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RESEARCH ARTICLE

TRICLOSAN MODIFIED STABLE MELAMINE RESIN AS LONG-LASTING ANTIMICROBIAL AGENT.

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Abstract

Antimicrobial property of triclosan is diminished due to leaching out from the substrate during washing. A non-leaching permanent antimicrobial finish for cotton fabrics in textile was prepared by condensation reaction of hexa-methylolated melamine with triclosan and further etherification with ethylene glycol to stabilize the resin. Resin was characterized for molecular structure by FTIR and molecular weight by viscometric method. Bacterial strains were used to test inhibition of triclosan modified melamine formaldehyde resin in comparison with conventional leachable triclosan. Triclosan modified melamine formaldehyde resin showed dynamic inhibitory action against bacteria such as *Escherichia coli*, *Staphylococcus aureous.* A rapid decrease was observed in bacterial count within 48 hours. It was also observed that resin treatment had significant effect on fabric strength, crease recovery angle, stiffness and whiteness index due to resin crosslinking reaction. Resin coating on fabric showed very good wash fastness with long lasting antimicrobial properties in comparison with triclosan treated fabric. All these properties were achieved due to crosslinking behavior of resin with cellulosic fabric.

Introduction:-

The compounds having antimicrobial functionality belong to organic compounds and metals. Organic compounds include triclosan, polybiguanideschitosan (naturally derived antimicrobial)[1-3],quaternary ammonium compounds and N-halamines. The metals include such as silver and zinc.

The polymer having antibiotic repeated units or antibiotic pendant groups in the chemical structure are known to be antimicrobial polymers.Addition of polymer material and antibiotic should not allow as a production method of antimicrobial polymers. In 1970s intrinsically modified antimicrobial polymers had been synthesized and their application was studied[4, 5].They are proposed to be cost-effective for making non-leaching antimicrobial surfaces.

Resorcinol based resins due to thermal stability and cost effectiveness, show antimicrobial properties, have wide range of application as polymer matrix and structural application in aerospace[6].Antimicrobial properties can be produced by the combination of Melamine formaldehyde with polyhexamethylenebiguanide (PHMB) on the outer layer of high pressure laminates[7].Due to wide range of antimicrobial properties of metal nanoparticles they are embedded in polymer matrices.[8]

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There is a growing demand for non-leaching antimicrobial surfaces in textile materials. Its mechanism is not fully understood due to limited literature as compared to traditional antimicrobial concept. Cationic antibiotic having low molecular weight act as a target site on cytoplasmic membrane of bacterial cells. The following routes had been assumed: (i) adsorption onto the negatively charged surface of bacterial cell, (ii) cell wall diffusion, (iii) cytoplasmic membrane binding, (iv) interruption of cytoplasmic membrane, (v) constituent and K^+ ion of cytoplasmic membrane, (vi) cell death. Due to higher charge density of polycation their adsorption onto negatively charge cell surface is higher than that of cationic molecules or monomers. Polycation form linkage easily with cytoplasmic membrane due to larger negative charges as compared to cations of low molecular weight.Thus, membrane interruption and release of potassium ions and cytoplasmic ingredient would be enhanced[9].

Resins are cross-linked polymer which are thermosetting[10, 11]. Thermoplastic can be extruded and moulded many time while thermosets processed only once at high temperature to make polymer hard and solid after the cross linking reaction. These polymers can't be softened on heating and are insoluble upon curing[12-14].In surface coating minor types of thermosetting resins are used such as unsaturated polyester resins, epoxy resins , urethane foams and alkyds[15]. These materials have drawback that during and after cure they have poor weather ability and also release formaldehyde. While in melamine moulding this is not the case and have better weather ability [16, 17].

In Germany melamine formaldehyde was discovered earlier but can't be processed due to non-commercial development. Reaction of amines or amides with formaldehyde was produced by the coined of amino plastic which cover range of resinous polymer[18, 19]. Urethane formaldehyde formation is similar to that of melamine formation. A complete hydroxyl methylation of melamine is occurred[20]due to addition of formaldehyde to amino group of melamine, which is faster than the urethane formaldehyde formation[21]. The reaction is also feasible in alkaline and neutral condition in addition to acidic condition[22, 23]. But in urethane formaldehyde the formation is only in acidic condition. Products are less susceptible to release of formaldehyde by melamine formaldehyde resins than urethane formaldehyde resins.

Triclosan is an antimicrobial agent with wide spectrum. It is active against bacterial species. Inhibition of bacterial growth by triclosan is processed by hindering lipid biosynthesis[24].There are numerous application , triclosan can be used as a stabilizer and bacteriostat, in the preparations of detergents and cosmetics, as well as disinfectant and antiseptic in medications[25].Washing resistance of triclosan is very poor and leached out during washing the material. Triclosan antimicrobial property is diminished after one or two washing cycles. In this study washing resistance of triclosan is improved by condensing it with stabilized etherified melamine resin.Characterized polymer is insoluble in ethanol, toluene and benzene and readily soluble in DMSO and acetone[26].

Experimental Procedure:-

Chemicals and apparatus:-

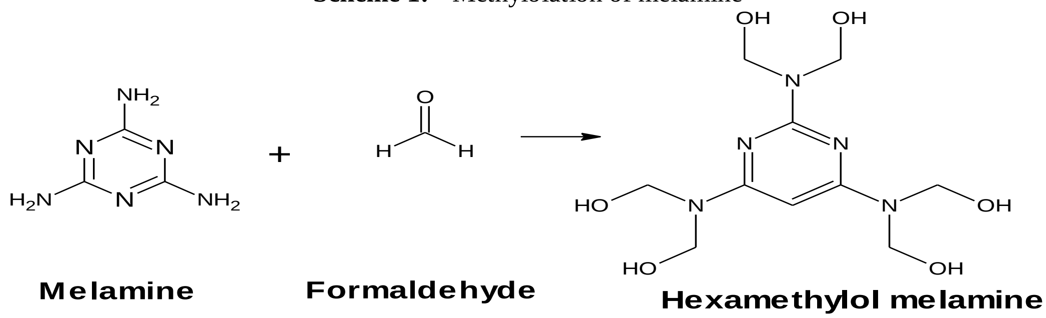
Melamine (99.8% purity, powder) was taken from Royal DSM Netherland and was processed as received. Technical grade formalin (370% strength) and commercial triclosan were used without purification.
Viscosity was determined by Brookfield viscometer LV DVE 230 USA at 25°C. FT-IR spectrum of the resins was recorded by Agilent Technology Carry 630 FTIR.

