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Review Article

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Review on Synthesis of *N*-glucosylamine Derivatives and Their Biological Activity

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ABSTRACT

The chemistry of glucosylamine derivatives has gotten among the synthetic compounds noticeable consideration as of late inferable from its significance in the pharmaceutical field. Glucosylamines carrying (R)-homoallylamines, benzoimidazole, phenanthrolineringshavebeenrepresente to display a wide range of pharmacological activities, which incorporates antibacterial, antimicrobial, antioxidant, anti-HIV and anticancer activity. These observations have been guiding for the synthesis of different derivatives of these glucosylamine derivatives enclosing biologically active nuclei and concentrate their pharmacological activities.

Keywords: Synthesis; *N*-Glycosylamines; Protected sugars; Monosaccharide

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INTRODUCTION

Glycosylamines are important intermediates in the synthesized of nucleosides and chemotherapeutic [1-3]. Carbohydrate derivatives use an productive stereo selective potential in various nucleophilic reactions on parochial imines, *β*-amino, and α-Amino acids and their derivatives can be preparation few synthetic steps with high enantiomer purity. A grouping of chiral heterocyclic can quickly be getting from glycosyl imines by stereo selective changes [4]. The asymmetric Staudinger reaction use 2,3,4,6-tetra-O-acetyl-β-D-galactopyranosylamine or 2,3,4,6-tetra-O-acetyl-β-D-glucopyranosylamine as the chiral auxiliary in the synthesis of 2-azetidinones has been told by the authors [5] and others [6]. 2-Azetidinonebase, determined as the central motif of the so-called β-lactam antibiotics, the most widely employed family of antimicrobial agents to date [7]. The importance of β-lactams as synthetic intermediates has been widely recognized in organic synthesis. *β*-Lactam molecules with a quaternary carbon center have been used as building blocks for biologically active compounds [8]. Heterocyclic compounds [9]

and their saccharide derivatives [10] display different applications (Figure 1A and 1B) and it is because of their structural features.

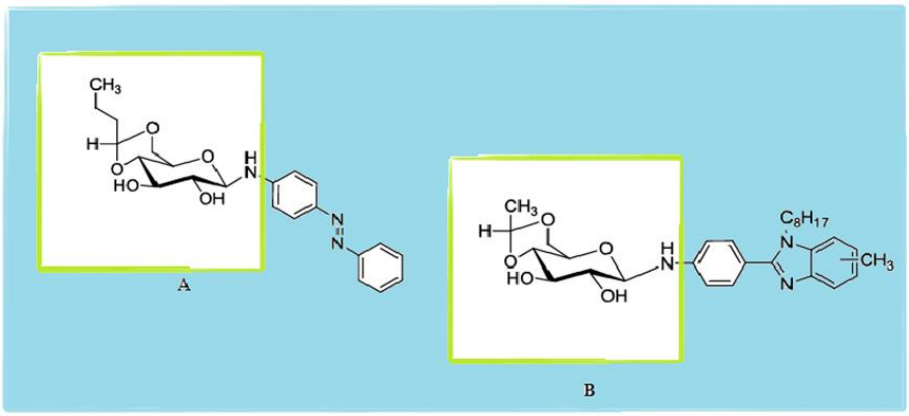


Figure 1. Examples of heterocyclic glucosylamine

LITERATURE REVIEW

Glucosamine Schiff bases [11] **2**, were synthesized by reaction of D-(+)-glucosamine hydrochloride (1) with *p*-methoxybenzaldehyde (2) or hydroxybenzaldehyde scheme 1 are the best Glucosamine Schiff bases synthesized from methyl benzaldehydes, 3-methoxybenzaldehyde,nitrobenzaldehydes and naphtylaldehydes. However, in this cases used another Schiff bases synthesized from aminoglucose. For this reason used 1,3,4,6-tetra-*O*-acetyl-β-D-glucosamine hydrochloridethat it's produced by three steps [12] (Figure 2).

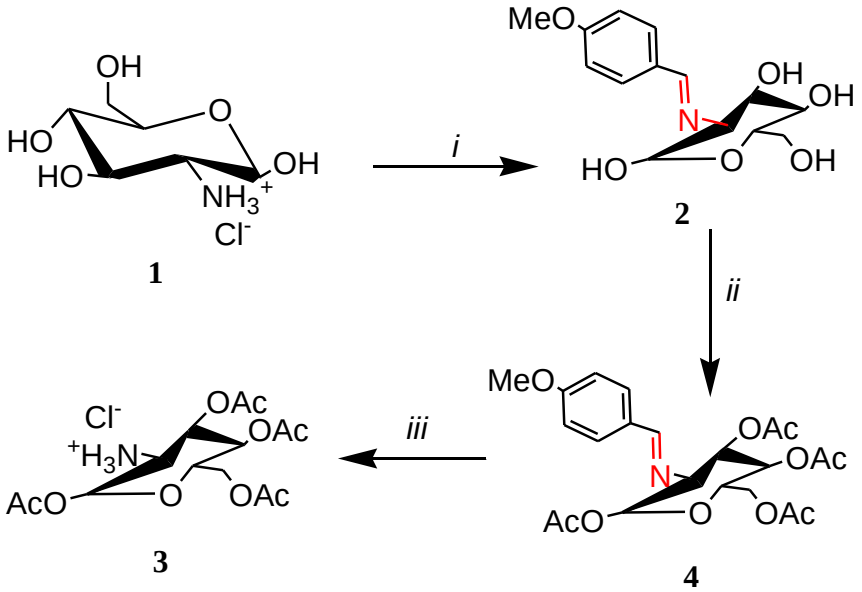


Figure 2. Synthesis of O-acylated glucosamine hydrochlorid [i: p-anisaldehyde, NaOH 1M; ii: Pyrridine, Ac2O; iii: acetone, HCl 5M]

Corresponding Schiff base molecules synthesized by the protection of its hydroxyl groups, followed by condensation with substituted different aldehydes **5a-k (**Figure 3). Furthermore; the practical has been application for conversion of D-Glucosamine into the corresponding Schiff base molecules through protection of its hydroxyl

groups, followed by condensation with substituted aldehydes. Handling, safety, light reaction conditions, good to excellent yields of the products and simple recovery of the reaction products make this method useful in organic synthesis [11].

