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Hygienic coatings by UV curing of diacrylic oligomers with added triclosan

Giuseppe Gozzelino, Antonio Giovanni Dell'Aquila, Diana Romero

Abstract Hygienic coatings have been obtained by UV photopolymerization of mixtures of urethane-diacrylate, tri-propylene-glycol diacrylate and 2,4,4′-trichloro-2′-hydroxydiphenylether (Triclosan). UV-dried coatings containing a weight fraction of Triclosan as low as 0.001 submitted to tests for antimicrobial activity evaluation with *Escherichia coli* colonies have shown the complete elimination of living bacteria in the thin liquid layer in contact with the film surface. A persistent biocide activity can also be observed after a prolonged water treatment of the coating. The biocide release from the coating has been studied through contact with water–ethanol mixtures of different compositions. The maximum release rate has been observed at an ethanol weight fraction of 0.85, where the crosslinked films show the maximum swelling. The phenomena can be explained on the basis of the interaction between the liquid and the polymer network through the Hansen solubility parameters.

Keywords Urethane-acrylate, Networks, Biocide, Films, Photopolymerization

Introduction

The crosslinking of an acrylic formulation through UV curing is well known as an environmental-friendly technology because it offers many advantages, the principal being the VOC-free formulation and the high speed of the drying process.

Antimicrobials are chemicals that influence the growth of microbial flora and they are frequently used in health care to reduce the transmission of pathogenic agents.

The inclusion of antimicrobial agents in a polymer matrix is of great importance in coating technology, where antimicrobial agents are classically used to protect the coating film from the aggression of the bio-environment. They play a key role in packaging materials for the preservation of products such as foods, beverages, cosmetics, and pharmaceutical formulations.[1,2]

Antimicrobial agents are also used in hygienic coatings to confer sterilizing properties to the surrounding environment.[3] To date, these coatings constitute a limited fraction of the potential industrial production but they could be developed, taking into account that the benefit of the antimicrobial property is not just to preserve the treated material against microbiological degradation, but also to provide health-related benefits when stringent hygiene measures are necessary, such as in high-care areas of medical institutions or in pharmaceutical or food industries.

Hygienic coatings can play an increasing role not only in helping to keep food premises, food store rooms, food equipment, and other food-contact materials clean and fit for their purposes, but also for products designed to be in continuous contact with the human body, such as medical devices.[4]

Chemicals with biocidal properties solubilized in the coating can confer antimicrobial properties, if they are allowed to migrate through the polymeric matrix and reach a sufficient concentration on the surface to perform their action,[5] but the efficiency of an antimicrobial agent contained in a polymer matrix cannot be

G. Gozzelino (✉), A. G. Dell'Aquila, D. Romero
Department of Materials Science and Chemical Engineering, Politecnico di Torino, C.so Duca degli Abruzzi, 24, 10129 Torino, Italy
e-mail: giuseppe.gozzelino@polito.it

A. G. Dell'Aquila
e-mail: giovanni.dellaquila@polito.it

D. Romero
e-mail: diana.romero@polito.it

easily predicted or controlled. Moreover, because of the migration, by diffusion, from the matrix toward the surface and from the surface toward the environment, the concentration of the active constituent decreases in time and the activity time of the polymer surface depends on the factors that influence the biocide migration into and out of the polymer bulk.

This study has been performed to show the performances of a coating and the releasing properties of the contained biocide. The effect of a chemical that is commonly used as an antiseptic for hand washing, 2,4,4′-trichloro-2′-hydroxydiphenylether (TCDPE), added to a UV curable formulation, has been analyzed. The antimicrobial agent, which is commercially known as Triclosan,[6] has been incorporated in a urethane-acrylic network by the fast UV photopolymerization of formulations based on acrylic oligomers, biocide, and UV photoinitiator.[7]

The antimicrobial activity of the film surface has been tested with colony forming bacteria that are typical of human pollution, the Gram-negative *Escherichia coli* (*E.C.*). The biocide release from the network has been evaluated by putting the film in contact with ethanol–water solutions of different compositions and by analyzing the biocide content in the liquid at different times.