Preparation of triclosan modified etherified melamine resin:-

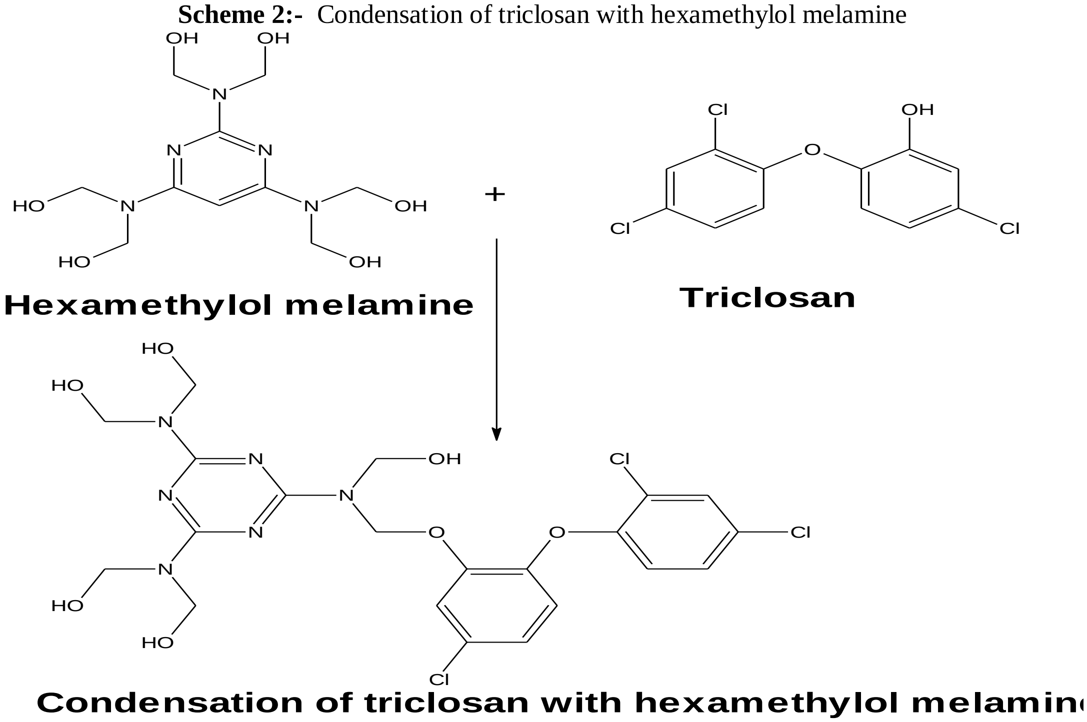
In a three necked round bottom flask assembled with condenser, thermometer and stirrer, 243.48g of formalin (3.0 moles, 37% strength) and 63g of melamine (0.5 moles) were taken. pH of reaction mixture was adjusted to 8-8.5 by 0.1 N sodium hydroxide. Heated the reaction mixture upto 85°C and mixed for half hour. Added 62.07g of triclosan (0.5 mole) and mixed for one hour. Added 62.07g of monoethylene glycol (1.0 mole) and mixed for one hour. Cool to 25°C and added 10g of sulfuric acid (36 N, 99% strength) controlling temperature not exceeding above 30°C to adjust pH up to 2.

During acidic conditions, ether linkages were developed between the monomer molecules. After half hour mixing at low pH of 2, reaction mixture was neutralized with 50% strength sodium hydroxide solution. Reaction mixture was heated to 85°C and mixed for two hours to stabilize the polymer. The reaction mixture was cooled to room temperature. The solid contents of the resin were about 70±1%. Schematic route for the synthesis of triclosan modified antimicrobial melamine formaldehyde resin as shown in scheme 1-4.

