



# Potential application of some coumarin derivatives incorporated thiazole ring as ecofriendly antimicrobial, flame retardant and corrosion inhibitor additives for polyurethane coating

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## ABSTRACT

Coumarin derivatives continue to draw the attention of synthetic organic chemists due to their versatile utility associated with them and their widely use as pharmaceuticals. Coumarin derivatives having various substituted thiazole rings at carbon-3 exhibit promising biological activities. In this research, two series of coumarin derivatives incorporated thiazole ring were prepared; coumarin thiazole derivative and benzo coumarin thiazole derivative, the structures of the products were confirmed by elemental analysis, FTIR, <sup>1</sup>HNMR and mass spectroscopy. The prepared coumarin derivatives I–III was incorporated physically into polyurethane coating as an additive. Experimental coatings were fabricated on a laboratory scale and applied by brush on steel and wood panels. The coated films were screened against gram-negative bacteria, gram-positive bacteria and fungi to evaluate the antimicrobial activity of the prepared coumarin derivatives. A flame retardant property, the corrosion resistance and the mechanical properties of these coated films were also examined to evaluate the drawbacks of the prepared coumarin derivatives as an additive. The obtained results showed that, the incorporation of a few amount of the prepared coumarin derivatives into polyurethane coating have led to improve the antimicrobial activity, flame retardancy, corrosion resistance, gloss and scratch hardness of the polyurethane coated films.

## 1. Introduction

Coumarin is a natural product; it's found in many plants, it was first isolated in 1820 and it has been used in perfume industries since 1882. Coumarin and its derivatives possess remarkable activities against bacteria [1] and fungi [2]. Coumarin compounds also form a group of more than 40 drugs, which are used in medicine. Recently, the synthesis of some new coumarin incorporated thiazolyl semicarbazones with good anticonvulsant activity, while analgesic and anti-inflammatory activities of thiazolyl coumarins [3] are also known. Some thiazolyl coumarin analogues are found to have potential as anticancer and antimicrobial agents [4]. On the basis of all of this evidence, a new series of biologically active agents containing both of thiazole and coumarin important pharmacophores, were synthesized and characterized by Afsheen et al. exhibited very good activities against all of the tested microbial strains [5]. Compounds containing thiazole rings are a

familiar group of heterocyclic compounds possessing chemotherapy, fungicidal and pesticidal properties. In addition, 2- aminothiazole derivatives are reported to exhibit significant biological activities [6]. The thiazole nucleus is an integral part of all the available penicillin, which have revolutionized the therapy of bacterial diseases. A lot of work has been done in the last few decades on the thiazole ring, to find new compounds to act as antimicrobial and antifungal [7–9]. Some thiazole derivatives were used as an antimicrobial additive in polyurethane coating was reported [10]. The corrosion behavior of some thiazole derivatives toward mild steel has been recently studied, thiazole derivatives are considered non-cytotoxic substances, with the potential to replace other toxic organic corrosion inhibitors used for the protection of metals [11]. Antifouling paints, i.e. paint formulations traditionally containing biocidal species, are used to protect the submerged surfaces from marine biofouling [12]. Organotin compounds, i.e tributyltin compounds was the most effective antifouling

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paint formulation [13], which was completely prohibited by 1 January 2008 by the International Marine Organization (IMO) [14], due to environmental issues [15]. The paint manufacturers' challenge was to develop alternative antifouling paints as effective as the organotin compounds [16–20]. One way to obtain environmental paints consists in developing coatings, which may contain a non-toxic antifouling molecule, i.e. natural products [21–24] were reported. Polyurethanes are a one of the most important class of polymers that have wide application in industrial sectors, they are commonly utilized for corrosion protection of steel structures used in highly corrosive atmospheres [25], they are susceptible to microbial attack, when they are exposed to the atmosphere [26,27], micro-organisms have been found to cause damage of coating materials under various service conditions [28,29]. Some arylhydrazone ligand and its metal complexes, and a pyrimidine derivative was used as flame retardant and antimicrobial additives in polyurethane coating were prepared and evaluated [30,31]. Based on the above mentioned facts, this study reports the synthesis, characterization and evaluation of coumarin derivatives incorporated thiazole ring, and study their potential application as an antimicrobial, flame retardant, and corrosion inhibitors additive in polyurethane surface coating. These compounds were anticipated to improve the biological activities and the flame resistance of polyurethane coating. The mechanical properties were also studied to evaluate any drawbacks associated with these additives.

## 2. Experimental

### 2.1. Materials

All the chemicals used during the research work were sourced both internationally or from local companies were of pure grade quality, and used without any additional purification. Salicylaldehyde, a product of Santa Cruz Biotechnology, USA. Ethyl acetoacetate, a product of Merck Schuchardt Co. German. Piperidine, a product of Merck Millipore laboratories, England. Thioacetamide (ethanethioamide) and 1-Hydroxy-2-naphthaldehyde, a product of Fluka Chemie AG, Buchs, Switzerland. Bromine, a product of sigma Aldrich Chemical Co. USA. Ethyl alcohol absolute, a product of CDH Chemical Co. India. Acetic acid glacial and benzene, a product of El-Naser Pharmaceutical, Chemical Co. Egypt.

