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Recent Modification of Polyvinyl Chloride (PVC) *via* Heterocyclic Compounds

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Abstract

Most of polymers polymers instead of Polyvinyl Chloride (PVC) were degraded by UV or thermal, so most researchers want to solve this problem by modifying these polymers and realizing their importance instability (thermal and photo), bioactivity, and …..etc. The driving force in polymer science is generating novel materials for variable applications. The success of the new materials relies mainly on the ability to be manufactured on a large scale and in high quality. Polyvinyl Chloride (PVC) is one of the famous polymers used by manufacturers all over the world. Therefore, we are going to consider polyvinyl chloride (PVC) as one of the polymers acting as a distinctive material with a vast application domain. Modification of PVC with synthesized heterocyclic derivatives such as pyrazole, imidazole, thiazole, triazoles, and …… etc, enhances its stabilization, bioactivity, and rel1atively high chemical resistance to be an abundant and affordable cost. As the world and the needs of civilization are changing, new creative ideas for the applications of PVC are continually emerging It is undoubtely one of the most promissing continually improved raw materials of the future.The PVC showed progress upon applying nucleophilic substitution reactions. The biological compatibility of PVC during the surface modification process is important to ensure the effectiveness of the enhanced PVC. The heterocyclicare the nucleophiles that facilitate the structural unit of the synthesized molecules. The chemical heterocycles are bonded to (PVC) forming new molecular adducts with various physical and chemical properties.

*Keywords***:** Polyvinyl Chloride; Modification of PVC; Quaternary PVC, Polymers, Heterocyclic compounds, Amino compounds

1. Introduction

Polymers play an important role in our lives because many objects we use daily are made of them, such as packaging, films, covers, bags, and containers. They are also used in construction, electrical, and electronic applications [1-4]. For over a century, polyvinyl chloride, abbbreviated as PVC, has been used in almost all branches of trade and industryas well as in our homes. Polyvinyl chloride is nearly everywhere you look[1–5].Polyvinyl chloride has a great impact on our environment. They are used in different applications ranging from domestic, construction to high tech application[6,7]. Poly(Vinyl chloride) is one of the world's largest thermoplastic materials [8], due to its valuable properties [9], widespread use, high chemical persistence, barriers, and low cost [10–13]. Since its synthesis in the early years of the last century; utilizing PVC is renowned worldwide as well as exponential research work has been performed [14–16]. The importance of PVC comes from the low creation costs and the extraordinary adaptability of vinyl chloride polymers are the two significant explanations behind their large portion of the plastic market [17], which was broadly utilized in enterprises including design, electronics, synthetic designing, bundling, transportation [18]. The chemical modification of PVC generates an opportunity to produce a novel material with interesting features. The great exhibition of PVC items has expanded the use of this polymer in buildings, basically in outside applications, for example, window profiles, cladding designs, and siding [10]. The polymer can be changed into various items displaying an incredibly wide scope of properties, both physical and chemical by utilizing altering specialists, like plasticizers, such as epoxidized vegetable oil [19–22], polymer plasticizer [23–26], polyol ester [27], phosphate plasticizer [21,28], and fliers and stabilizers [29–31].

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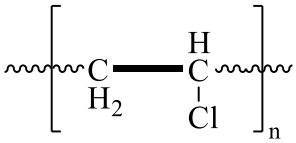
PVC homopolymers are linear and strong. The monomers are mainly arranged head to tail, meaning that chloride is located on altrnating carbon centres. PVC has mainly an atactic stereochemistry, which means that the relative stereochemistry of the chloride centres are random. Some degree of syndiotactcity of the chain gives a few present crystallien that is influentail on the proprties of the



material, (figure 1)[32,33].

Fig. 1, Repeat-unit (PVC polymer)

PVC is commercially produced through different methods comprising emulsion polymerization, bulk polymerization and suspension polymerization .
PVC is used in so many frequent and ubiquitous applications that it creates a waste stream that needs to be handled safely. These Modification's, which include replacement and elimination, are essentially related to the dechlorination process. PVC's chemical functionalization appears to be permanent due to the characteristics of the material that are pertinent to its uses. The most innovative aspect was the way PVC and other polymers' surfaces were modified using the recently created by graft polymerization process but in this review the modifications were carried out by new synthesized heterocyclic compounds.

2. Modification of PVC using Heterocyclic compounds:

PVC, in particular, is very important polymer in terms of industerial applications. One of the drawbacks of PVC is that suffering from degredation which affects its performance under working conditions [1–5]. Therefore our review is focused to cover some modifcations of PVC using heterocyclic compounds.