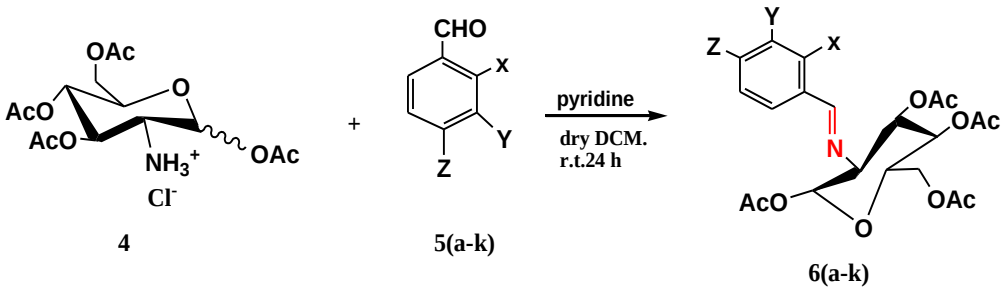


Figure 3. Synthesis of different Schiff bases from different aldehydes

5a: X: Methoxy, Y: H, Z: H

5b: X: Methyl, Y: H, Z: H

5c: X: Nitro, Y: H, Z: H

5d: X: Acetyl, Y: H, Z: H

5e: X: H, Y: Methoxy, Z: H

5f: X: H, Y: Methyl, Z: H

5g: X: H, Y: Nitro, Z: H

5h: X: H, Y: H, Z: Methoxy

5i: X: H, Y: H, Z: Methoxy

5j: X: H, Y: H, Z: Nitro

5k: 2-naphtyl

Reaction of phenyl isothiocyanated **7** reacted with guanidine carbonate to form thioguanidine derivatives **8** which it cycliztion to form 1,2,4-thiadiazolines **9** in presence of iodine as oxidizing agent. The N-(3-tetra-O-acetyl-β-D-glucopyranosyl)-1H(1,2,4-thiadiazolo (3,4-C) (1,2,4) thiadiazol-5(3H)ylidene) arylamines [13**] 11** were synthesized by following the interaction of 3 amino-5 arylimino-1,2,4 thiadiazolines**9** and N-tetra-O-acetyl β-D-glucopyranosylimino chloromethane sulfonyl chloride **10** (Figure 4).

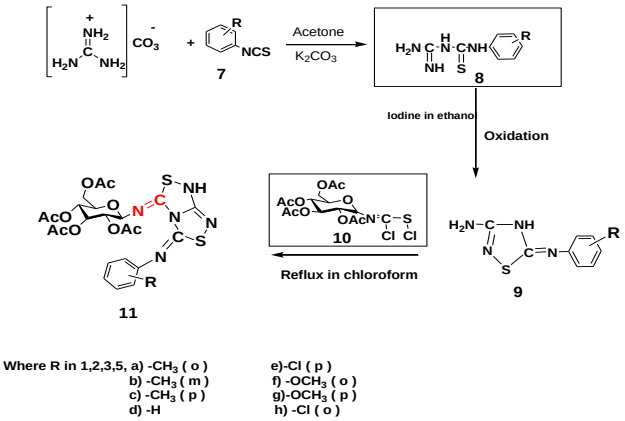
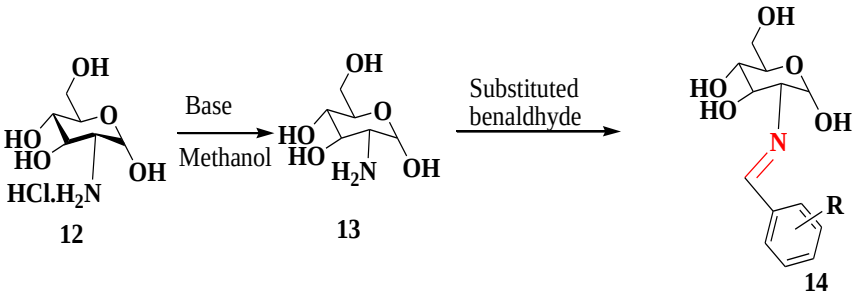


Figure 4. Reaction of phenyl isothiocyanated7 reacted with guanidine carbonate to form thioguanidine derivatives 8

Irvine [14] and other authors [15] had used sodium bicarbonate for conversion of α-D-glucosamine hydrochloride into α-D-glucosamine base, whereas, another authors [16-18] had used NaOH for elimination HCl. For finding the most suitable basic conditions in this conversion, we have investigated the role of some inorganic bases (such as NaOH, $NaHCO_3$ and Na_2CO_3) and organic bases (such as triethylamine, DABCO, pyridine and piperidine) in synthetic reaction of azomethines from α-D-glucosamine hydrochloride using methanol assolvent (Figure 5).