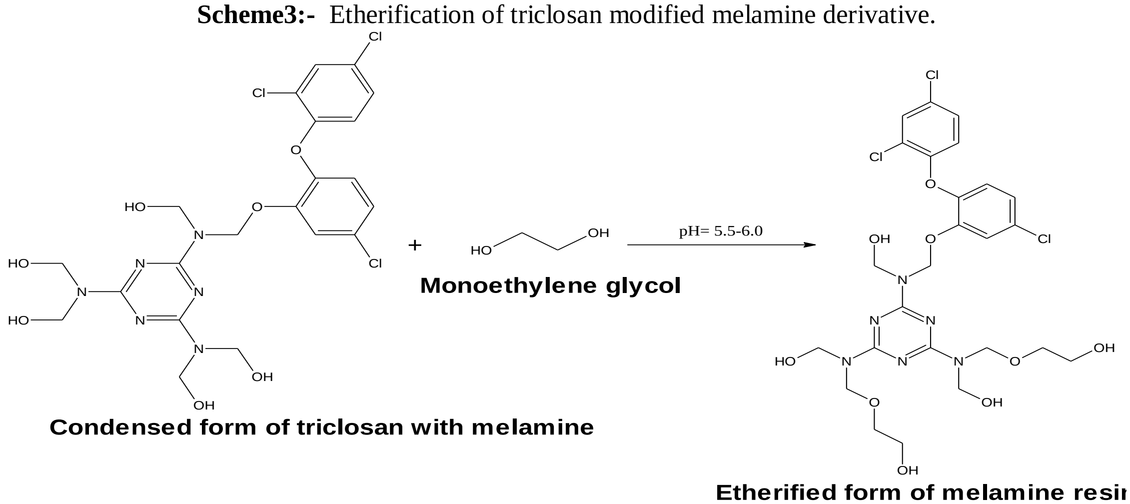
Scheme 1:- Methylolation of melamine



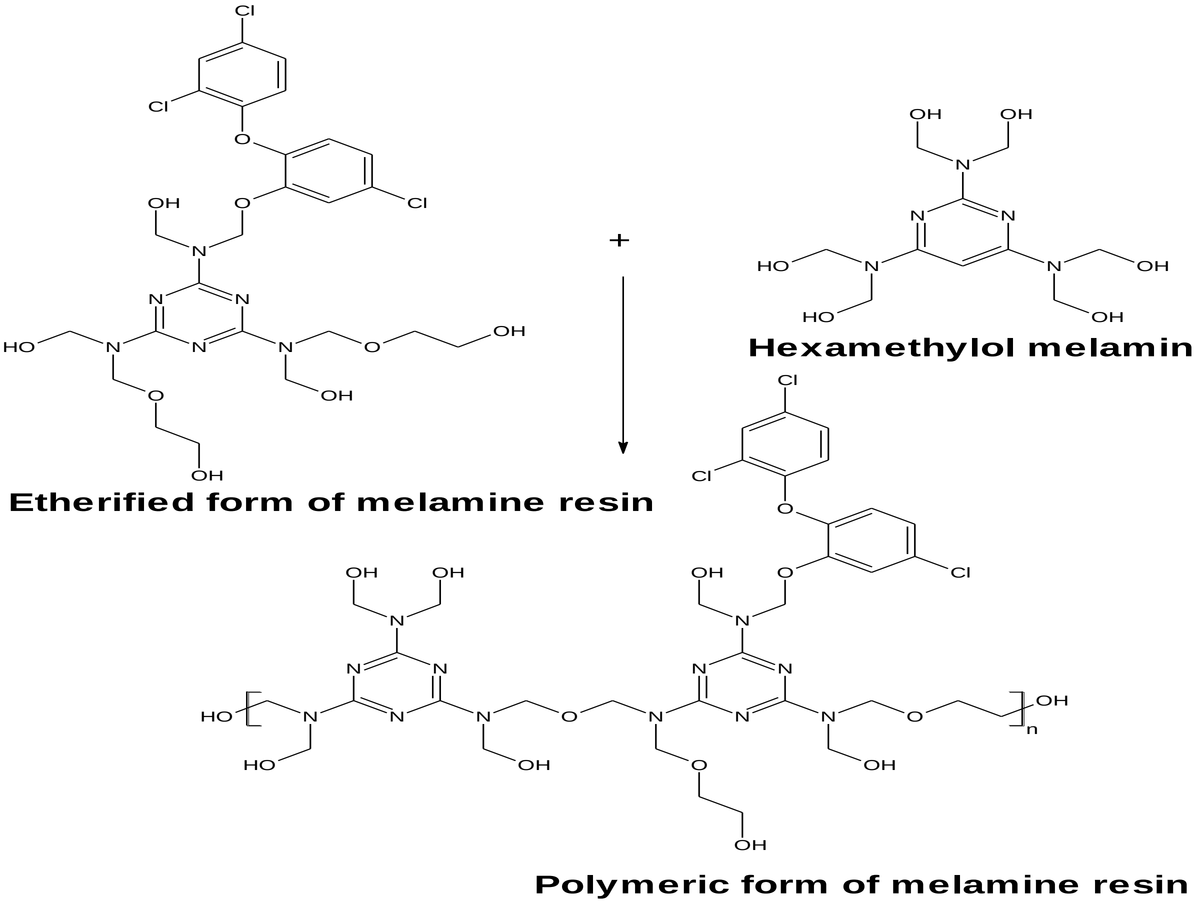
Scheme 2:- Condensation of triclosan with hexamethylol melamine



Scheme3:- Etherification of triclosan modified melamine derivative.



Scheme 4:- Resinification



Characterization Of Resin:-

Estimation of solid Content:-

Solid content of modified melamine resin was determined by weighing known quantity of the material in a petri dish and drying at 105±1^{o}C for one hour as per standard procedure[27]. Solid contents of the resin were calculated on dried weight basis and was found 70±1%.

Viscosity Determination:-

Viscosity of liquid modified melamine resin was determined by Brookfield viscometer LV DVE 230 at temperature of 25^{o}C and was found250 cp.

Viscosity average molecular weight:-

The molecular weights of etherified synthetic resin was measured by using viscometery technique. Following procedure was used to determine molecular weight of the resin. Flow time was measured by Cannon Ubbelohde viscometer of each dilution of resin solution. All the measurements were performed at constant temperature of 25^{o}C ± 0.1^{o}C. Flow time for the water (solvent) was determined by taking measured volume of water in the viscometer. Three consecutive readings of flow time were determined and calculated average value for achieving maximum accuracy in the results. The viscometer was rinsed, dried. 10 ml of resinous solution at concentration of 0.08 g/10 ml of water was taken and flow time measured by three concordant readings. The solution was further diluted to concentration of 0.07g/10 ml and flow time was determined. Similarly, concentration of 0.06g/10 ml, 0.05g /10 ml, 0.04g /10 ml dilutions were prepared and their flow time was determined. The respective viscosity average molecular weight was determined.

Efflux time of the pure water is determined by flow time measurement. The relative viscosity is the viscosity of the polymer solution to the viscosity of the pure solvent. This is done by taking the efflux time of the polymer solution at a given concentration (we call this t) and dividing it by ‘efflux time of solvent as explained in Equation 1

$$\text{Relative viscosity} = \frac{\text{Efflux time of solution}}{\text{Efflux time of solvent}}$$

$$\eta_r = \frac{t}{t_o} \text{ Equation 1}$$

The specific viscosity was measured by taking the difference in the efflux times of the solution and the pure solvent. In other words, the efflux time of the pure solvent 't_o' was subtracted from the efflux time of the solution as explained in Equation 2:

$$\text{Specific viscosity} = \frac{\text{Efflux time of solution} - \text{Efflux time of solvent}}{\text{Efflux time of solvent}}$$

$$\eta_{sp} = \frac{t - t_o}{t_o} \text{Equation 2}$$

The intrinsic viscosity of the sample solution was determined by extrapolating graph between the concentration and Δ /c^2. This plot gives the value of 1/2 $[\eta]^2$ at zero intercept.

From the flow time, relative velocity (η_r), specific viscosity (ηsp) and intrinsic viscosity were calculated for the etherified resin. Then, the molecular weight of the etherified resin was determined by using Mark Houwink equation 3.

$$[\eta] = K[M]^a \qquad \text{Equation 3}$$

Where, [η] is intrinsic viscosity, 'M' is the molecular mass of the synthetic resin, 'K' is the characteristic of the synthetic resin and solvent and 'a' is a constant and a function of the shape of the synthetic resin structure in the solution.

For melamine formaldehyde type of resins, the value of 'a' is usually 0.6 and 'K' is 0.076.

Cotton Fabrics:-

Cloth, plain weave, 102 $g/m^2$was purchased from textile factory which was desized and scoured. Before treatment the fabric material was washed with solution of Na_2CO_3(3 g/L) and kierlon (1 g/L, non-ionic surfactant) at 100 ^oC for 30 min. After washing, the fabric was dried at room temperature.

Preparation of antimicrobial cotton Fabrics:-

To attain a wet pick up of 100 % (with respect to weight of fabrics) laboratory padderwas used to saturate cotton fabric with triclosan modified melamine resinsolution (2%). For complete reaction between cotton fabric and functionalized melamine resin the polycondensation reaction was carried out. By drying and curing at 80 ^oC for 30 min. The same procedure was adopted for the application of triclosan alcoholic solution (2%).

