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Biomedical Applications And Oxidative Aromatization Of Hantzsch 1,4-Dihydropyridines: A Review

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Introduction

The study of dihydropyridines began early in 1882, when Hantzsch disclosed the first synthesis of these compounds. Major landmarks were the isolation of NADH (reduced nicotinamide adenine dinucleotide, **Fig. 1**) and its role as a reductive cofactor, and the breakthrough of Hantzsch dihydropyridines as antihypertensive drugs. Afterwards, research also focused on NADH mimics and on the synthetic aspects of these heterocyclic systems, especially with regard to natural products and bioactive agents.[1,2]

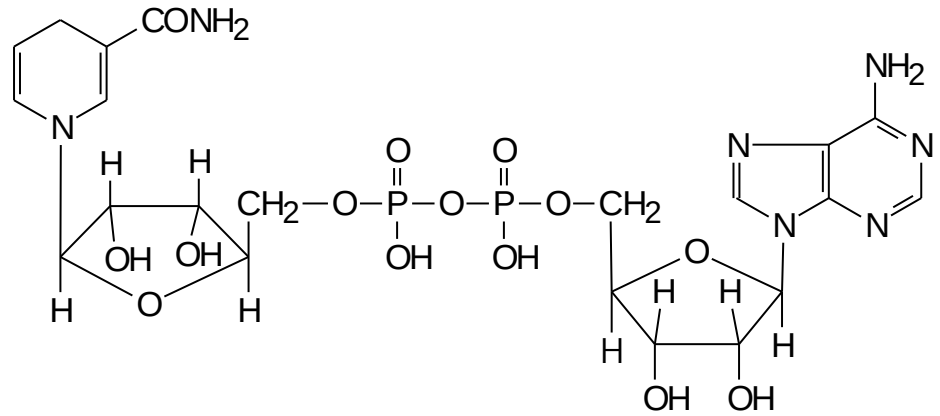

Fig. 1

The most interesting aspect of dihydropyridines can be attributed to the coenzyme NADH and the unique ability of these compounds in biological systems to reduce unsaturated functionalities and also strained ring systems (carbonyls, conjugated olefins, epoxides, etc.).[3] The mechanism of reactions mediated by NAD(P)H and its models has been extensively studied and continues to attract considerable attention because of the need to understand the finer details of the mechanism.[4,5] Concerning the reaction mechanism, one critical yet controversial issue is whether the formal hydride transfer from NAD(P)H and its model compounds to the substrates occurs by one-step direct detachment or by a multi-step sequential e^-–H^-–e^- process. Evidence has been reported in support of the e^-–H^-–e^- transfer mechanism for many thermal reactions in which strong oxidants are involved.[6] Evidence for the direct hydride transfer mechanism has also been reported in reactions with

aldehydes and ketones, particularly in the cases of most $NAD(P)^+$ / NAD(P)H mediated enzymatic reactions.[7]

Biomedical Applications

1,4-Dihydropyridines (DHPs) are an important class of drugs which are potent blockers of calcium (Ca^{2+}) currents through voltage-dependent L class Ca^{2+} channels.[8] They induce relaxation of vascular smooth muscle, preferentially in arterial beds, and display a negative inotropic effect on isolated cardiac muscle. In therapy, this class of drugs is principally used in the treatment of cardiac arrhythmias, peripheral vascular disorders, and hypertension.[9] DHPs have received most attention as calcium channel blockers, as exemplified by commercial therapeutic agents such as Nifedipine,[10] Nitrendipine[11] and Nimodipine.[12] Second-generation calcium antagonists include DHPs with improved bioavailability, tissue selectivity/stability, such as antihypertensive/antianginal drugs Elgodipine,[13] Furnidipine,[14] Darodipine,[15] Pranidipine,[16] Lemildipine,[17] Dexniguldipine,[18] Lacidipine[19] and Benidipine.[20] Following the discovery of compound Bay K 8644,[21] a number of DHPs acting as calcium agonists rather than antagonists were introduced as potential drug candidates for the treatment of congestive heart failure.[22] DHPs have also exhibited neuroprotectant and cognition enhancer lacking neuronalspecific calcium antagonist properties.[23] While the majority of DHP therapeutic agents have been originally developed as cardiovascular and antihypertensive drugs, recent studies suggest several other medicinal applications. Thus, preclinical data have demonstrated the potential of DHPs in the treatment of Alzheimer's disease,[24] diabetic nephropathy[25] and a series of platelet anti-aggregatory DHPs[26] including the drug Trombodipine.[27] The antiproliferative effect of Ca^{2+} channel blockers has also been shown in tumor cell lines.[28-32] The potential of dihydropyridines as antifungal agents has also been investigated.[33] Some 4-substituted phenyl-2,6-dimethyl-3,5-bis-N-(substituted phenyl)carbamoyl-1,4-dihydropyridines were tested as potential antitubercular agents.[34,35] The NO-dependent vasodilator activities of 4-phenyl-1,4-dihydropyridine substituted compounds were evaluated on rat aorta.[36]