D Thanh, et al. [19], showed that azomethines of α-D-glucosamine with substituted salicylaldehydes **14a-c** are thermal stable, almost other remained azometines, if formed, are thermal unstable, so the recrystallization of these azomethines lead to decrease the reaction yield, or to decompose the formed product, even some azomethines **14** become black in air. In case of azomethines of α-D-glucosamine with substituted salicylaldehydes **14a-c**, intramolecular hydrogen bond between nitrogen atom of azomethine bond and hydroxyl group on carbon C-1 of monosaccharide component through lone-pair electron on this nitrogen atom make azomethe molecule to be stable [20] (Figure 6).



R= 2-OH (3a); 2-OH-5-Cl (3b); 2-OH-5-Br (3c); 4-OMe (3d);
3-OMe (3e); 3-OEt (3f); 4-Me (3g); 3-OMe-4-OH (3h);
3-OEt-4-OH (3i); 4-iPr (3j); 4-Cl (3k); 4-OH (3m);
4-NMe_2 (3n); 3-NO_3 (3p); H (3q)

Figure 5. Reaction of α-D-glucosamine substituted benzaldehyde

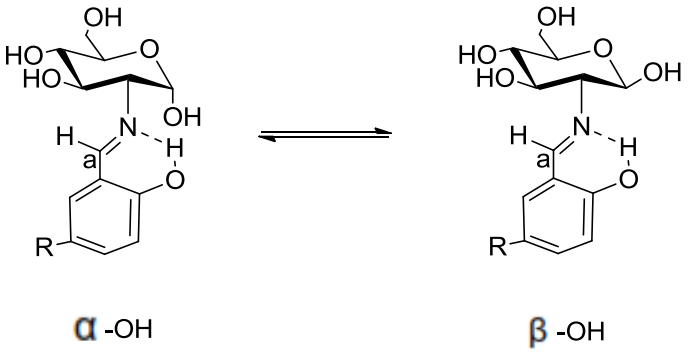


Figure 6. Intramolecular hydrogen bond in substituted salicylidene α-D-glucosamines

Synthetic methods were reporting for the direct *N*-glycosylation of aromatic compounds and carbohydrates without hydroxyl protection or activation were few [21-28]. Generally, the reaction performed by heating at reflux in protic solvents and, in some reactions, the best results achieved under mildly acidic conditions [21-28]. That, the reaction is strongly influenced by the reactivity of the starting amine and the best preparative yields were obtaining for aromatic amines of moderate basicity [29-31]. A series of *N*-(1,10-phenanthrolin-5-yl)-β-glycopyranosylamines

16a-e were synthesized by treatment of 5-amino-1,10-phenanthroline with different unprotected monosaccharides **15a-e** using $(NH_4)_2SO_4$ as a convenient catalyst promoter. *N*-phenanthrolineglycosylamine **16a-e** was reacted with ethanoic solution of Cu $(NO_3)2{\cdot}3H_2O$ in dimethyl sulfoxide and heating the mixture at 65 ºC for 6 h., to yield. The copper (II) complexes **17a-e.** Cu complexation improved the solubility properties of the *N*-glycopyranosylamines. This behavior is particularly important for the potential applications of these metallo-organic derivatives (Figure 7).

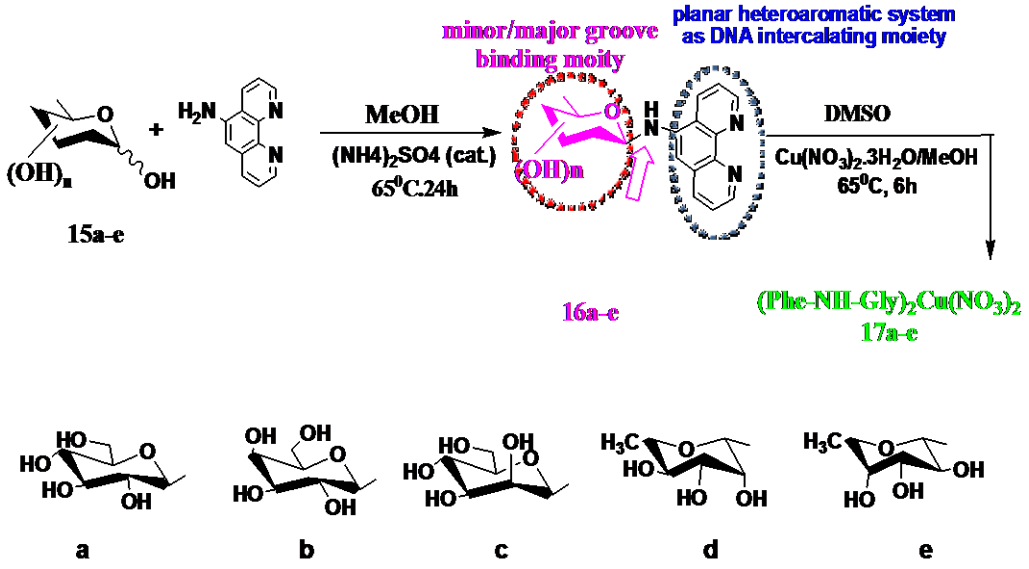


Figure 7. Synthetic methods for N-glycosylation of aromatic compounds and carbohydrates

Lately, we reported that S-configured homoallylamines were synthesized diastereoselectively by used Lewis acid that it causes addition of allylsilanes to Schiff bases of 2,3,4,6-tetra-*O*-pivaloyl-*β*-*D*-galactopyranosylamine **18** (Figure 8) [32-33]. Strecker, [34] Ugi [35] Mannich [36] and tandem Mannich-Michael reactions [37,38] have been used chiral auxiliary in diastereoselective. The stereocontrol in these reactions was causing by the complexing ability of them oxygen functions of the carbohydrate toward Lewis acids in combination with the pronounced chirality of the carbohydrate. Analogously, the (R)-homoallylamines **23** are available by using the corresponding 2,3,4-tri-O-pivaloyl-8-L-fucopyranosylamine (**21**) [39].