2.1. General Considerations

It is important to consider that PVC modification aimed to three main goals:
1. Biological compatibility of PVC during surface modification process, especially in the medical field.
2. Improvement of the mechanical properties in terms of stretching and ductile strength with maintaining translucency.
3. Metal Chelation especially in heavy water and municipal wastes.
The term (heterocyclic) in the retro-synthetic analysis means a molecular part, which is an effective starting reagent that plays an active role in targeting the molecule during the chemical reaction. In our case, the heterocyclic derivatives are the nucleophiles that are the structural unit of the desired molecules to be synthesized. The combination of these chemical derivatives with PVC leads to new molecular adducts. The latter are different in their physical and chemical properties. A chemical modification of PVC is investigated as a worthy method to upgrade and improve the quality and intrinsic characteristics of the PVC by maintaining the major features of the base polymeric matrix [36–39]. Moreover, optimized features with unprecedented properties of the modified PVC matrix are expected. Without any chemical additions, PVC would be a solid rigid dense material with inferior mechanical properties. The optimum method to produce flexible PVC is to introduce a nucleophilic chemical modification to the matrix. During the past two decades, research papers concerning the modification of PVC have been carried out [40,41]. The focus of these reports is the formation of a (CPVC-X) bond between the PVC carbon and the modifier X (e.g., N, O, S, etc…) [42–47]. This modification was applied *via* the introduction of a modifier of nucleophilic nature as shown in Figure 2 [48]. Scientists are concerned primarily with the biological tolerance of the modified PVC materials which are tested against nosocomial infections as the main assessment in the modification process. The most commonly known infections are *Staphylococcus aureus* (Gram-positive (G +ve)) pneumonia associated with ventilators and surgery. Moreover, *Escherichia coli* (Gram-negative (G -ve)) is mostly related to urinary tract catheters and the gastrointestinal tract. The second concern is the internal plasticization of PVC by chemical modification to improve its mechanical properties. Thus, it provides enduring stabilized materials.

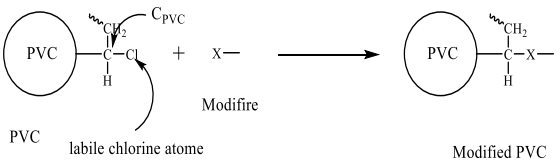


Fig. 2, Proposed Structures of PVC and modified PVC

2.2. PVC –N modification:

a) Diamino group

Statistics have confirmed that 40% of the medical wastes in hospitals are PVC-based materials. This high percentage is related to what is known as biomaterial-related infections (BRI), especially bacteremia due to the usage of PVC [49]. It is reflected in the exponential increase in the number of research articles to enhance the PVC properties [50–52]. Approaching this idea, the decreased bacteremia risk associated with blood storage and

transfusion, in PVC bags, and *IV* tubes is crucial. Additional restrictions are to be taken into consideration to provide polymeric surfaces with biological compatibility [12,53]. This is the reason for the cationic PVC to be introduced on a positively charged surface exhibiting antibacterial characteristics figure 3 [54–57].

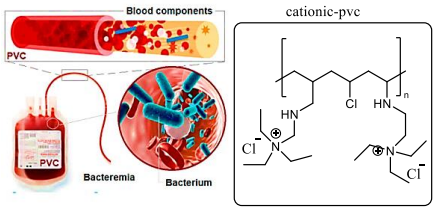


Fig. 3, suggests the biomedical capacity of surface cationic PVC

In this reaction, the quaternary PVC formation mechanism can be accomplished and can be perceived by the primary amine -NH_2 bonds appearance (Figures 4a & b). This is followed by a second step of the reaction, the quaternary ammonium groups in the modified PVC appear simultaneously with the secondary amine -NH bonds.

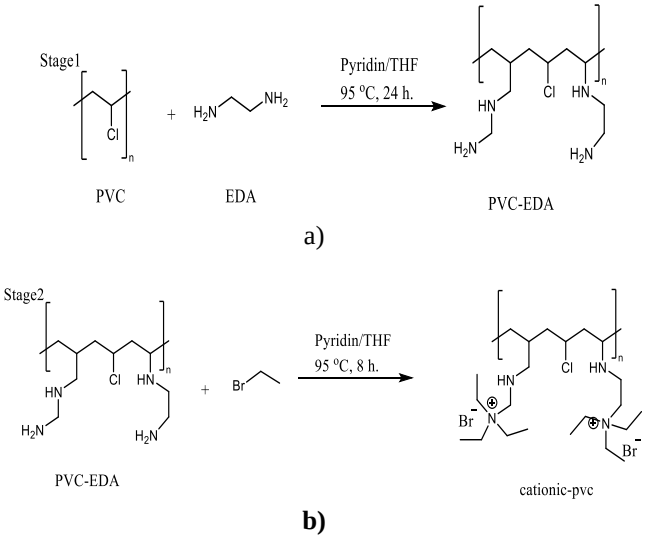


Fig. 4 a,b, Chemical reactions associated with obtaining cationic PVC

In addition, the functionally enhanced derivative spectroscopy (FEDS) algorithm has been implied as a reference. Further confirmation is asserted for the structure *via* the administration of the ^{1}H-NMR technique [58].