The biological tests have shown that the crosslinked films are endowed with strong antimicrobial activity with a lower antibacterial content than 1 wt%. The extraction experiments have shown that the amount of the biocide released depends on the interaction between the liquid and the network.

An analysis of the experimental data suggests that the difference in surface solubility parameters between the network and the liquid in contact with the surface could be a good approach to explain biocide release behavior.

Experimental

Materials

The aliphatic urethane-diacrylate oligomer (Eb270, MW: 1500, d: 1.05 g/mL) was a product from UCB Chemicals, Belgium. The biocide TCDPE, the tripropylene-glycol diacrylate (TPGDA, d: 1.10 g/mL) used as a reactive thinner, and the pure ethanol were Aldrich products. The 2-hydroxy-2-methylphenyl-propanone (Darocur 1173) used as a UV photoinitiator was a Ciba product. All products were used as received.

Coating preparation

Reactive mixtures for UV coating were obtained using the Eb270 and TPGDA in different weight ratios, with 3% (w/w) of the photoinitiator. The mixture, with the addition of a suitable quantity of biocide, was spread, using a calibrated wound applicator, on a glass plate in a 200-μm-thick layer.

The liquid film was crosslinked under N_2 atmosphere by UV irradiation with a Hg vapor lamp (25 mW/cm^2). The films were submitted to three irradiations, for a total of 90 s, to obtain the maximum crosslinking of the sample. After the irradiation, the coatings on the glass plate were tested for antibacterial activity.

The biocide release experiments were performed on free films detached from the glass plate.

Kinetics of UV photopolymerization

The kinetics of the curing under UV irradiation were followed by an analysis of the acrylic double bond content using a real-time infrared spectroscopy apparatus. A 10-μm-thick liquid film, placed on a Si plate and submitted to UV irradiation, was analyzed in real time by infrared spectroscopy. The conversion of the acrylic double bond was determined, on the spectra collected at different times, through the ratio of the absorbance peak area of the acrylic double bond (1640 cm^{-1}) with the area of the same peak before the UV irradiation.

Test methods

Antibacterial activity

Only sterile films were submitted to the biological tests. The sterilization was made exclusively with a physical method: 10 min in an autoclave at 120°C. Preliminary tests excluded the possibility of using a chemical sterilizer, such as ethanol, because of the residual antibacterial activity caused by the sterilizer absorbed into the polymer matrix.

The method adopted for the antibacterial activity evaluation was derived from an international protocol for the evaluation of bactericide activity of thermoplastic films,[8] modified to make it applicable for crosslinked films.

A 200 μL volume of a bacterial suspension of *E.C.* in sterile water (1.0×10^5–1.0×10^6 CFU/mL) was spread over a 10 cm^2 area of the sterilized coating. Water evaporation of the biological suspension was avoided by placing a sterile PET film over the spread suspension.

After different contact times, the film was washed with 35 mL of sterile water and a 1.0 mL sample of the *E.C.* suspension was allowed to grow in a Petri dish filled with Agar. The living colonies were counted after 24 h of incubation at 35 ± 1°C and 90% humidity. The result was expressed as the mean of the absolute values of the counted CFU (colony forming units) of at least three samplings.

Biocide release in liquid media

The release of TCDPE was tested in water–ethanol mixtures by monitoring the biocide concentration in the liquid by UV spectrophotometry. Figure 1, where the UV spectra of TCDPE and Darocur are reported, shows the strong TCDPE absorption at 282 nm, which was used as the analytical band for the biocide determination.

In order to follow the kinetics of the biocide release, a film was introduced into an optical quartz system filled with the liquid, which was mechanically stirred and continuously monitored by an Unicam UV2 Spectrometer instrument.

Swelling

The swelling of the crosslinked film was expressed as the percent weight increase of the film sample in contact with the liquid. The samples, placed into different composition liquids at room temperature for 150 min, were dried with blotting paper and weighed.