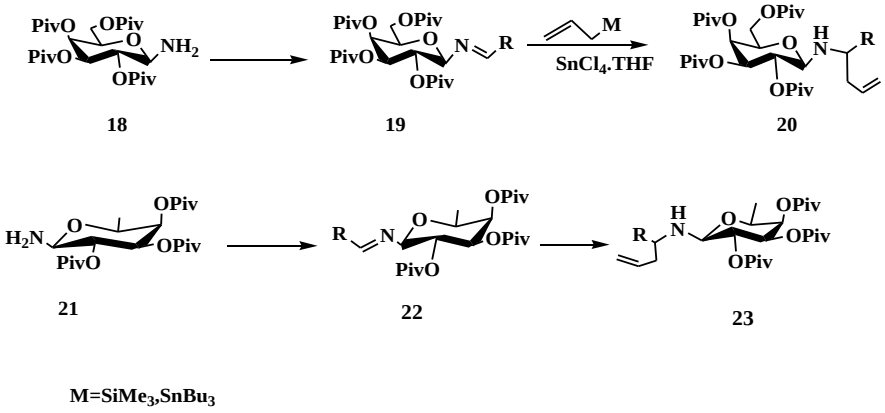


Figure 8. Synthesis of N-Glycosyl-N-homoallylamines

The differentiation between the studies of glycosylcarbamides and other substituted carbamides without introducing glycosyl moiety has higher effect on antimicrobial activity. The carbamides have an important role in pharmacological field [40] and many of these compounds have a wide applying in carbohydrate chemistry [41]. Therefore, a series of novel glycosyl-3-o-tolyl carbamides **26** produced by the interaction of different glycosylamines **24** with o-tolylisothiocyanate [42] **25** in pyridine solvent for 24 h at room temperature. The chemical structures of the title compounds **25**were deduced by IR, ^{1}H NMR, mass spectral analysis (Figures 9 and 10).

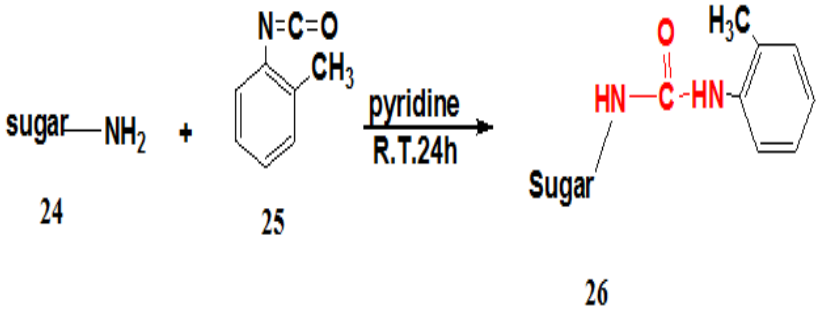


Figure 9. Glycosyl-3-o-tolyl carbamides (Sugar)

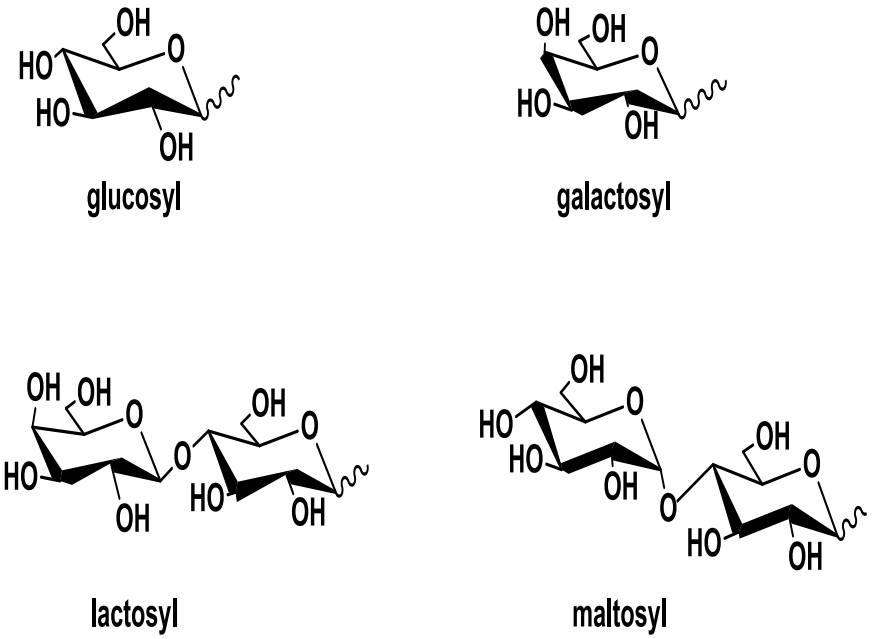


Figure 10. Structure of Glucosyl, galactosyl, lactosyl and maltosyl

The observation that the introduction the hydrazide moiety [43] such as diacylhydrazines (**B**, Figure 10) [44], semicarbazides (**C**, Figure 10) [45], and acylhydrazones (**D**, Figure 10) [46] derivatives that contain 5-phenyl-2-furan showed diverse and significant bioactivities such as fungicidal, insecticidal, and antitumor activities. Glycosylhydrazides **29** [47-56] synthesized by reaction of equimolar amounts of the corresponding 5-substituted phenyl-2-furoyl hydrazide **28** with the monosaccharides **27** D-glucose, D-galactose, D-mannose, D-fucose, and D-arabinose in ethanol (Figures 11 and 12).

