



Correlation between the scratch resistance of UV-cured PUA-based coatings and the structure and functionality of reactive diluents

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ABSTRACT

UV-curing technology is a powerful technique for a wide range of applications. The advantages of UV-curing, including speed and process controllability, and the wide range of raw materials and solvent-free formulations are well-known. However, some specific end-product issues such as the migration of components in food packaging, the odor in thiol-based formulations, the scratch or abrasion resistance in the automotive and aeronautic industry still need to be addressed. In this work, the predictable tendency of the scratch resistance of a polyurethane acrylate (PUA)-based coating is proposed with respect to the structure and functionality of the reactive diluent used. The correlation between the scratch resistance and abrasion resistance of or adhesion to the substrate was also demonstrated.

1. Introduction

The interest in UV-curable coatings has been growing during the last few decades [1,2]. The low-cost, controllable process [3], the speed of the curing and the environmental benefits [4] make radiation curing technology extremely convenient for various industries [5]. Coatings are mostly used for substrate protection, so they must exhibit specific resistance properties [6] and satisfy some applicative requirements from various industries such as the automotive [7], aeronautic and building industries. Among the different properties, the scratch and abrasion resistances are crucial ones. Indeed, damage can easily originate from several agents, which can ruin the entire substrate surface and decrease the product lifetime. Therefore, knowledge regarding the structural properties of the coating involved in scratch and abrasion phenomena is required for proper enhancement of the coatings. According to the force applied, different degrees characterize the scratch damage [8] abrasion, cracking, delamination and chipping. When low normal force values are applied to a coating, abrasion mar damages may occur. This type of damage will impact the surface of the coating ruining the appearance of the film. Even for lightest damage, the loss of gloss and the incoming optical haze at the surface for automotive, outdoor and indoor applications are visible to the naked eyes. When increasing the force applied to a coating, scratch damage is achieved, which leads to appearance issues and partial or complete loss of adhesion. The scratch is characterized by plastic deformation and fracture

(cracking) [6], depending on the ductility or brittleness properties of the coating. The plastic deformation occurs in soft coatings, in which the force applied to the film leads a matter movement in front of the tip [9,10]. Fracture damage occurs more easily in brittle coatings than in soft coatings. In the literature, scratch resistance measurements are achieved via several techniques (abrasion, pencil and stylus tests, etc.). However, approaching the problem with different methodologies leads to contradictory results. Numerous technical factors such as the geometry of the tip [11,12], the scratch loads, the speed of the tip motion, and the friction coefficient strongly affect the final scratch result. Scratch testing with a diamond indenter is the most reported tribological method, which provides the dynamic surface mechanical properties [11,13].

Scratch resistance not only depends on the coating adhesion on the selected substrate [14] but is also frequently linked to the coating hardness. Adhesion is influenced by the nature of the polymer material and its thickness; the surface properties of the substrate and the coating application process have also to be taken into account. Therefore, substrate surface treatments are commonly implemented in industrial applications [15]. Coating hardness mostly relies on the polymer material properties and more particularly on the chemical skeleton of the polymer, the monomer or oligomer functionality and the crosslinking density of the polymer network [16,17]. Different approaches have been reported to increase the intrinsic polymer hardness in thermal- or light-cured coatings: the use of alumina or silica nanoparticles

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(nanofillers) [18], the *in situ* generation of an inorganic network [19–21] are efficient ways to improve the scratch and abrasion resistance [18,22,23]. Interestingly, identification of the key parameters governing the relationship between the UV-cured coating and its resistance to scratch is mostly based on empirical results. Therefore, the aim of the present paper is to focus on the polymer network characteristics by leaving aside the addition of an inorganic moieties or substrate surface treatment that could contribute to increase the scratch resistance. The correlation between the scratch resistance and chemical structure has been investigated for an aliphatic urethane acrylate oligomer, which is part of a valued oligomer family due to its flexibility and moderate toughness. Aliphatic urethane acrylate oligomers are commonly used in exterior applications due to their resistance to UV radiations. Different UV curable formulations were prepared with various monomers usually added to achieve the desired viscosity corresponding to the application process. The influence of the monomer structure, its functionality and the monomer/oligomer ratio on the final mechanical properties and more specifically on scratch resistance was studied. The correlation between scratch resistance and adhesion was investigated by comparing the impact of three types of steel substrates. Finally, the interdependency between scratch and abrasion resistance was addressed.