In addition, the biological properties revealed a relative increase in antibacterial activity against gram-positive and gram-negative (*S. aureus* and *E. coli*), respectively. In Table 1, these results are evident to form a biomedical surface with modified biomedical properties in the comparison with amoxicillin and mannitol as antibiotics used for these gram-positive and gram-negative bacteria.

b) Pyridyl groups

4-Vinylpyridine(4VP) and vinyl acetate (VA) was chosen to graft PVC for enhancement of flux and selectivity [59,60]. In graft copolymerization, PVC takes part in the polymerization of 4VP or copolymerization of VA by forming active sites which makes covalent bond with the growing polymer chains of 4VP or 4VP and VA. The active site on PVC is generated by irradiation with Co60γ rays in radiation grafting [59–61]. PVC macro radical is formed by abstraction of hydrogen and chlorine from PVC backbone [62]. The initiator benzoyl peroxide (BPO) first decomposes to generate BPO free radicals. The BPO radicals initiate polymerization of 4VP or copolymerization functional groups of clay and the polymers as shown in figure 5.

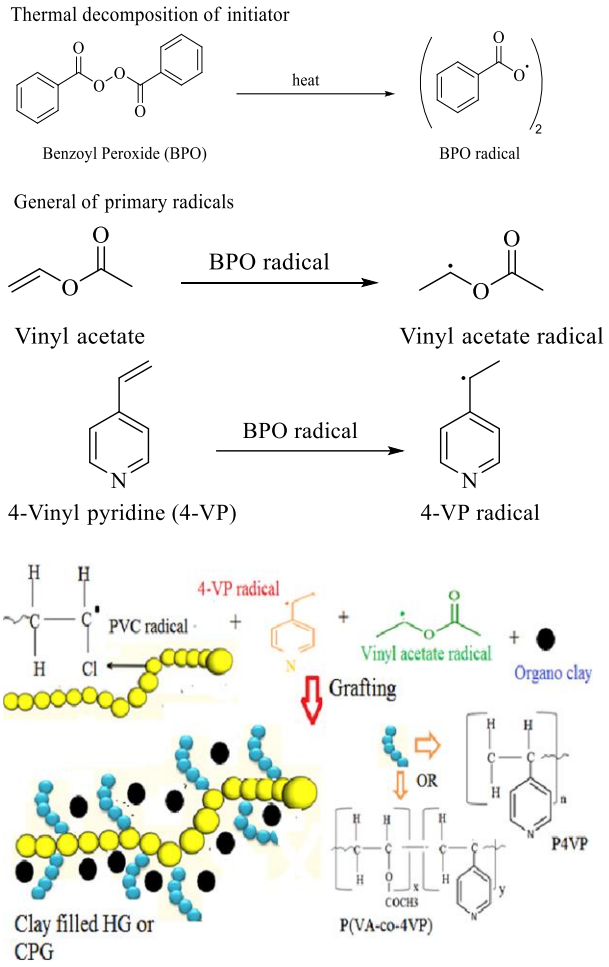


Fig. 5, Synthesis mechanism for grafted PVC

Table 1: Mean inhibition percentage for PVC, PVC-EDA, and cationic PVC against *E. coli* and *S. aureus* [66]

sample	*E. Coli*		*S. aureus*	
	CFU/ml. (x10^9)	Inhibition (%)	CFU/ml. (x10^9)	Inhibition (%)
PVC	2.15 ± 0.12	5.5 ±5.4	2.20 ± 0.07	4.9 ±3.0
PVC-EDA	1.90 ± 0.16	16.5 ± 6.9	2.17 ± 0.09	6.6 ± 4.1
Cationic –PVC	1.54 ± 2.25	32.2 ± 2.1	1.44 ± 0.29	37.8 ± 9.0
Mannitol	2.15 ± 0.08	5.17 ± 3.7	2.29 ± 0.10	0.9 ± 4.5
Amoxicillin	0.13 ± 0.06	95.0 ± 2.5	0.31 ± 0.16	89.8 ± 6.6

c) Imidazole groups:

Recently, it has been reported that modifying PVC [63–65] has been carried out using eight different imidazole derivatives producing new polymer membrane adducts with improved quality such as removal of heavy metals from the aqueous solution (Figure 6) [66,67]. ^{1}H-NMR and FTIR were applied to confirm some of the new structures *via* SN_2 substitution as depicted. The decrease in band intensities at 2905 cm^{-1} and 602 cm^{-1} are due to -CH and -CCl groups in PVC [15]. The appearance of new bands at 3363 cm^{-1} and 1455 cm^{-1} can be correlated to the presence of -OH and -C=N groups. Simultaneously, the imidazolium groups showed the absorption bands at 2920 cm^{-1} and 2850 cm^{-1} due to the saturated alkyl imidazole -CH chain [49,65].