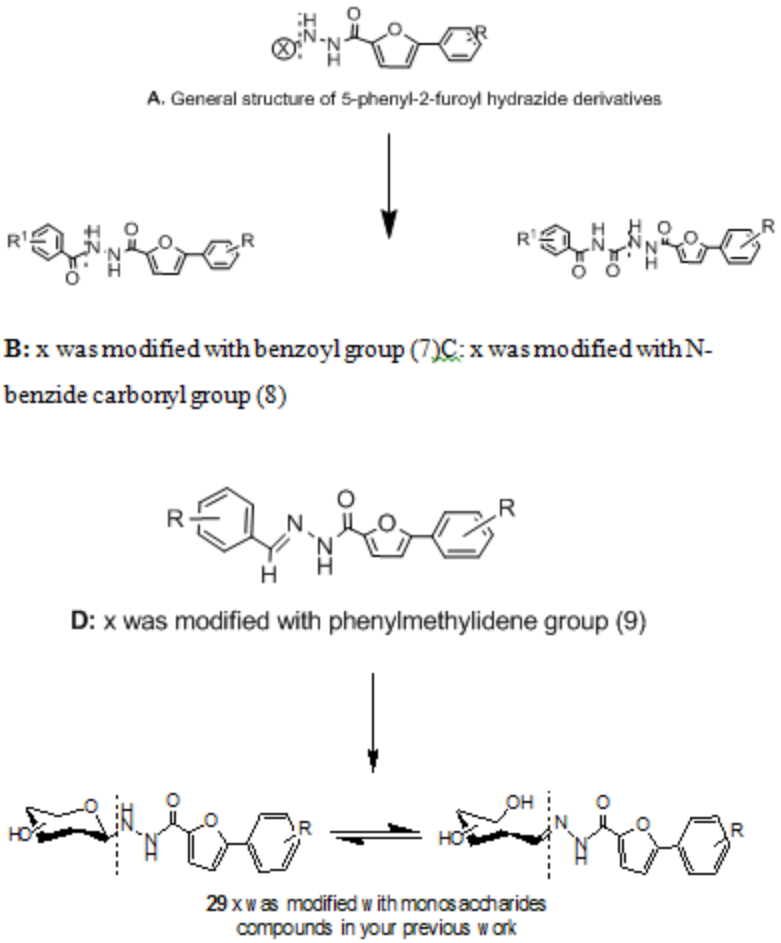


Figure 11. Designed Strategy for title compound

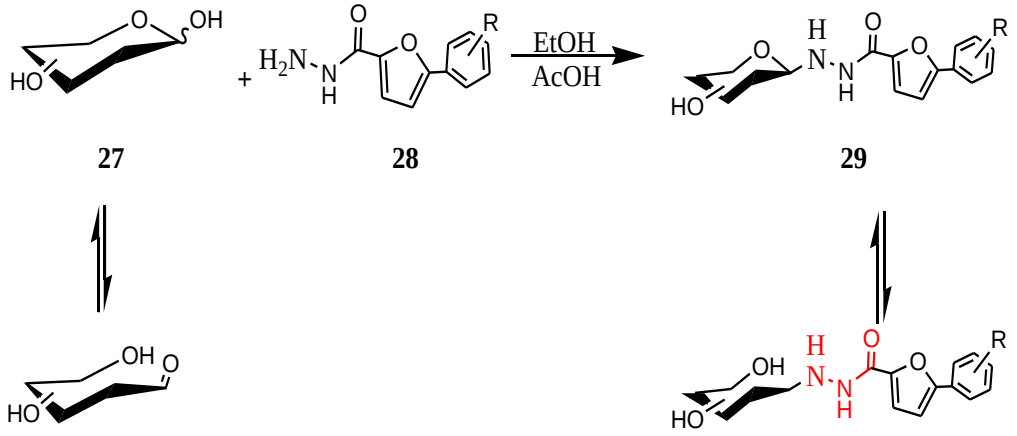


Figure 12. Condensation reactivity and equilibrium between cyclic and acyclic formsof the monosacchride

Ghoneim, et al. [56] was preparation of *N*-(2,3,4,6-tetra-*O*-acetyl-*β*-D-glucopyranosyl)-*N*`-(-4-amino-5-cyano-2-(phenylimino) thiazol-3 (*2H*)-yl)propylideneamino) benzenesulfonic acid) thiourea **32** by nucleophilic addition of (*4E*)-4-(1-((*2E*)-4-amino-5-cyano-2-(phenylimino)thiazol-3(*2H*)-yl) propylideneamino) benzenesulfonic acid **30** on 2,3,4,6,-tetra-*O*-acetyl-*β*-*D*-glucopyranosylisothiocynate **31**. The reaction was proceeded in the microwave oven for

25-30 min, using dioxaneas as a solvent. IR spectrum of compound **9** showed stretching band of C=S bond at 1374 cm^{-1}, another band at 3489-3165 cm^{-1} due to N-H bands and C=O bonds of glucopyranosyl at 1745cm^{-1}. The mechanism for the formation of derivative **32** is proceed by nucleophilic attack of the lone pair of electrons on NH_2 group at the Carbon of isothiocynate group followed by H transfer to form the thiourea derivative as shown in Figure 13.