Washing Procedure:-

An AATCC Atlas Launder-O-Meter standard instrument was used for evaluating washing fastness of antimicrobial treatment. The instrument has highest efficiency because five times home washing is equal to one wash with this instrument (ISO 105-C01:1989(E) standard method). 5 g/L standard reagent of SDC was added and mix for the duration of 30 min then the material was rinsed with distilled water and dried.

Antimicrobial Activity:-

Branched polymers, including star-shaped, dendritic and hyper-branched architectures, have attracted attention as potential antimicrobial agents[28].AATCC 100 standard method was used to evaluate the antimicrobial activity[29].Bacterial strains like *Escherichia coli*(Gram negative) and *Staphylococcus*(Gram positive) were used as testing bacterial strains. Untreated control, triclosan treated and functionalized melamine resin treated swatches of cotton fabrics were inoculated with 1-2 × 10^5 CFU (each experiment was performed for separate bacterial strain) of bacteria and 1.0 ml of broth culture of nutrients were incubated for 2 h at 37 ^{o} C, then shaked in neutral solution for separating bacteria from cotton swatches. Distilled water is used for dilution. The nutrient agar was used for suspension plating and then incubated at 37 ^oC for 2 h. After incubation sample was washed with phosphate buffer saline (PBS pH 7.2)[30]. Percentage of bacterial reduction is calculated by count down the number of bacterial forming units (CFU) by the following equation 4.

$$\%BR = \frac{B-A}{B} \times 100 \qquad \text{Equation 4}$$

A= CFU value recovered from the resin treated or triclosan treated cotton fabric which were inoculated.

B=CFU value recovered from the control cotton fabric which were inoculated.

Textile strength, stiffness, crease recovery angle, and whiteness Index:-
ASTM D 5035 test method was carried out by UTR-4 (Uster) for measuring the tensile strength of control, triclosan treated and resin treated cotton fabrics, whereas ASTM D 1388-96 test method was performed by stiffness tester (SDL international) to determine the bonding strength. The AATCC-066 method was used for evaluation of crease recovery angle of the antimicrobial cotton fabrics. Data color550$_{TM}$ was used to evaluate the whiteness index (WI).

Result and Discussion:-

Viscosity average molecular weight:-

The molecular weight of triclosan modified melamine resin was determined by viscometric method. Δ/c^2 for the triclosan modified melamine resin sample was calculated at different concentrations from relative viscosity, specific viscosity and given in Table 1. The intrinsic viscosity of the triclosan modified melamine resin sample was determined by extrapolating graph between the concentration and Δ/c^2 as shown in figure 1. This plot gives the value of 1/2 $[\eta]^2$ at zero concentration which was 2463.46. Thus, intrinsic viscosity was calculated to be 70.192. From the intrinsic viscosity, molecular weight of the resin was calculated by applying Mark Houwink equation and calculated to be 57590.392.

Table 1:- Dependence of the flow time, relative viscosity, specific viscosity, Δ = **ŋsp – lnŋr**and Δ/c^2 on the concentration of the triclosan modified melamine resin.

Conc.g/ml	Flow time seconds	ŋr	ŋsp	lnŋr	Δ = ŋsp - lnŋr	Δ/c2
0.008	171.1	1.443	0.439	0.367	0.072	1127.475
0.007	166.9	1.408	0.404	0.342	0.061	1256.505
0.006	160.4	1.353	0.349	0.302	0.046	1290.016
0.005	155.2	1.309	0.305	0.269	0.035	1420.592
0.004	152.8	1.289	0.285	0.254	0.030	1927.898

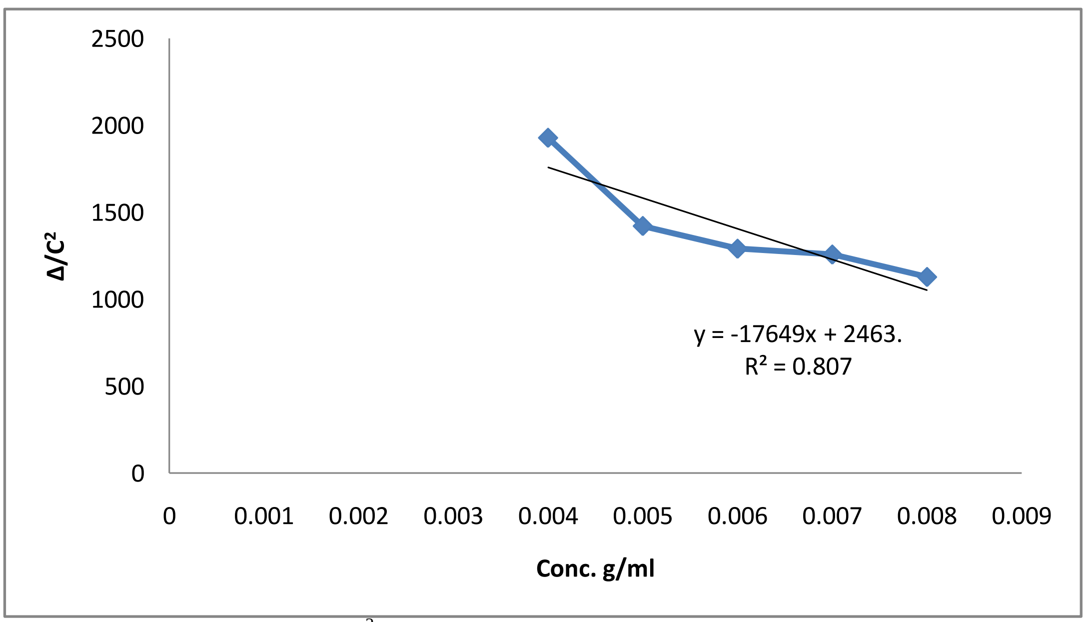


Fig.1:- Dependence of Δ/c^2 on the concentration of triclosan modified melamine resin.