Synthetic derivatives of 1,4-dihydropyridine (1,4-DHPs) show modulating activity on corticosteroid regulatory circuits,[37] and prevent inflammatory[38] and diabetic processes[39,40] and some show antimutagenic,[41,42] antineoplastic,[43] geroprotective,[44] radioprotective,[45] and radiosensitizing effects.[46] DHP derivatives also act as a fertile source of valuable drug for cancer[47] and AIDS.[48]

Particularly, for optimal activity of 1,4-DHPs as antagonists, the following essential moieties have been detected: (a) the presence of the intact 1,4-DHP ring; (b) the secondary nitrogen in the heterocycle; (c) a space-filling substituent in para-position of the 1,4-DHP ring; (d) the presence of ester groups in 3- and 5-position and the methyl groups in 2- and 6-position on the 1,4-DHP ring.[49]

1,4-Dihydropyridine (DHP) derivatives possess pronounced free radical quenching and antioxidant properties[50] that may contribute to their pharmacological activity. This effect is not due to the Ca^{2+} antagonist effect, but it is related to the reactivity of these compounds toward radical species.[51]

In the human body, these compounds are oxidized to pyridine derivatives by the action of cytochrome P-450 enzymes in the liver.[52] These oxidized compounds are largely devoid of the pharmacological activity of the parent compounds. The oxidation of 1,4-DHPs to the corresponding pyridine derivatives constitutes the principal metabolic route in biological systems,[53] as well as a facile access to the corresponding pyridine derivatives, which show antihypoxic and antiischemic activities.[54]

Aromatization Reactions

Consequently, this aromatization reaction continues to attract the attention of organic and medicinal chemists for the discovery of a plethora of protocols applicable to a wide range of DHPs. Many of the reported procedures involve the use of N_2O_3,[55] ceric ammonium nitrate,[56] homogeneous complex of palladium as a catalyst,[57] clay-supported cupric nitrate accompanied by ultrasound promotion,[58] chromic acid,[59] elemental sulfur,[60] potassium permanganate,[61] chloranil,[62] p-nitrosodimethylaniline,[63] hydrogen peroxide,[64] diisoamyl disulfide,[65] silver nitrate,[66] platinum in acetic acid,[67] mercuric acetate,[68] iodine,[69] iron or nickel carbonyls,[70] manganese dioxide or dichlorodicyanoquinone (DDQ),[71] nitric oxide,[72] bismuth nitrate pentahydrate,[73] PCC,[74] tetrakis-(pyridine) cobalt(II) dichromate,[75] nicotinium dichromate,[76] S-nitrosoglutathione,[77] N_2O_4 complex of 18-crown-6,[78] 3-carboxypyridinium chlorochromate (CPCC),[79] MnO_2/bentonite/microwave irradiation,[80] HNO_3,[81-84] HNO_2,[85] tert-butylhydroperoxide,[86] CrO_3,[87] photo-oxidation,[88] tetraethylammonium bromate,[89] $CO(NH_2)_2.H_2O_2/I_2$,[90] $K_2S_2O_8$,[91] clayfen,[92] diphenylpicrylhydrazyl/benzoylperoxide,[93] $NaHSO_4/Na_2Cr_2O_7$/wet SiO_2,[94] microwave under solid phase condition,[95] iodine/ultrasound irradiation,[96] inorganic acidic salts/sodium nitrite or nitrate,[97] silica chloride and sodium nitrite in the presence of wet SiO_2,[98] polystyrene-bound $Mn(TPP)Cl/NaIO_4$,[99] $Mn(TPP)Cl/(n\text{-}Bu)_4NIO_4$,[100] 4-phenyl-1,2,4-triazole-3,5-dione,[101] $Zr(NO_3)_4$,[102] $H_2O_2/Co(OAc)_2$,[103] urea nitrate and peroxydisulfate-Co(II),[104] hypervalent iodine reagents,[105] I_2-MeOH,[106] selenium dioxide,[107] heteropolyacid/$NaNO_2$/wet SiO_2,[108] cytochrome P-450,[109] electrochemical catalysis,[110] manganese triacetate,[111] N-hydroxyphthalimide/O_2/$Co(OAc)_2$,[112] $Fe(ClO_4)_3$/HOAc,[113] Mn(III)-salophen/$NaIO_4$,[114] Pd/C in acetic acid,[115] vanadomolybdophosphate heteropolyacid ($H_6PMo_9V_3O_{40}$),[116] benzyltriphenylphosphonium peroxymonosulfate,[117] 9-phenyl-10-methylacridinium/molecular oxygen,[118] bismuth(III) chloride supported on to wet HZSM-5 zeolite,[119] barium manganate,[120] electrogenerated superoxide,[121] iodobenzene diacetate,[122] Co(II) naphthenate,[123] solid acids,[124] sodium nitrite in the presence of a catalytic amount of acidic silica gel,[125] 2,6-dicarboxypyridinum chlorochromate (2,6-DCPCC),[126] aqueous hydrogen peroxide–acetic acid,[127] DMSO/O_2,[128] $KBrO_3/SnCl_4.5H_2O$,[129] potassium ferrate(VI) supported on montmorillonite K-10 under microwave irradiation,[130] activated carbon/molecular oxygen,[131] Dess-Martin periodinane under classical heating and microwave irradiation,[132] zeofen,[133] manganese dioxide supported onto HZSM-5 zeolite,[134] N, N'-ethylene-bis(benzoylacetoniminato) copper (II), ($Cu[C_{22}H_{22}N_2O_2]$),[135] N-nitroso-2-aryl-1,3-oxazolidines,[136] in situ generated acetyl hypoiodite or bromite,[137] tetraethylammonium superoxide[138,139] and silica chromate/$NaHSO_4.H_2O$/wet SiO_2.[140]