Results and discussion

The dynamics of the curing under UV irradiation of the formulations containing different ratios of the two diacrylic oligomers are shown in Fig. 2. The figure reports the acrylic double bond (a.d.b.) conversion in time that was observed using real-time infrared analysis of the coating sample during the UV irradiation. The conversion of the liquid formulation into a solid coating, for diacrylate mixtures with a weight fraction of TPGDA in the 60–100 wt% range, was fast. Less than two minutes were required to fully develop the polymerization and to reach a constant concentration of a.d.b.

The presence of a limit in the a.d.b. conversions shows that the glass transition temperature of the system becomes higher than the reaction temperature during the photopolymerization. In other words, the progress of the polymerization increases the crosslinking density of the system and reduces the mobility of the reacting species up to the point where the polymerization stops.

A decrease in the a.d.b. conversion with the increase in the TPGDA in the diacrylic oligomer mixture was also observed. This behavior shows that the TPGDA does not act as a reactive plasticizer, a function that has been reported for diacrylate monomers in similar systems.[9]

The reaction of a single a.d.b. generates a branching of the growing chain, i.e., a crosslinking point of the growing network. The higher the crosslinking density of the network, the lower the mobility of the reacting species. In the case of a total conversion of the a.d.b., the theoretical values of the crosslinking densities, evaluated from the properties of the liquid formulations, result to be 2.0, 2.6, 3.2, and 3.7 mmol cm^{-3} for mixtures containing 10, 20, 30, and 40% (w) of TPGDA, respectively. If the crosslinking density hinders the mobility of the reacting species, it is obvious that the increase in TPGDA reduces the conversion of the a.d.b.

This is in agreement with the data reported in Fig. 2, which shows conversions that decrease with an increase in the TPGDA content. The observed values of a.d.b. conversion show that the curing reaction stops when the crosslinking density reaches a value of about 2.5 mmol cm^{-3}.

The presence of the TCDPE in the formulation does not modify the kinetics of the photopolymerization up to a content of about 10%. An increase in reaction rate and conversion can be observed at higher concentrations than 10%, which is probably caused by the plasticizing effect of the TCDPE.

These concentrations are two magnitude orders higher than the minimum TCDPE concentration needed to confer hygienic properties to the coating: films of crosslinked coating with added TCDPE, submitted to evaluation of antimicrobial activity,

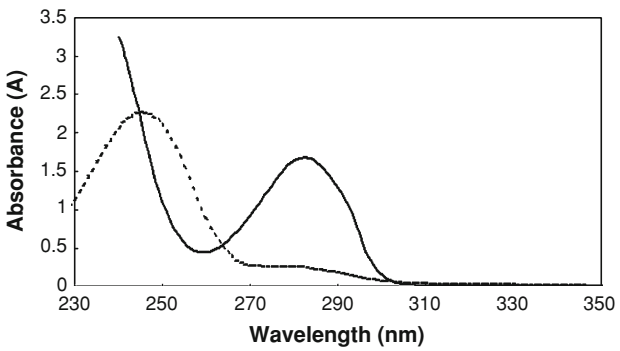

Fig. 1: UV spectra of Triclosan (full line) and photoinitiator (broken line) in the ethanol solution

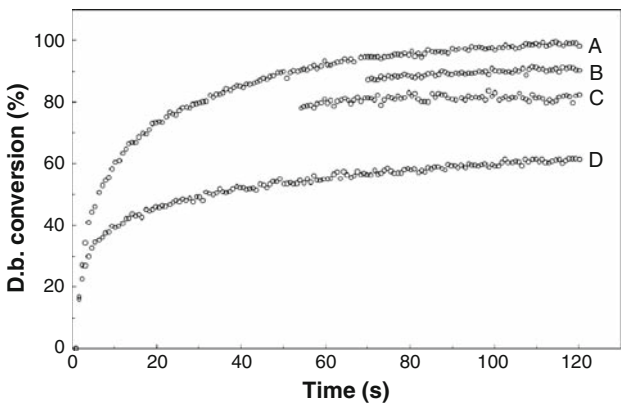

Fig. 2: Kinetics of acrylic double bond conversion under UV irradiation of films with different oligomer compositions. Eb270/TPGDA weight ratio: A = 90/10, B = 80/20, C = 70/30, and D = 60/40

showed evident *E.C.* bacteria growth inhibition properties at a biocide concentration as low as 0.1%.