Structure Elucidation:-

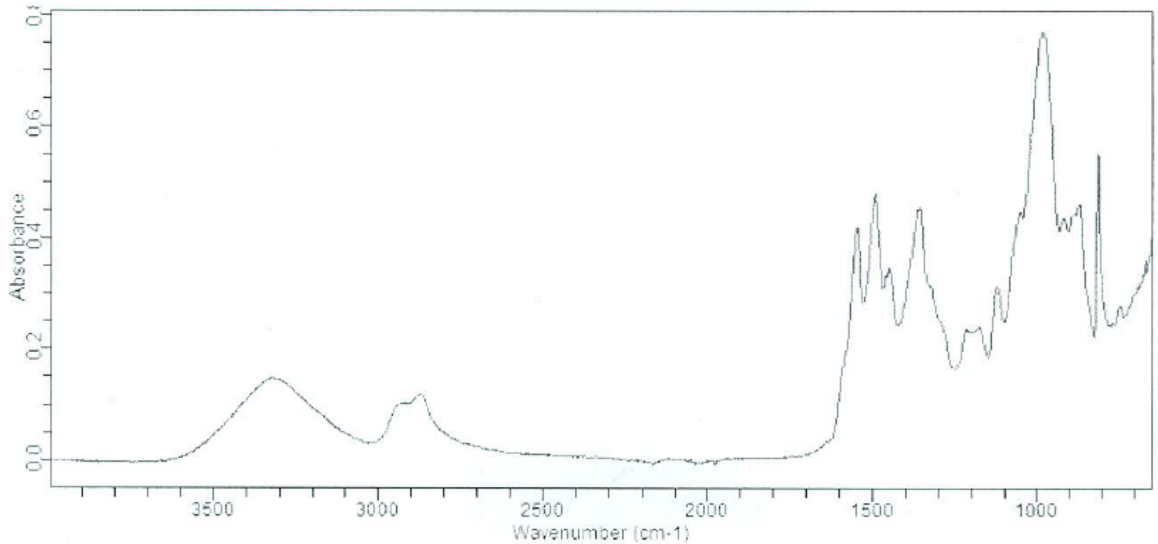


Fig. 2:- Spectra ofhexa-methylolated melamine formaldehyde resin

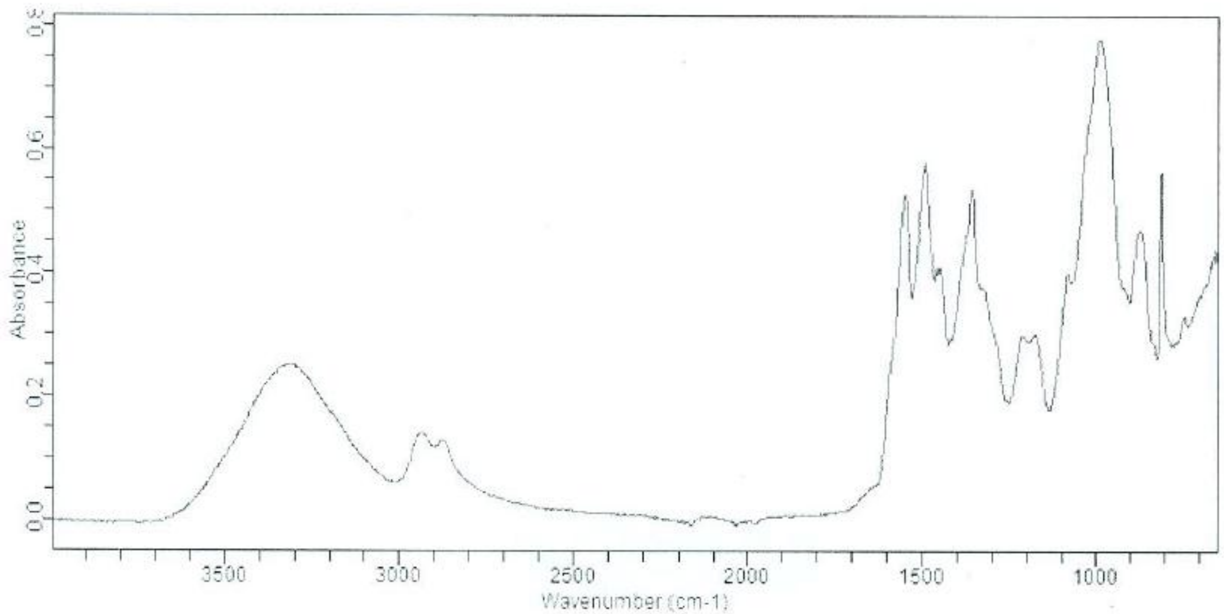


Fig. 3. Spectra of triclosan modified melamine formaldehyde resin.

Fig 2 and 3shows spectra of hexa-methylol melamine formaldehyde resin and triclosan modified melamine resin. The spectra were measured in spectral range from 4000 to 400 $cm^{-1}$by Agilent Technology Carry 630 FTIR. In fig (2,3) the broad band at 2875 to 2925 shows aromatic C-H stretching. Benzene ring of aromatic compounds is observed at 1485 to 1615 cm^{-1}. In figure 1 there is no absorption signal at 900 cm^{-1}, when triclosan is condensed with melamine formaldehyde resin a sharp peak is observed at 900cm^{-1} indicating C-Cl linkage. The broad band at 3400cm^{-1} to 3500cm^{-1} in both spectra represents hydroxyl and amino functionality in the synthetic resin. Aromatic amine peaks is observed at 1180 to 1280 cm^{-1}.

Antimicrobial Activity:-

Antimicrobial activity of untreated cotton fabric as control and both cotton fabrics antimicrobially treated with triclosan and functionalized melamine resin were determined against bacterial strains *E. Coli (Gram negative)* and *S. aureus (Gram positive)* according to modified AATCC 100. After the period of 24 h on the cloth treated with functionalized melamine resin both strains showed very high % bacterial reduction.*E. Coli* showed 89 % in decrease and *S. aureous* showed 94 % in decrease. After the period of 48 h both strains vanished up to 98 % of viable colonies.Result also indicates that untreated cotton fabric showed no bacterial reduction. Similarly after the period of 24 h on the cloth treated with triclosan both strains shows very high % bacterial reduction of *E. coli* up to 80 % and *S. aureous* up to 82 %. After the period of 48 h both strains vanished up to 90% of viable colonies. Results are given in Table 2. Untreated cotton fabric showed an increasing bacterial recovering, which as a result indicating cotton fabric was most suitable substrate for both the growth of strains.

Antibacterial activity relationship between triclosanand antimicrobially functionalized melamine resinwas also evaluated.