An elegant photocatalytic aromatization of 1,4-DHP by platinum(II) terpyridyl complexes has been reported leading to the generation of H_2.[141] Photoinduced oxidations of Hantzsch 1,4-dihydropyridines have been performed through SET processes with CCl_4,[142] and quinines.[143]

The electrooxidation of 1,4-DHP consists of removal of one electron from the starting 1,4-DHP molecule with formation of a cation radical, 1,4-$DHP^{+\cdot}$. This cation radical becomes deprotonated at a high rate forming a neutral radical, $Py^{\cdot}$, which oxidizes further to produce pyridinium cations, Py^+, and then deprotonated to form the pyridine derivative. The decay of the primary product, cation radical, 1,4-$DHP^{+\cdot}$, seems to be the key stage of the electrooxidation process and two ways of the decay of these cation radicals can occur, i.e. via loss of H^+ or via loss of $H^{\cdot}$. Competition between these two ways depends on the influence of temperature and on electronic effects of the substituents attached to the 1,4-DHP ring.[144-146] Electrochemical oxidation of 4-(pyridinium-3-yl)-1,4-dihydropyridines and 1,2-dihydropyridines (arising from [3+2] cycloadditions) affords the aromatized compounds, occasional cyclizations at the 4-substituent to yield indolizines have also been reported.[147]

Remarkable oxidations of enantiomerically pure Hantzsch-type dihydropyridines have allowed the enantioselective synthesis of atropoisomeric γ-arylpyridines, for instance $NOBF_4$ reacts with dihydropyridine to yield pyridine in 95% ee, whereas MnO_2 or $TEMPO^+BF_4^-$ affords the same product in 93% ee.[148]

In a conceptually different approach, oxidation of dihydropyridines can occur through bonding with electronegative atoms, bypassing the natural (biomimetic) electron transfer to form pyridinium salts. These, so-called nonbiomimetic oxidations constitute a family of chemically productive processes, yielding functionalized tetrahydropyridines or piperidines with potential use in organic synthesis. In this respect, 1,2- and N-acyl-1,4-dihydropyridines have been oxidized through interaction with MCPBA and OsO_4 to form the corresponding trans-hydroxyester and the tetraacetoxypiperidine respectively.[149] Similar reactions have been used in the synthesis of azasugars and alkaloid derivatives.[150-151]

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