The counting of the *E.C.* living bacteria units, after different permanence times of the microbial culture on the surface of a crosslinked film containing the TCDPE, is reported in Fig. 3. The data show that there is a consistent increase in living bacteria units on the inert surfaces of sterilized glass as a consequence of the natural bacteria reproduction, but a decrease in the living units, which leads to a total disappearance of bacteria on the coating surface after 24 h of contact.

The antimicrobial activity should be the result of biocide migration from the coating into the liquid bacteria suspension in contact with the surface. The water solubility of TCDPE is very low, 10 ppm at 20°C,[10] but it has been reported that a biocide concentration of 0.1–0.3 ppm is sufficient to inhibit the growth of *E.C.* bacteria.[11]

If the long-term effectiveness is considered, it should be taken into account that continuous contact with a liquid could affect the hygienic efficiency of the coating as a consequence of the biocide depletion of the polymer. In order to have evidence of this effect, the influence of a prolonged water washing of the surface was tested. Table 1 reports the evaluation of the antibacterial activity after the coating surface was submitted to a continuous flow of water at temperatures of 10 and 50°C. As can be seen, the prolonged water washing at the lower temperature does not lead to any decrease in the biocide activity. Despite the presumable partial loss of TCDPE, the coating maintains its hygienic property. On the other hand, the experiments performed at 50°C highlight a complete loss of bioactivity of the coating.

These results suggest that the release is controlled by a very slow diffusion phenomenon of the biocide from the coating into the liquid. The amount of transferred TCDPE in time depends on the parameters that influence the diffusion, such as the temperature, the network swelling, the polymer plasticization caused by the liquid–polymer interaction, and the solubility of the biocide.

Experiments with water–ethanol mixtures were carried out on films containing TCDPE to study the release of biocide into the liquid in contact with the coating surface. The results obtained using liquid mixtures containing ethanol in the 50–95% range are reported in Fig. 4. As can be seen, the shape of the curves of the extracted biocide vs time shows an asymptotic trend, which is typical of Fick's diffusion phenomenon.

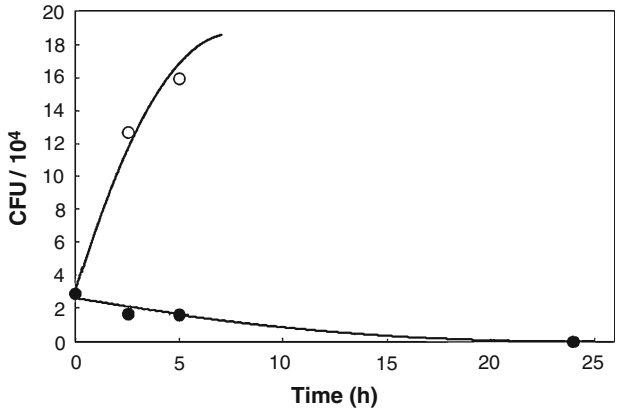

Fig. 3: Counting of living *E.C.* bacteria colonies (CFU) after different contact times with Eb270/TPGDA (60/40) coating containing TCDPE 0.1% (●) and with a glass surface (○)