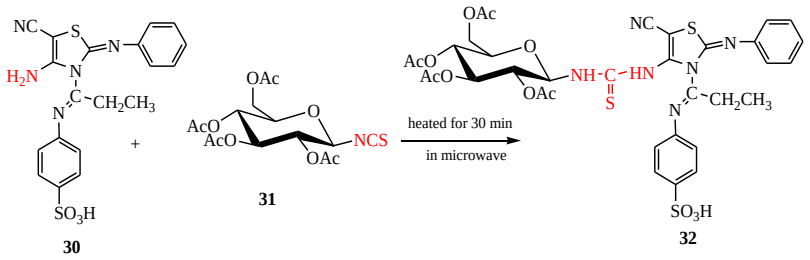


Figure 13. preparation of *N*-(2,3,4,6-tetra-*O*-acetyl-*β*-D-glucopyranosyl)-*N*`-(-4-amino-5-cyano-2-(phenylimino) thiazol-3 (*2H*)-yl)propylideneamino) benzenesulfonic acid) thiourea32

A series of new thiourea and urea derivatives have synthesized by the reaction of multicomponent such as phenol, benzaldehyde and urea or thiourea to yield a new amides derivatives **34a, 34b** and **34c** respectively. The amide compounds have condensed with a different monosaccharide sugars like (D-glucose, D-galactose, and D-fructose) in the presence of absolute ethanol at 80 °C for 8 h., in yield 78% toafford new N-glucoside The structure of all compounds have confirmed by ^{1}HNMR and IR spectra. The configuration of all glycosides derivatives were β based on indication from ^{1}HNMR where the coupling constant (J=6 to 7 MHz) of anumeric protons in these compounds (Figures 14 and 14A) [49].

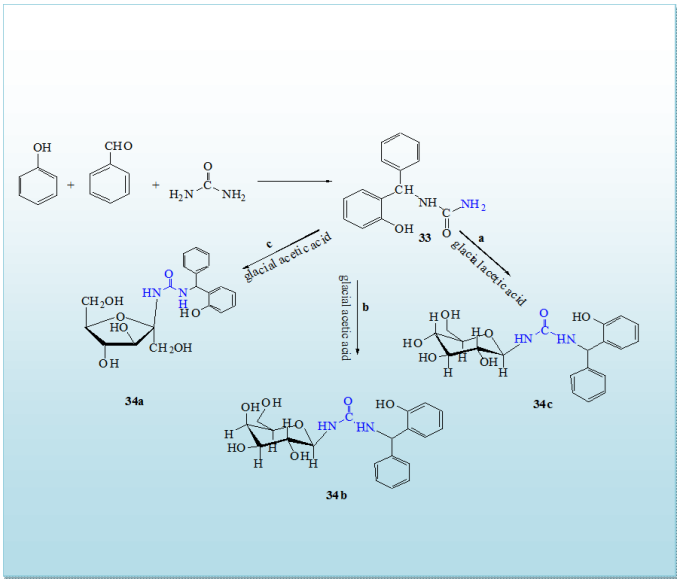


Figure 14. Synthesis of amides derivatives from phenol, benzaldehyde and urea or thiourea

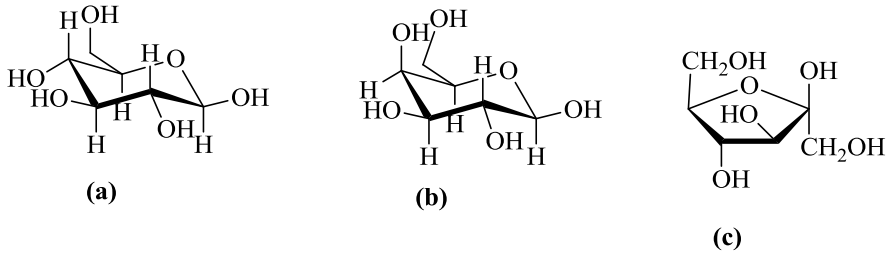


Figure 14A: D-glucose (a), D-galactose (b), and D-fructose(c)

N-Glycosylaminecompounds **37** [50] were synthesized by reaction of 2-amino quinoline **35** with active hydroxyl group in the 4,6-O-protected-*D*-glucose **36** (Figure 15) [51,52].

Furthermore, aminoazobenzene derivatives **38** [53] reacted with 4,6-*O*-protected-D-glucose **36** to give Azobenzene based *N*-Glycosylamine derivatives **39** (Figure 16). Synthesized of *N*-glycosylamines, derivatives used of partially protected saccharide [54] gel formation were determined and this observation prompted us to go for the study of the gelationproperty of azobenzene/quinoline containing saccharide compounds (**37** and **39**).