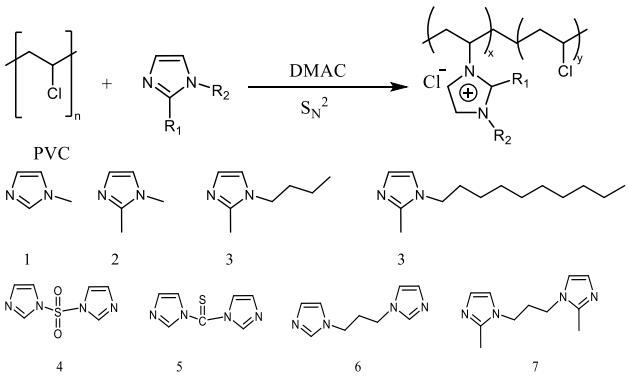


Fig. 6, The synthesis of imidazolium from PVC

The increase in the imidazole percentage has expanded the PVC matrix and upgraded the mechanical stability of the modified PVC. The flexibility and relatively ordered matrix of the highly saturated PVC can be noticed. These new

modified PVC membranes are subjected to further studies, purposing to decrease the internal plasticization. In other words, it is desired to maintain the flexibility of PVC; maybe by increasing the spaces between the chains with a simultaneous decrease in chain interaction.

d) Pyrazolone derivatives:

Nitrogen heterocycles; as azoles are favored for structure stability due to their electronic ring cloud distribution. This feature facilitates their ability to attach to receptors and enzymes [76]. The modification of the backbone PVC polymer has been carried out with pyrazolone derivatives followed by the loading of silver nanoparticles. This procedure has led to the enhancement of the biological properties of the modified PVC (e.g., antibacterial and antifungal activities) (Figure 7) [15,77]. The two-step mechanism is performed *via* an initial nucleophilic substitution of the PVC labile chlorine forming an intermediate. The second step is a substitution reaction carried out by the active pyrazolone methylene group forming a two-strain PVC attachment. To improve the biological compatibility of the enhanced PVC, silver nanoparticles were added at 3% and 5% levels. A scanning electron microscope (SEM) has clarified the homogeneity of the modified pyrazolone PVC with silver nanoparticles. Moreover, transmission electron microscopy (TEM) was employed to confirm the ranging size of Ag nanoparticles in the range of 16-22 nm. Finally, biological compatibility has been examined. A noticeable increase in antibacterial activity was observed. The addition of pyrazolone and silver nanoparticles to the backbone of the PVC matrix has led to the inhibition of bacterial growth efficiently as tested against *Escherichia Coli* and *Staphylococcus Aureus* (Table 2).

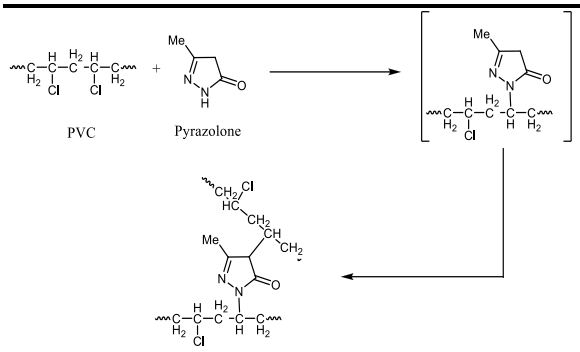


Fig. 7, modification of the backbone PVC polymer by methylpyrazolone

e) Vinyl imidazole:

A relevant work was performed to examine the effect of silver and copper nanoparticles effect of the modified PVC vinyl imidazole (Figure 8). The antibacterial and antifungal efficiencies of modified PVC/Ag-NPs and PVC/Cu-NPs have revealed promising results compared to the standard used (Table3)[91]

Table 2: Antimicrobial activity of PVC-pyrazolone derivatives in the absence and in the presence of 3% and 5% by weight of AgNPs in comparison with PVC [89]