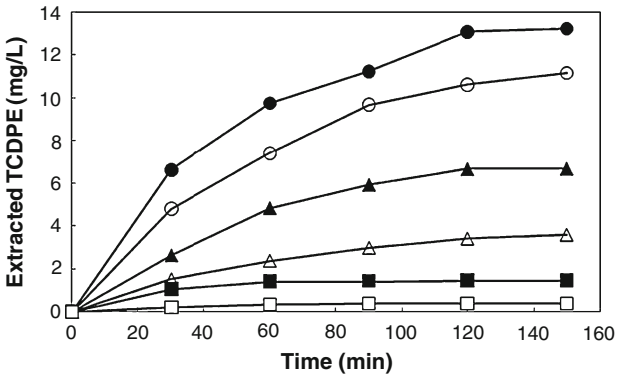

Fig. 4: Biocide extraction in time from crosslinked films by water–ethanol solutions. TCDPE content of the films (Eb 270/TPGDA: 60/40): 0.1% (w/w). Ethanol content in the solution (w/w): (■) 50%; (Δ) 65%; (▲) 75%; (●) 85%; (○) 90%; (□) 95%

Table 1: Counting of *E.C.* bacteria colonies after different time of contact with a coating (Eb270/TPGDA: 60/40, 0.1% of TCDPE) submitted to 48 h of a water flow[a] at different temperatures

Time (h)	T = 10°C		T = 50°C	
	Reference[b] (CFU × 10^{-4})	Coating (CFU)	Reference[b] (CFU × 10^{-5})	Coating (CFU × 10^{-5})
0	1.9 ± 0.2	–	1.0 ± 0.1	–
3	8.5 ± 1.2	0	5.9 ± 1.0	5.2 ± 1.0
6	7.8 ± 1.2	0	6.7 ± 1.0	5.2 ± 1.0

[a] A 2-mm-thick layer of water that flowed on the coating at 3.6 cm s^{-1}

[b] Bacteria colonies in contact with a glass surface

The quantity extracted at longer times, when extraction equilibrium should be reached, cannot be explained by the TCDPE solubility in the liquid. Initially, the extracted TCDPE increases with an increase in ethanol, but a decrease in the amount of the extracted biocide can be observed at higher ethanol contents than 85%.

These data are in contrast with the expected behavior of TCDPE extraction by the liquid, since TCDPE is quite insoluble in water but highly soluble in ethanol, and suggest that the extraction is controlled more by the interaction of the extracting liquid with the network structure than by the TCDPE solubility.

The extraction rates, reported in Fig. 5, show similar behavior in function of the liquid composition: the initial extraction rate has the maximum value with an ethanol concentration of 85%.

The CED of the liquid can be evaluated through the vaporization heat ΔH_v and the molar volume V_m of the liquid mixture

$$\mathrm{CED} = \frac{\Delta H_v - RT}{V_m}$$

The CED values calculated for the different water–ethanol mixtures using the literature data are reported in Table 2.

The CED of the polymer can be evaluated using the group contribution method suggested by Hansen.[13]

In order to build a model of the coating network, the following structures of the two diacrylate oligomers were considered:

$[CH_2=CH\text{-}COO\text{-}(CH_2)_2\text{-}O\text{-}]_2\text{-}CONH\text{-}(CH_2)_6\text{-}NHCO\text{-}[\text{-}O\text{-}CH_2\text{-}CHOH\text{-}CH_2\text{-}O\text{-}CONH\text{-}(CH_2)_6NHCO]_4$

(urethane-diacrylate oligomer)

$CH_2=CH\text{-}COO\text{-}CH_2\text{-}CH\text{-}CH_3\text{-}[\text{-}O\text{-}CH_2\text{-}CH\text{-}CH_3\text{-}]_2\text{-}OCO\text{-}CH=CH_2$

(tripropylene-glycol diacrylate oligomer)

These results can be explained if the swelling data of the coating in contact with liquids of different compositions are analyzed. Figure 6, where the values of the swelling of the coating with water–ethanol mixtures in the 50–100% range of ethanol content are reported, clearly shows a maximum swelling at an ethanol concentration of about 85%.

The swelling of the network, the consequent mobility of the solubilized TCDPE, and the TCDPE–polymer and TCDPE–liquid interactions are the parameters that control the release phenomena from the coating. The volume expansion of the network allows an easier diffusion of the TCDPE in the coating bulk and increases the transport rate from the polymer to the liquid. In other words, when the film is in contact with differently interacting liquids, the rate of extraction and the swelling must behave in the same way.