2. Experimental section

2.1. Materials

An aliphatic polyester-based urethane diacrylate oligomer (CN991 from Sartomer) with a molecular weight of 1500 g/mol was selected for this study. Different reactive acrylate diluents were used to study the structure-scratch resistance relation, changing their functionality and their backbone structure. These diluents are reported in Table 1. 2-Hydroxy-2-methyl-1-phenylpropan-1-one ((D1173 from BASF) was chosen as the photoinitiator. Ebecryl 170 (Allnex) was used as the adhesion promoter. It is an acidic diacrylate with a viscosity of 3200 mPa.s at 20 °C. All the chemicals were used without further purification.

The substrate used for all the experiments was black steel (from Hussor) plates (75 × 75 × 5 mm). For specific experiments, two other metal substrates were tested: hot-laminated stainless steel (Uginox F17 by Hussor) and cold-laminated stainless steel treated with microballs (by Aqua Decoupe).

2.2. Methods

2.2.1. Formulation and curing

The metal plates were washed with soap and dried with acetone prior to the coating deposition. For all the formulations, the amounts of photoinitiator (D1173) and Ebecryl 170 were constant and set at 3 and 5% w/w of the overall formulation weight, respectively. Different monomer ratios were studied at 30%, 50% and 70% w/w. After 24 h of stirring, the formulation was applied onto the substrate by a motorized film applicator equipped with a 30 μm thick spiral bar coater. The coated substrates were UV cured under a Hg lamp (Fusion H bulb, intensity of 4.5 W.cm⁻² and light dose of 1.4 J.cm⁻²) at a belt speed of 9.5 m.min⁻¹ under air.

For thermomechanical characterization by dynamic mechanical analysis (DMA), the formulations were coated on a polypropylene foil in order to peel off the UV-cured films.

2.2.2. Characterization and analysis

The final coating properties depend on the monomer M (its nature, functionality, molecular weight and amount) combined with the aliphatic polyester-based urethane diacrylate (oligomer O).

Therefore, the M/O ratio described in Equation 1 was calculated for all the formulations.

Table 1

List of all the reactive diluents used in this study. Their abbreviation, structure, IUPAC name, functionality, molecular weight and T_g provided by Sartomer are reported.

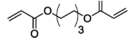
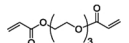
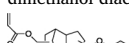
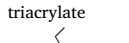
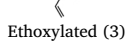
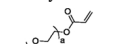
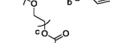
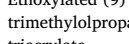
Abbreviation	Name and structure	Functionality	Molecular Weight (g/mol)	T_g (°C)
HDDA	1,6-hexanediol diacrylate 	2	226	86
TIEGDA	Triethylene glycol diacrylate 	2	258	54
TCDDMDA	Tricyclodecane dimethanol diacrylate 	2	304	220
TMPTA	Trimethylolpropane triacrylate 	3	296	85
TMP3EOTA	Ethoxylated (3) trimethylolpropane triacrylate 	3	428	100
TMP9EOTA	Ethoxylated (9) trimethylolpropane triacrylate 	3	692	N/A
PETTA	Pentaerythritol triacrylate 	4	352	70
4EOPETTA	Ethoxylated (4) pentaerythritol tetraacrylate 	4	528	93

Table 2

Adhesion, scratch resistance and T_g of a photocured film containing 97 wt.% CN991, 3 wt.% D1173 and 5 wt.% Ebecryl 170 with respect of the total weight.

CN991 oligomer	
Adhesion	3B
Scratch Resistance (N)	0
T_g (°C)	30.1

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