Sample	Inhibition zone diameter (mm/mg Sample)							
	B. Subtilis	*E. Coli*	*N. gonorrhoeae*	*P. aeruginosa*	*S. aureus*	*S. faecalis*	*A. flavus*	*C. albicans*
Activity/Gran reaction	(G^+)	(G-)	(G^-)	(G^-)	(G^+)	(G^+)	(Fungus)	(Fungus)
Control: DMSO	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Standard								
Ampicillin	20	22	20	17	18	18	-	-
Amphotericin B	-	-	-	-	-	-	17	19
1:PVC	13	16	15	12	12	10	6	8
2:PVC/Pyrz	15	18	16	14	13	11	8	10
3:PVC/Pyrz-Ag 3%	17	21	19	15	15	14	11	13

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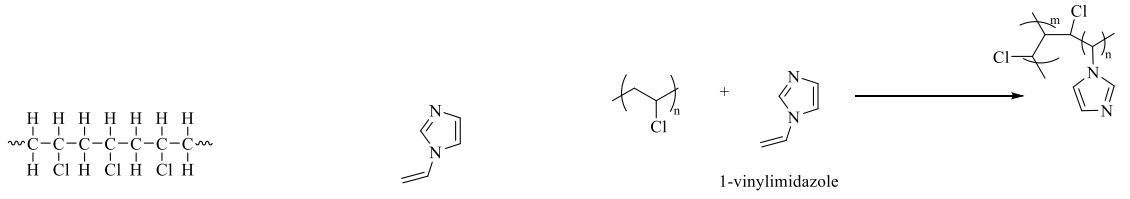

Fig. 8, PVC, and Imidazole derivative structure

Table 3: Antibacterial activity of PVC/VI in the absence and in the presence of Ag-NPs and Cu-NPs compared to blank PVC [82]

Sample	Gram +ve bacteria inhibition zone (mm)			Gram –ve bacteria inhibition zone (mm)		
	B. subtilis	*S. aureus*	*S. faecalis*	*E. coli*	*N. gonorrhoae*	*P. aeruginosa*
DMSO	-	-	-	-	-	-
Ampicillin	20 ± 0.25	18 ± 0.22	18 ± 0.17	22±0.28	20 ± 0.22	17 ± 0.19
PVC	12 ± 0.15	12± 0.24	9 ± 0.21	15± 0.18	12 ± 0.20	11 ± 0.27
PVC/VI	16 ± 0.21	14 ± 0.19	10 ± 0.23	18±0.22	17 ± 0.25	15 ± 0.23
PVC/VI-AgNPs 0.5%	16 ± 0.19	15 ± 0.20	11 ± 0.06	20±0.29	17 ± 0.28	15 ± 0.26
PVC/VI-AgNPs 1%	17 ± 0.23	15 ± 0.18	12 ± 0.24	22±0.32	18± 0.23	16 ± 0.18
PVC/VI-AgNPs 2%	18 ± 0.09	16 ± 0.23	13 ± 0.31	23±0.24	19 ± 0.17	17 ± 0.25
PVC/VI-AgNPs 4%	19 ± 0.17	17 ± 0.24	18 ± 0.26	26±0.19	19 ± 0.09	20 ± 0.28
PVC/VI-CuNPs 0.5%	16 ± 0.13	14 ± 0.32	12 ± 0.22	18±0.28	17 ± 0.34	15 ± 0.42
PVC/VI-CuNPs 1%	17 ± 0.41	15 ± 0.08	13 ± 0.27	18±0.17	17 ± 0.26	16 ± 0.24
PVC/VI-CuNPs 2%	17 ± 0.22	15 ± 0.25	15 ± 0.18	18±0.32	18 ± 0.24	16 ± 0.28
PVC/VI-CuNPs 4%	18 ± 0.17	16 ± 0.21	16 ± 0.23	20±0.25	19 ± 0.31	18 ± 0.31

f) Triazole derivatives and click reaction:

PVC having a side group capable of undergoing a substitution reaction that provide a suitable platform to alter the original properties of the polymer towards more desirable charactrestics to be used in different applications [81].A modification of the PVC was carried out using Schiff base originating from a 1,2,4-triazole and aldehyde reaction. The successfully modified adduct was produced *via* the SN^2 mechanism, followed by the addition of Cu(II),the latter enhanced the photostability characteristics (Figure 9) [82] of the enhanced polymer. The copper ligand attachment has been subjected to further studies. It allows the adsorption of heavy metals by chelating into the PVC-Schiff base matrix.