Table 2:- Percentage bacterial reduction antibacterial properties of treated cotton fabric against *S. aureus* and *E. coli*

	E. coli				*S. aureus*		
Time	24 h	48 h			24 h	48 h	
Control	0	0			0	0	
Functionalized melamine resin	89	98			94	97.5	
Triclosan	80	90			82	91	

Major problem faced in the antimicrobial treatment was the washing fastness. Use of silver nanoparticles was most popular method for rendering textile material. During the process silver nanoparticles leached from the surface, which as a result after few washing surface, becomes inactive and antimicrobial efficiency is reduced. An excellent method was developed by inserting AgCl in silica matrix but this method had also limitation as well because antimicrobial property was lost after several washing[29]. An AATCC Atlas Launder-O-Meter was used for to determine the washing characteristics of currently used method. It is worth noticed that five washing at home correspond to one wash with this instrument (ISO 105-C01:1989(E) standard method).

Antimicrobial activity of both cotton fabrics antimicrobially treated with triclosan and functionalized melamine resin after 15 washing cycles were determined against bacterial strains *E. Coli (Gram negative)* and *S. aureus (Gram positive)* according to modified AATCC 100.After the period of 24 h on the cloth treated with functionalized melamine resin both strains showed very high % viable colonies. *E. coli*showed up to 87 % in decrease and *S. aureous* showed up to 88 % in decrease. After the period of 48 h *E. coli* strains vanished up to 95% and*S. aureous* vanished up to 93 %of viable colonies. Similarly after the period of 24 h on the cloth treating with triclosan shows very low % viable colonies E. *coli* showed 25% in decrease and *S. aureous* showed 29 % in decrease. After the period of 48 h *E. Coli* strains vanished up to 30% and *S. aureous* vanished up to 34 % of viable colonies. Results are given in Table 3. Results indicate that functionalized melamine resin is reactive towards fabric and give non-leachable wash resistance antimicrobial performance.

Table 3:- Percentage viable colonies antibacterial properties of treated cotton fabric after 15 washing cycles against *S. aureus* and *E. coli*

	E. coli				*S. aureus*		
Time	24 h	48 h			24 h	48 h	
Functionalized melamine resin	87	95			88	93	
Triclosan	25	30			29	34	

Further, functionalized melamine resin was evaluated at different concentrations of 0.1, 0.2, 0.3, 0.4 and 0.5% to determine the optimum dosage concentration, as given in Table 4. Result indicated that an excellent antibacterial property was observed at 0.4 %concentration as demonstrated Figure 4.

Table 4:- Effect of different concentrations of triclosan modified melamine resin.

Concentration	***E.coli***	***S. aureous***
0.1	75	80
0.2	80	85
0.3	89	91
0.4	94	98
0.5	98	98

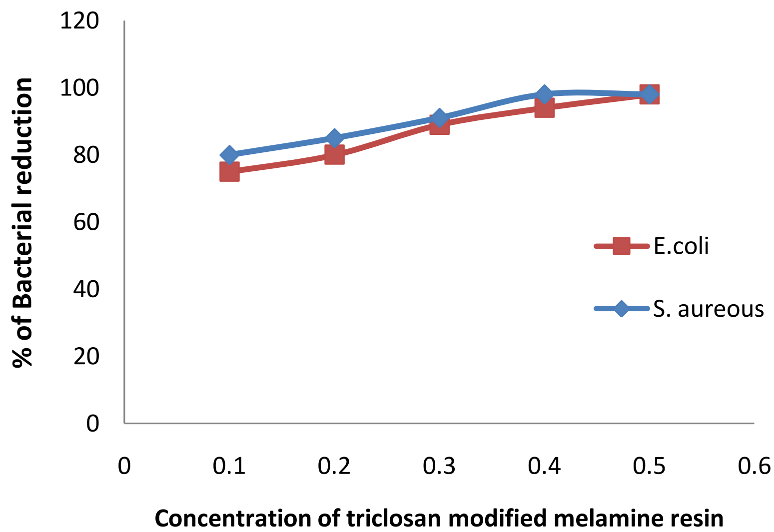


Fig. 4:- Relationship between triclosan modified melamine resin concentrations, and antibacterial activity

In order to observe the effect of different concentration of triclosan modified melamine resin, tensile strength and bending length of treated cotton fabric has been measured. The results for tensile strength are given in Table 5, whichshowed positive result of tensile strength of cotton fabric after treating with varying concentration of triclosan modified melamine resin and results are also displayed graphicallyin Figure 5. Results indicate that increase in tensile strength was due to more crosslinking of resin with cellulosic fabric

Table 5:- Effect of different concentrations of triclosan modified melamine resin on fabric tensile strength

Concentration	Tensile strength (N/m^2)
0.1	2500
0.2	2690
0.3	2750
0.4	2930
0.5	3040

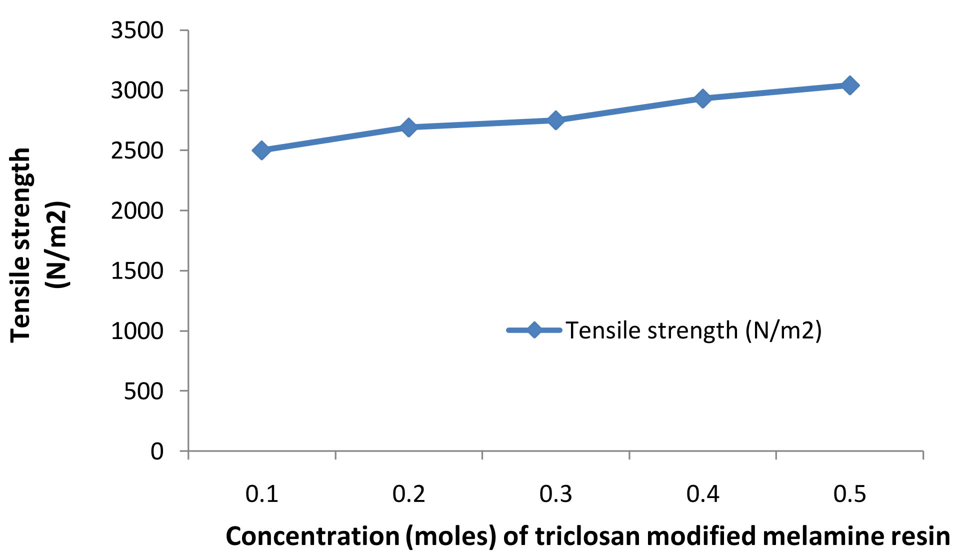


Fig. 5:- Effect of triclosan modified melamine resin concentration on fabric tensile strength.

It was also noticed that increase in tensile strength correlates with increase in bending strength as data is given in Table 6 and graphically demonstrated in Figure 6.