On the other hand, the higher the swelling, the higher the liquid–network interaction and, if the TCDPE is adsorbed on the polymer network, the increase in the liquid–polymer interaction reduces the TCDPE–polymer interaction and increases the maximum amount released into the liquid. The rate of the release is controlled by diffusion[12] but the maximum amount of biocide released into the liquid appears to be controlled by the interaction between the liquid and the polymer network.

One way of analyzing the polymer–liquid interaction is through the solubility parameter δ, which is evaluated as the square root of the cohesive energy density (CED)

$$\delta = \sqrt{\mathrm{CED}}$$

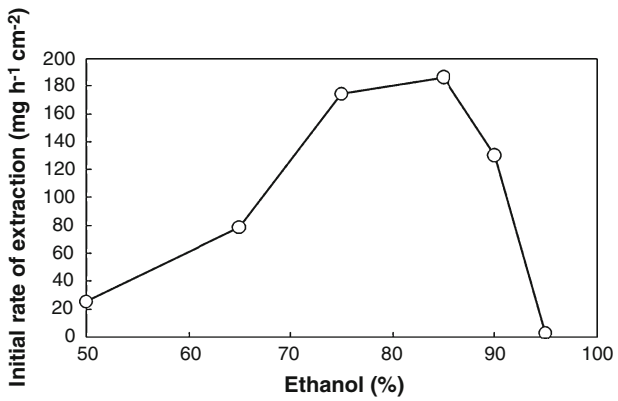

Fig. 5: Initial rate of TCDPE extraction as a function of ethanol content of the liquid mixture

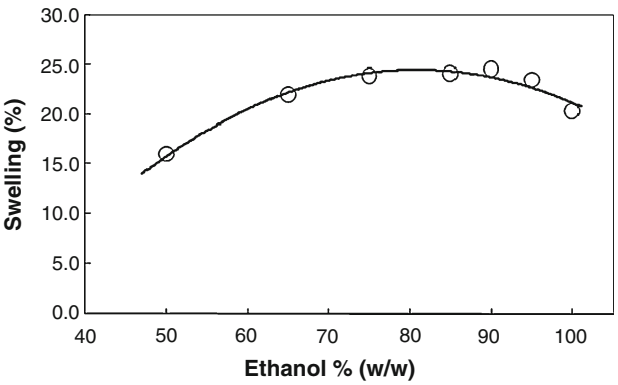

Fig. 6: Swelling of crosslinked coating (Eb270/TPGDA: 60/40) in the presence of water–ethanol mixtures

Table 2: Solubility parameters of water–ethanol mixtures evaluated through the heat of vaporization

EtOH (% mol)	EtOH (w/w)	ΔH mix[a] (cal g^{-1})	ΔH vap[b] (cal cm^{-3})	CED (cal cm^{-3})	δ (J$^{0.5}$ cm$^{-1.5}$)
100.000	1.000	0.000	172.294	162.115	25.8
95.000	0.984	0.213	179.746	169.204	26.4
90.000	0.967	0.458	187.707	176.774	27.0
80.000	0.928	1.188	205.618	193.809	28.3
70.000	0.883	1.882	226.592	213.754	29.7
60.000	0.829	2.753	251.250	237.187	31.3
50.000	0.764	3.539	280.937	265.391	33.1
40.000	0.683	4.933	317.845	300.465	35.2
30.000	0.581	6.728	364.225	344.522	37.7
20.000	0.447	8.644	423.164	400.421	40.6
10.000	0.264	8.962	496.262	469.369	44.0
5.000	0.145	6.046	537.483	507.890	45.7
0.000	0.000	0.000	585.240	552.344	47.7

[a] From reference (16)

[b] From reference (17)

The components δ_d, δ_p, and δ_h of the solubility parameter of these structural units of the network can be calculated using the Hansen method by adding the contributions of all the i groups that constitute the structural units. Through the evaluation of the dispersive ($F_{d,i}$), polar ($F_{p,i}$), and hydrogen ($F_{h,i}$) contributions of each group, the components of the solubility parameter can be obtained as:

$$\delta_d = \frac{\sum F_{d,i}}{V} \quad \delta_p = \frac{\sqrt{\sum F_{p,i}^2}}{V} \quad \delta_h = \sqrt{\frac{\sum F_{h,i}}{V}}$$

and the solubility parameter as:

$$\delta = \sqrt{\delta_d + \delta_p + \delta_h}$$

The values of the molar volume V_m and of the dispersive, polar, and hydrogen components of the urethane-diacrylate and tripropylene-glycol diacrylate structural units on which the network is based, calculated from group contribution values available in literature,[14] are reported in Table 3.