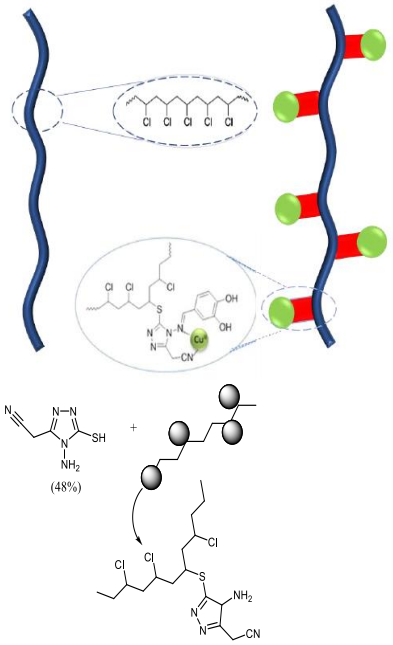

Fig. 9, Structure of Schiff base with suggested ligand coordination of PVC-L-Cu(II).

Triazoles and PVC have the capacity to separate heavy metals from aqueous solutions [83,84]. Chemical modification of PVC was carried out using triazoles *via* click reaction for the extraction of heavy metals such (Cd, Cu, Ni, Cr, and Pb) (Figure 10)[84,85].

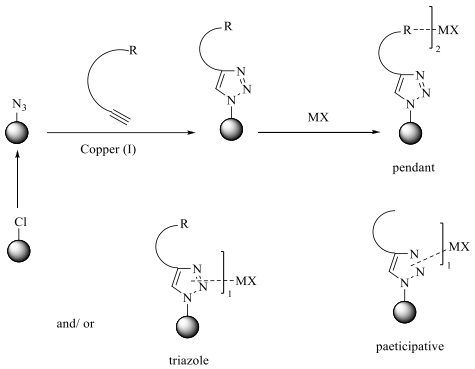

Fig. 10, click reaction for the extraction of heavy metals such (Cd, Cu, Ni, Cr, and Pb)

Moreover, similar reported articles investigated the PVC-triazoles modification [40,78,82,86–88]. It was reported that (PVC-0.75CX[4]) was synthesized viaclick reaction between dialkylnyl- p-tert-butylcalix[4]arene and p oly(vinylchloride) azide (PVC-N3). dialkylnyl- p-tert-butylcalix[4]arene with triazole groups grafted onto PVC polymer (figure 11). (PVC-0.75CX[4]) has an excellent extraction capability for chromium ions from aqueous solutions. The ability of calixarene-based covalent polymers to extract chromium (VI) ions from aqueous solution regarding to the soft cavity, the presence of π triazole rings and hydrogen bonds. Cr(VI) ion sorption capacity is of 95% at pH = 3 [85].

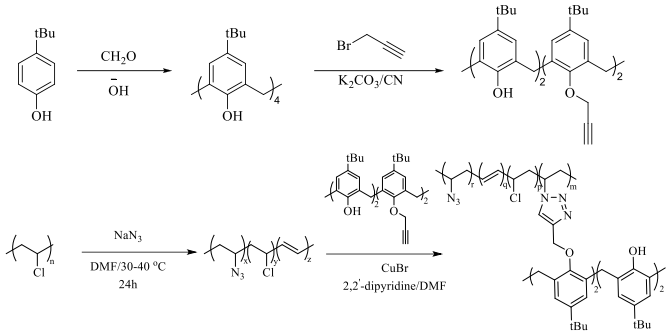

Fig. 11, Chemical modification of PVC using triazoles *via* click reaction

g) Oxazoles derivatives:

Chemical modification of PVC with oxazoles was carried out by Abdelaal and coauthors to be metal uptaken from the aqueous solution [88]. Figure 12 shows the reaction of PVC with 1,4-diaminobenzene (Phenylenediamine) and complete to oxazole and triazoles.

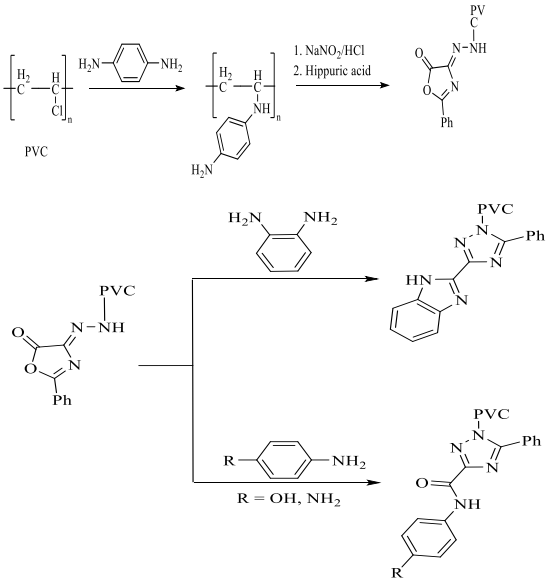

Fig. 12, modification of PVC with oxazoles and triazoles

h) Thiazole derivatives:

PVC modification was carried out *via* amino thiazole nucleophilic substitution [89–92]. The thiazole ring substitution is considered a main component of the PVC matrix. Generally, thiazole plays a role in drug development e.g. allergy [93] and fungal [94] infections. In addition to their occurrence penicillin antibiotics [46,95]. The modified PVC adduct was further utilized to produce the corresponding Ag and Cu nanocomposites (3% metal nanoparticles ratio) (Figure 13). The antibacterial study for both types Gram +ve and Gram -ve showed promising results.