Table 6:- Effect of different concentrations of triclosan modified melamine resin on fabric bending strength

Concentration	Bending length (cm)
0.1	3.5
0.2	3.9
0.3	4.2
0.4	4.3
0.5	4.9

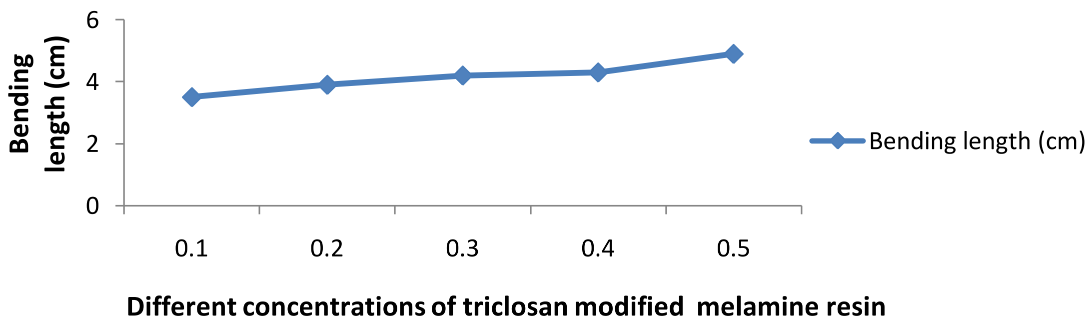


Fig. 6:- Effect of triclosan modified melamine resin concentrations on fabric bending length.

Effect of different concentration of triclosan modified melamine resin was also evaluated against wrinkle recovery angle (WRA). Results are given in Table 7 and 8, showing that there was a significant increase in WRA along warp and weft by increasing concentration of functionalized melamine resin that caused increased in stiffness, results also demonstrated graphically in Figure 7 and 8.

Table 7:- Effect of different concentrations of triclosan modified melamine resin on wrinkle recovery angle (warp)

Concentration	Wrinkle Recovery Angle (warp)
0.1	50
0.2	59
0.3	65
0.4	74
0.5	79

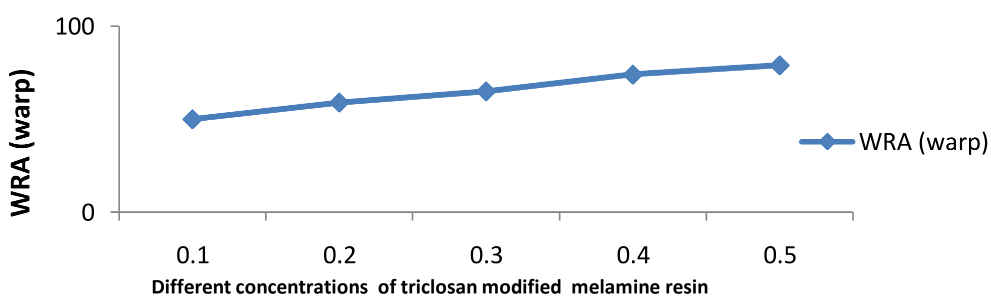


Fig. 7. Effect of triclosan modified melamine resin concentrations on wrinkle recovery angle (warp).

Table 8. Effect of different concentrations of triclosan modified melamine resin on wrinkle recovery angle (weft)

Concentration	Wrinkle Recovery Angle (weft)
0.1	55
0.2	61
0.3	69
0.4	78
0.5	82

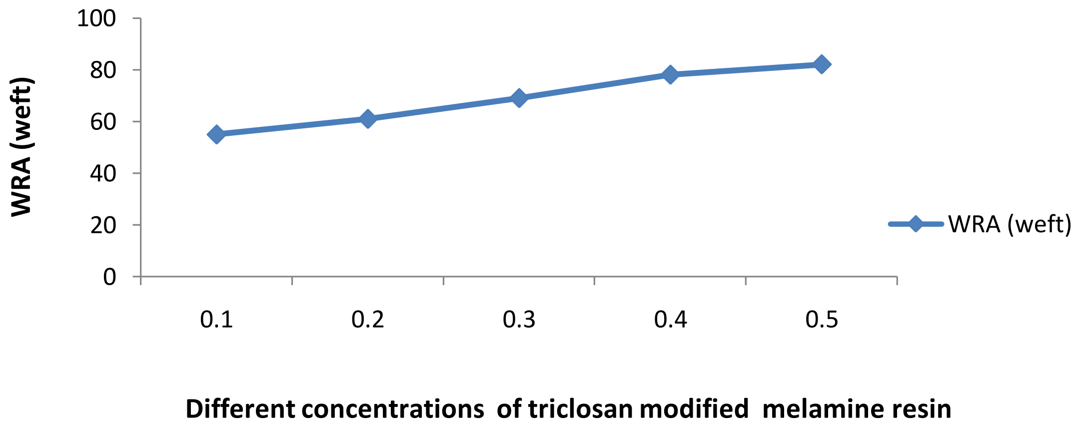


Fig. 8:- Effect of triclosan modified melamine resin concentrations on wrinkle recovery angle (weft).

By varying concentrations, a positive effect was also observed in whiteness index as data is given in Table 9 and graphically demonstrated in Figure 9.

Table 9:- Effect of concentration of triclosan modified melamine resin on degree of whiteness

Concentration	Degree of whiteness
0.1	70
0.2	79
0.3	85
0.4	89
0.5	92

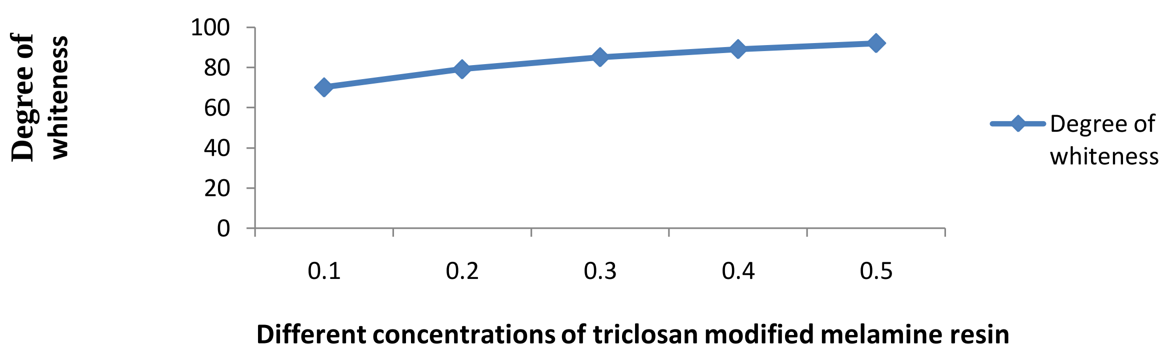


Fig. 9:- Effect of triclosan modified melamine resins concentrations on degree of whiteness.