The network solubility parameter δ_{net} formed by the two oligomers was obtained by applying the equation for the solubility parameter of a mixture:

$$\delta_{net} = \frac{x_1 V_1 \delta_1 + x_2 V_2 \delta_2}{x_1 V_1 + x_2 V_2}$$

where V and x are the molar volume and the molar fraction of the two components of the network structure, respectively.

For the network based on the 60/40 mixture (urethane-diacrylate)/(TPGDA), a δ_{net} = 27.4 J$^{0.5}$ cm$^{1.5}$ can be obtained.

This value agrees with the solubility theory of polymers, which allows the interaction of a polymer with a solvent to be predicted on the basis of the difference between their solubility parameters.[15]

In Fig. 7, where the differences between the calculated solubility parameters of the liquid mixture and of the network, $\Delta\delta$, are reported, it is evident that the predictable composition of the water–ethanol mixture

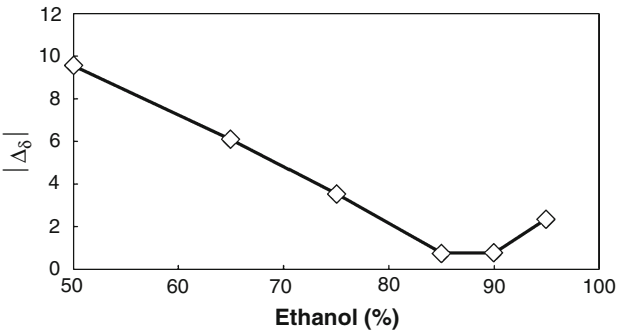

Fig. 7: Difference between solubility parameters of the water–ethanol mixture and of the coating network

Table 3: Solubility parameter and dispersion, polar, and hydrogen components of the structural units of the network obtained by the group contribution method[14]

	V_m (cm^3 mol^{-1})	δ_d (J$^{0.5}$ cm$^{1.5}$)	δ_p (J$^{0.5}$ cm$^{1.5}$)	δ_h (J$^{0.5}$ cm$^{1.5}$)	δ (J$^{0.5}$ cm$^{1.5}$)
Urethane-acrylic	989	22.3	8.16	14.0	27.6
TPGDA	197	24.5	5.55	10.1	27.0

that causes the maximum interaction between the liquid and film must be in the 85–90% of ethanol range. The maximum swelling of the network and, as a consequence, the higher rate and amount of released additive, should occur in this range of composition.

The results obtained in the evaluation of the solubility parameters of a polymer network using the Hansen method suggest that this could be a suitable way of predicting the behavior of hygienic coatings containing a soluble biocide in contact with liquids that interact with the polymer.

Conclusions

Crosslinked films obtained by UV photopolymerization of urethane-diacrylate and tri-propylene-glycol diacrylate oligomers with added 0.1% (w/w) of TCDPE (Triclosan) show bactericide activity toward *E.C.* bacteria and perform like hygienic coatings.

The antimicrobial activity of the coating is maintained even after prolonged washing with cool water.

Extraction of the bactericide with water–ethanol mixtures shows that the loss of bactericide is more related to the liquid–network interaction than to the solubility of the bactericide in the liquid.

A good prevision of the biocide release behavior of the hygienic coating in contact with liquid mixtures can be obtained through a comparison of the solubility parameters of the network and the solubility parameter of the liquid mixture, evaluated through the Hansen method and using the heat of vaporization, respectively.

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