The PVC/AgNPs had a significant increase in antimicrobial efficiency (Table 4). This is due to Ag nanoparticles' ability in cell wall penetration to cause damage by binding to the existing thiol groups in the enzyme of microorganisms leading to final deactivation [96].

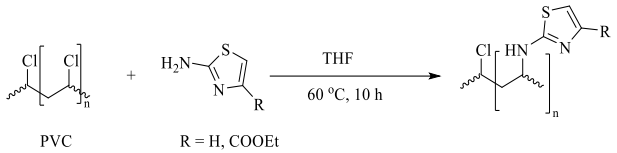

Fig. 13, modification of the PVC using thiazole derivatives

Table 4: Antibacterial behaviors of thiazole-functionalized PVC and thiazole-functionalized PVC nanocomposites with AgNPs or CuNPs. [16]

Sample	Gram +ve bacteria inhibition zone (mm)			Gram –ve bacteria inhibition zone (mm)		
	B. subtilis	*S. aureus*	*S. faecalis*	*E. coli*	*N. gonorrhoae*	*P. aeruginosa*
DMSO	0	0	0	0	0	0
Ampicillin	20 ± 0.21	22 ± 0.25	20 ± 0.22	17±0.18	18 ± 0.20	17 ± 0.23
PVC	6 ± 0.18	5 ± 0.29	5 ± 0.22	4± 0.31	6 ± 0.18	5 ± 0.21
Thz-PVC	10 ± 0.32	12 ± 0.17	9 ± 0.14	12±0.27	9 ± 0.15	10 ± 0.15
Thz-PVC/AgNPs	14 ± 0.19	15 ± 0.20	13 ± 0.06	14±0.29	13 ± 0.28	15 ± 0.26
Thz-PVC/CuNPs	12 ± 0.16	13 ± 0.07	12 ± 0.26	13±0.12	11 ± 0.23	12 ± 0.23
Thz ester-PVC	14 ± 0.18	16 ± 0.13	14 ± 0.22	16±0.18	13 ± 0.15	14 ± 0.23
Thz-ester-PVC/AgNPs	19 ± 0.22	20 ± 0.17	18 ± 0.20	21±0.23	17 ± 0.19	16 ± 0.11
Thz-ester-PVC/CuNPs	16 ± 0.21	17 ± 0.26	16 ± 0.13	18±0.15	15 ± 0.24	14 ± 0.27

i) Pyrimidine derivatives:

Pyrimidine derivatives have been used to modify PVC. The photostability and bioactivity were investigated. The results indicated a stabilization rate [46,98]. The *p*-chloro adduct has shown the highest photostability in comparison to the other modified PVCs. In addition, the same compound has shown promising antibacterial results as tested against *Staphylococcus aureus* (Gram-positive) bacteria [46,98]. (Figure 14) showed the reaction of pyridopyrimidine and adenine with PVC to the modifier which gives high photostability and high bioactivity. The antibacterial study for both types Gram +ve and Gram -ve showed promising results. The modified PVC showed a significant increase in antimicrobial efficiency (Table 5).

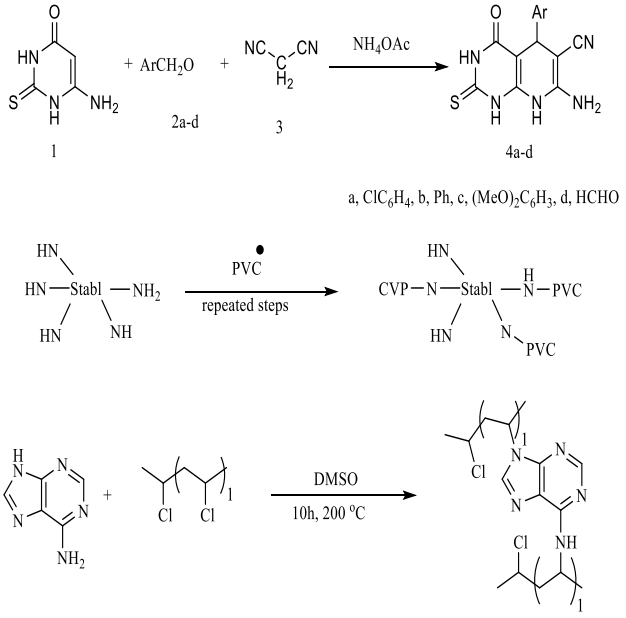

Fig. 14, Synthesis of pyridopyrimidine derivatives, and PVC modification

Table 5. The bioactivity activity of PVC in the presence of some pyridopyrimidine derivatives. [109]