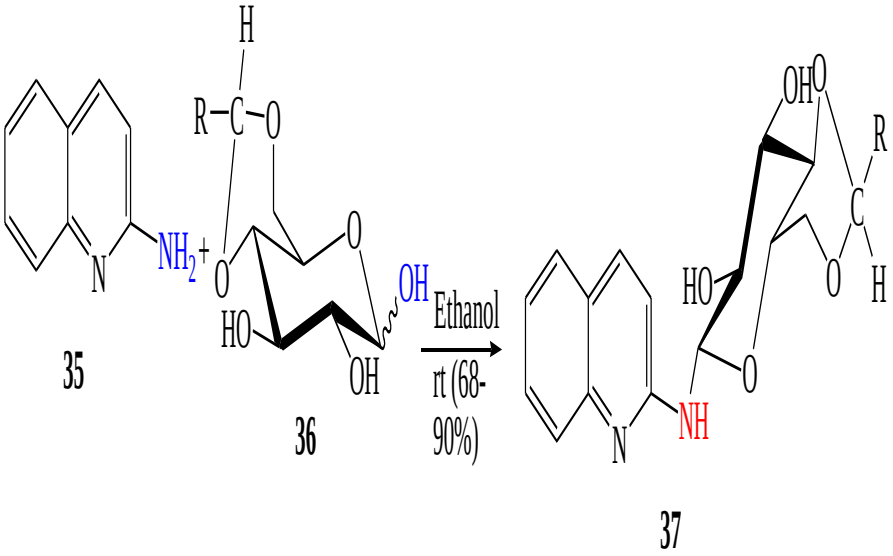


Figure 15. Synthesis of N-Glycosylaminecompounds (R = C_3H_7, CH_3)

The chacterised of all the synthesized *N*-glycosylamines were confirmed using NMR (^{1}H and ^{13}C) andelemental analysis. 2,3,4,6-tetra-O-acetyl-β-D-galactopyranosyl bromide and 2,3,4,6-tetra-O-acetyl-β-D-glucopyranosyl bromide **41** were prepared by a reported method [55]. The thermodynamically more stable α-anomers were forming. The halogen in the acylglycosy **41** halide is reactive and may be readily displaced by an azido group. In the case of D-(+)-galactose and D-(+)-glucosederivatives, the replacement involves inversion of configuration at the anomeric site and thus the glycopyranosyl halide yields a *β*-glycopyranosylazide through an oxonium ion. Schiff bases **46** were obtained by condensation of 2,3,4,6-tetra-O-acetyl-β-D-galactopyranosyl amine and 2,3,4,6-tetra-O-acetyl-β-D-glucopyranosyl amine **43** with different aromatic aldehydes **45** in refluxing ethanol (Figure 17). The IR spectrum of these Schiff bases showed an absorption band at 1627-1635 cm^{-1} for the imine group. The ^{1}H-NMR spectrum showed a singlet for azomethin (CHN) of Schiff-bases at 8.50 ppm.

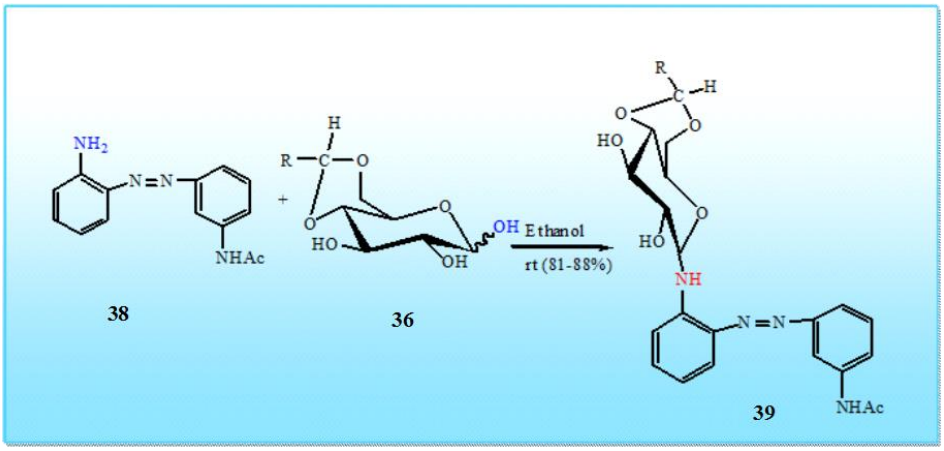


Figure 16. Azobenzene based N-Glycosylamine derivatives (R = C_3H_7, CH_3, C_6H_5)

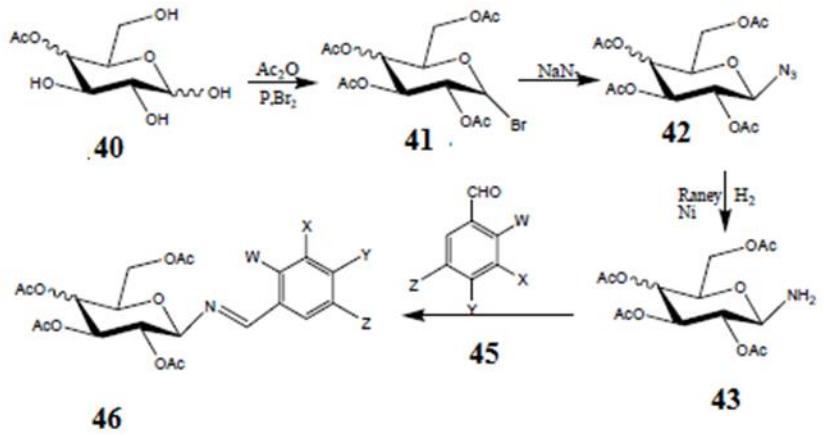


Figure 17. Schiff bases was obtained by condensation of 2,3,4,6-tetra-O-acetyl-β-D-galactopyranosyl amine and 2,3,4,6-tetra-O-acetyl-β-D-glucopyranosyl amine 43 with different aromatic aldehydes

In 2000, Ichikawa reported a similar reduction of a-azide **47** and subsequent treatment with cyclohexyl isocyanate **48** produces an inseparable mixture of α- and β-ureas, **49** (Figure 18) [56]. This result confirmed an earlier one from Ogawa that the a-azide reduction proceeds without retention of anomeric stereochemical integrity [57-66].