Positive trends in tensile strength, stiffness, wrinkle recovery angle and whiteness index byincreasing concentration of functionalized melamine resin is due to availability of more reactive group which form stable bond with the cellulosic fabric and give long lasting effect.

Conclusion:-

In the present work simple non-leached and significant antimicrobial functionalized melamine resin for cotton fabric has been developed, which was further stabilized by etherification. Results indicate that treatment with functionalized melamine resin gave excellent wash fastness and long-lasting antimicrobial activity. However it has positive effects on the tensile strength, whiteness index, wrinkle recovery angle and bending strength. Antimicrobial property achieved by novelly developed functionalized melamine resin on textile fabric could be easily adapted on various other substrates like leather and coating industries.

References:-

1. Gao Y,CranstonR, (2008): Text. Res. J,Recent advances in antimicrobial treatments of textiles, 60-72 78(1).
2. Purwar RM, Joshi, (2004): Aatcc. Rev,Recent Developments in Antimicrobial Finishing of Textiles--A Review, 4(3).
3. Simoncic BB, Tomsic, (2010): Text. Res. J,Structures of novel antimicrobial agents for textiles-A review, 1721-1737 80
4. Donaruma LG, (1975): Prog. Polym. Sci,Synthetic biologically active polymers, 1-25 4
5. Ackart WR, Camp W, Wheelwright J, Byck, (1975): J. Biomed. Mater. Res,Antimicrobial polymers, 55-68 9(1).
6. Chandra, MR, Neelamma B, Mouli BC, Rao DS, (2016): Int J Pharm Pharm,Synthesis, antimicrobial screening studies of some terpolymer resins derived from 2, 4-dihydroxyphenyl alkyl ketone, melamine and furfural,
7. Sandra M, MD, João F, Paulo C, InêsP,Dmitry E, (2016): Materials,High Pressure Laminates with Antimicrobial Properties, 100 9(2).
8. Palza H, (2015): Int J Mol Sci,Antimicrobial Polymers with Metal Nanoparticles, 2099 16(1).
9. Tashiro T, (2001): Macromol. Mater. Eng,Antibacterial and Bacterium Adsorbing Macromolecules, 63-87 286(2).
10. Dante RC, DA Santamaria, JM Gil, (2009): J. Appl. Polym. Sci,Crosslinking and thermal stability of thermosets based on novolak and melamine, 4059-4065 114(6).
11. Naz MY, Sulaiman SA, Ariwahjoedi BS,Ku K, (2014): Scientific. World. J,Characterization of modified tapioca starch solutions and their sprays for high temperature coating applications, 2014
12. Santhanalakshmi J, (1987): Thermochim. Acta,Studies on the thermal decomposition of thermosetting aniline-formaldehyde resins, 321-327 119(2).
13. Stark W, (2010): Polym Test,Investigation of curing behaviour of melamine/phenolic (MP) thermosets, 723-728 29(6).
14. Christensen G, (1977): Prog Org Coat,Analysis of functional groups in amino resins, 255-276 5(3).
15. Ricciotti L, Roviello G,Tarallo O, Borbone F, Ferone C,Cioffi R, (2013): Int J Mol Sci,Synthesis and Characterizations of Melamine-Based Epoxy Resins, 18200 14(9).
16. Kim SH,J Kim, (2006): Thermochim Acta,Thermal stability and viscoelastic properties of MF/PVAc hybrid resins on the adhesion for engineered flooring in under heating system; ONDOL, 134-140 444(2).
17. Batista MA, J Moraes, RP BJC S, Oliveira P,CSantos AM, (2011): Prog Org Coat,Effect of the polyester chemical structure on the stability of polyester–melamine coatings when exposed to accelerated weathering, 265-273 71(3).
18. Bauer DR, (1986): Prog Org Coat,Melamine/formaldehyde crosslinkers: characterization, network formation and crosslink degradation, 193-218 14(3).
19. Ullah SF, AhmadP.S.M.M. Yusoff, (2013): J Appl Polym Sci,Effect of boric acid and melamine on the intumescent fire-retardant coating composition for the fire protection of structural steel substrates, 2983-2993 128(5).
20. Bal A I, AcarG, Güçlü, (2012): J Appl Polym Sci,A novel type nanocomposite coating based on alkyd-melamine formaldehyde resin containing modified silica: Preparation and film properties, 125(S1).
21. Zanetti M A, Pizzi M, Beaujean H, Pasch K, RodeP, Dalet, (2002): J Appl Polym Sci,Acetals-induced strength increase of melamine–urea–formaldehyde (MUF) polycondensation adhesives. II. Solubility and colloidal state disruption, 1855-1862 86(8).

22. A PizziK, L Mittal, (2003): Handbook of Adhesive Technology,"Melamine-formaldehyde resins," A. Pizzi and K. L. Mittal, Eds., Marcel Dekker, New York, NY, USA, 2nd edition
23. Nemli GM, Usta, (2004): Build Environ,Influences of some manufacturing factors on the important quality properties of melamine-impregnated papers, 567-570 39(5).
24. levy CW, (1999): nature,molecular basis of triclosan activity, 383-384 398(6726).
25. Pract J.C.D.P.C.D.: HSDB (Hazardous Substance Data bank), search: Triclosan. http://toxnet.nlm.nih.gov/cgibin/sis/search,
26. Assadi K.A.R.A.A, (2016): Global J. Pure Appl. Sci,Synthesis and characterization of new oxime-formaldhyde polymer and their biological activities, 1-12 4(1).
27. (1981): American Journal of Public Health,Association News, 362-448 71(4).
28. Xue Y, H Xiao,Y Zhang, (2015): Int J Mol Sci,Antimicrobial Polymeric Materials with Quaternary Ammonium and Phosphonium Salts, 3626 16(2).
29. Tomšič B, Simončič B, Orel B, Žerjav M, Schroers H, Simončič AS, (2009): Carbohyd Polym,Antimicrobial activity of AgCl embedded in a silica matrix on cotton fabric, 618-626 75(4).
30. Toda C M,M, Rodriguez LS, Paleari AG, Pero AC, Compagnoni MA, (2015): J Contemp Dent Pract,Antimicrobial activity of a tissue conditioner combined with a biocide polymer, 101-106 16(2).