Sample	*S. aureus*	*P. aeruginosa*	*A. Flavus*
Ciprofloxacin (100 µg/ml)	24	36	-
Fluconazole (20 µg/ml)	-	-	12
DMSO	-	-	-
4a	18	16	30
4b	12	14	19
PVC/**4a**	15	18	12
PVC/**4b**	11	12	10

j) Chitosan:

Antimicrobial polymers represent a great challenge to many researchers. Therefore, the proposed modification for PVC is recommended to be applied in medical devices with anti-infective properties. Chitosan has been implemented to present a dual effect of a spacer in the PVC matrix in addition to the biological compatible nucleophile. This modification has been carried out in a multistep route. Primarily, the Bromo-acetylation of the chitosan [99–101] was carried out. This was followed by the addition of benzothiazole to PVC. Finally, the silver nanoparticles have been loaded into the new PVC [102]. Functionalized chitosan (Cs1, Cs2, Cs3) have been synthesized, respectively, as shown in Figure 15, Cs1 is the Chitosan PVC, Cs2 is chitosan PVc

with silver nanoparticles (Ag NPs), and Cs3 chitosan-PVC with Ag/TiO_2 NPs [103]. The biomedical applications against *E. aureus* (gram-positive) and *E. coli* (gram-negative) has been tested. The results indicate the effectiveness of Cs2 against *E. coli*[104], the rupture of the bacterial cell walls, especially Cs2 is illustrated. It is suggested that further investigations would be an added value in the field of biochemical applications.

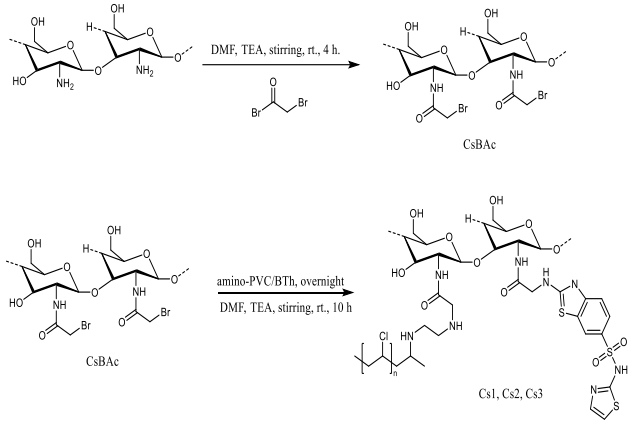

Fig. 15, Synthesis of benzothiazole -functionalized Cs-PVC (Cs1), benzothiazole -functionalized Cs-PVC containing AgNPs (Cs2), benzothiazole -functionalized Cs-PVC containing Ag/TiO_2-NPs (Cs3)

Conclusion

In this review, we give some examples of the PVC modification with different synthesized heterocyclic derivatives and polysaccharides which give high stability and bioactivity to PVC. Moreover, some of the modifiers such as oxazole, imidazole, and triazole derivatives give high activity for heavy metals uptaken from the aqueous solution. The trend of interacting (PVC) with heterocyclic derivatives relies on the utility of nucleophilic substitution reactions. Functionalizing heterocyclic compounds can produce electrophilic and nucleophilic and electrophilic cores. Hence, this may improve the diverse applications of these organic molecules in the modification of PVC. Different reactions with changeable conditions provide a versatile and safe transformation to PVC by forming new covalent bonds. The modified PVC has various applications in the medical field due to the biocompatibility of the surface after modification such as pyrazole, imidazole, pyrimidine, oxazole, and chitosan derivatives. The originated derivatives of heterocyclic compounds are able to contribute to manufacturing, pharmaceutics, and biotechnology. The kind of additives with different loadings in PVC has a role in affecting its photodegradation and phytostabilization as well. Efficient additives are able to minimize the photolysis rate of PVC. The heterocyclic units bound to PVC chains enhance the photostability of PVC. Moreover, the improvement in the mechanical properties in terms of stretching and ductile strength is achieved by maintaining the translucency of PVC. The additives can extend by applying PVC widely with longer durability upon being compared with other plastic materials.

CONFLICT OF INTEREST

The author declares no conflict of interest, financial or otherwise.

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