	102-104/23mm	100-104/23 mm ¹⁵
4			5	85	145-150/15mm	145/13mm ¹⁶
5			4	78	268	C ₁₈ H ₁₅ NO ₂
6			4	80	264	C ₁₉ H ₁₇ NO ₂
7			4	71	271	C ₁₉ H ₁₇ NO ₂
8			4	75	153	C ₁₄ H ₁₅ NO ₂

IR spectra (KBr) of **c. 5b - 8b** showed ν_{\max} at 1695 - 1720 cm^{-1} (-CO-CH₂-CH₂-); ¹H NMR spectra (CDCl₃): **1a** δ 2.9 - 3.4 (4H, coupled triplets), 7.4 - 7.7 (5H, m, ArH), 7.9 - 8.2 (5H, m, ArH); **5a** δ 3.31 (t, 2H, C1-H), 3.94 (t, 2H, C2-H), 7.22 - 8.55 (m, 10H aromatic, phenyl protons), 10.51 (s, 1H, NH)

Satisfactory analytical data were obtained for all compounds.

refluxed for 4-5 hr under nitrogen, cooled, filtered, poured into ice cooled water/crushed ice. For **1a - 4a**, product was extracted with chloroform. The organic layer was dried over anhyd. Na₂SO₄, evaporated and the residue was either purified by recrystallisation from ethanol or by distillation under reduced pressure. For **5a - 8a**, the product obtained as solid was filtered and recrystallised from ethanol.

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