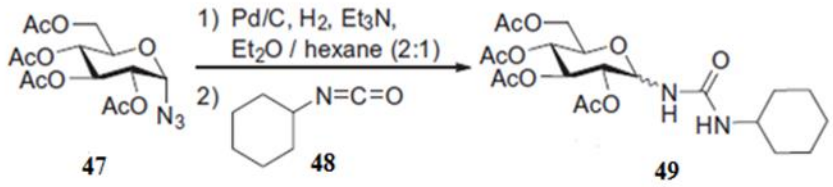


Figure 18. Reduction of a-azide 47 and subsequent treatment with cyclohexyl isocyanate 48 produces an inseparable mixture of α- and β-ureas 49

The 4,6-O-ethylidene-N-glucosylamines **53** and **54** containing thiazole **51** and benzothiazole **52** ring have been synthesized from 4,6-*O*-ethylidene-*D*-glucopyranose **50** as following reactions: A simple procedure has been adopted to synthesize different *N*-glucosylamines of the partially protected glucose with different type of amines. Glacial acetic acid has been used as catalyst for these reactions. The amines were substituted 2-amino-4-phenylthiazoles and substituted 2-aminobenzothiazoles. Obtained *N*-glucosylamines were crystalline solid with high melting point, dissolving in common organic solvents (ethanol, DMF, dioxane …). Their structures have been confirmed by using spectral methods (FTIR-IR, ^{1} H-NMR-, ^{13}C-NMR and mass spectra) (Figure 19).

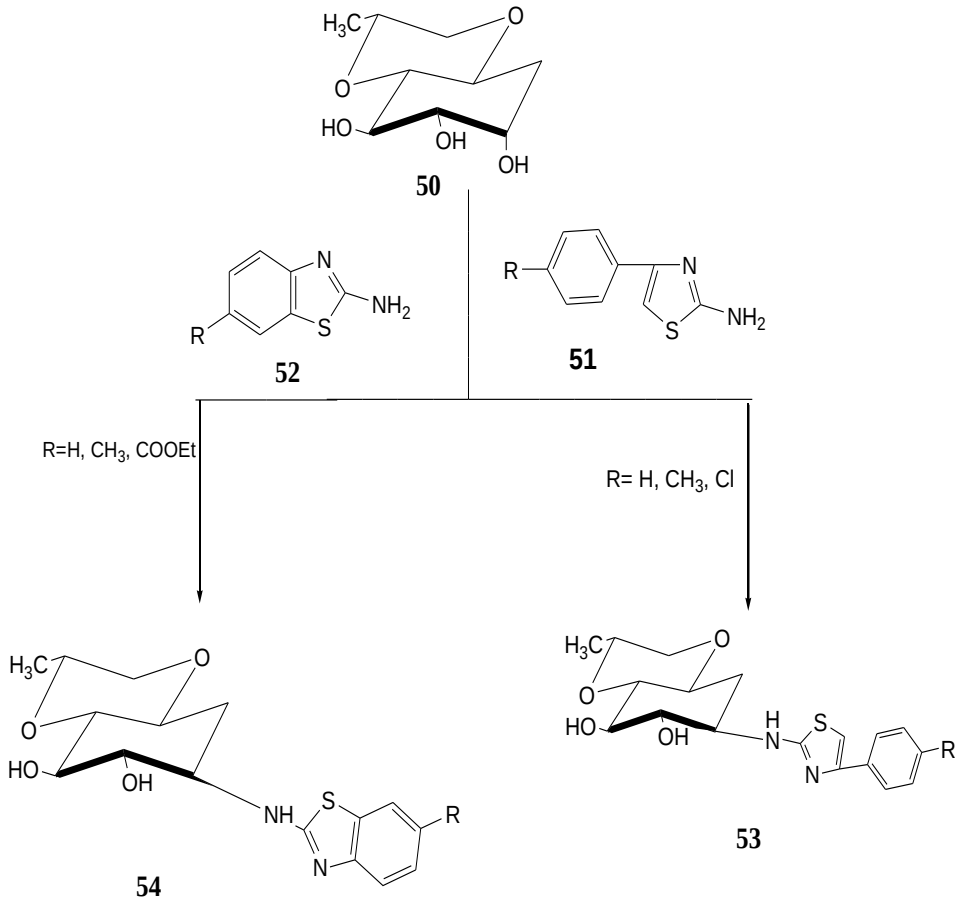


Figure 19. The 4,6-O-ethylidene-N-glucosylamines 53 and 54 containing thiazole 51 and benzothiazole 52 ring have been synthesized from 4,6-O-ethylidene-D-glucopyranose 50

DISCUSSION AND CONCLUSION

In this review, we report on the efficient procedures for the synthesis of *N*-glycosyl amine derivatives from different heterocylic compounds such as (aminoazobenzene derivatives, 2-amino quinoline5-amino-1,10-phenanthroline, thiazoleand benzothiazole) with glycopyranosylisothiocyanates, 2,3,4,6-tetra-*O*-pivaloyl-*β*-*D*-galactopyranosylamine and different monosaccharide sugars like (D-glucose, D-galactose, and D-fructose). The chemistry of glucosylamine derivatives has an important in in the pharmaceutical field.

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