### 2.2. Methods and techniques

The prepared coumarin and benzocoumarin derivatives incorporated thiazole ring, compounds I–III was based on the method of C.F. Koelsch; when 3- acetocoumarin is treated with bromine, there is obtained a monobromo derivative, such derivative would be a useful intermediate in the synthesis of compounds related to coumarin and benzocoumarin [32].

#### 2.2.1. Synthesis of 3-acetyl-2H-chromen-2-one (3-acetylcoumarin)

A mixture of salicylaldehyde (0.01 mol, 1.22 g) and ethyl acetoacetate (0.01 mol, 1.3 g) was mixed, stirred and cooled. Few drops of piperidine were added to this mixture with continuous shaking, the mixture was kept in the refrigerator for 3 h. The separated solid was washed with ethanol. Crystallization of the solid from the water gave yellow needle crystals with m. p. 120–122° C and 82% yield. M F: C<sub>11</sub>H<sub>8</sub>O<sub>3</sub>, MW: 188.18

#### 2.2.2. Synthesis of 3-(2-bromoacetyl)-2H-chromen-2-one (3-(bromoacetyl) coumarin)

3-acetyl-2H-chromen-2-one (0.01 mol, 1.88 g) was dissolved in 30 ml acetic acid, then bromine (0.01 mol, 0.51 ml) soluble in the fewest amount of acetic acid being added drop wise under sunlight. After the addition of the bromine, the reaction mixture was left over for 2 h. The reaction mixture was then poured into 100 ml of cold water,

and the solid obtained was filtered and re-crystallized from benzene. A pale yellow solid with m. p. 164–168° C and 53% yield was obtained. M F: C<sub>11</sub>H<sub>7</sub>O<sub>3</sub>Br, MW: 267.08

#### 2.2.3. Synthesis of 3-(2-methylthiazol-4-yl)-2H-chromen-2-one (compound I)

A mixture of 3-(2-bromoacetyl)-2H-chromen-2-one (0.01 mol, 2.67 g) and thioacetamide (0.01 mol, 0.75 g) were dissolved in 30 ml absolute ethanol and a few drops of acetic acid was added. The reaction mixture was then refluxed for 2 h. A precipitate formed on cooling, which was collected by means of filtration, washed with ethanol and dried under vacuum. The solid obtained was then re-crystallized with an ethanol/benzene solvent mixture, resulting in greenish – yellow colored crystals.

#### 2.2.4. Synthesis of 2-acetyl-3H-benzo[f]chromen-3-one

A mixture of 1-hydroxy-2-naphthaldehyde (0.01 mol, 1.72 g) and ethylacetoacetate (0.01 mol, 1.3 g) were taken in 50 ml ethanol, 3–4 drops of piperidine were added and the reaction mixture was stirred for 10 min at room temperature, the mixture was then kept in refrigerator overnight. A yellow solid obtained by filtration was washed with ethanol and dried over the vacuum pump, m. p. 188–190° C and 83% yield. M F: C<sub>15</sub>H<sub>10</sub>O<sub>3</sub>, MW: 238.24

#### 2.2.5. Synthesis of 2-(2-bromoacetyl)-3H-benzo[f]chromen-3-one

2-acetyl-3H-benzo[f]chromen-3-one (0.01 mol, 2.38 g) was stirred in 30 ml acetic acid, with the bromine (0.01 mol, 0.51 ml) soluble in a few amount of acetic acid being added drop wise under sunlight. After the addition of the bromine, the reaction mixture was left over a 2 h. The reaction mixture was then poured into 100 ml of cold water, and the solid obtained was filtered and re-crystallized from benzene. m. p. 182–184° C and 76% yield. M F: C<sub>15</sub>H<sub>9</sub>O<sub>3</sub>Br, MW: 317.13

#### 2.2.6. Synthesis of 2-(2-(4-fluorobenzylidene)hydrazinyl)thiazol-4-yl)-3H-benzo[f]chromen-3-one (compound II)

A mixture of 2-(2-bromoacetyl)-3H-benzo[f]chromen-3-one (0.01 mol, 3.17 g) and 2-(4-fluorobenzylidene) hydrazine carbothioamide (0.01 mol, 1.97 g) was refluxed in 30 ml ethanol for 2 h. The obtained product that formed was collected by filtration and recrystallized from ethanol to give yellowish brown needle.

#### 2.2.7. Synthesis of 2-(2-methylthiazol-4-yl)-3H-benzo[f]chromen-3-one (compound III)

A mixture of 2-(2-bromoacetyl)-3H-benzo [f] chromen-3-one (0.01 mol, 3.17 g) and thioacetamide (0.01 mol, 0.75 g) were dissolved in 30 ml absolute ethanol. A few drops of acetic acid were added to this mixture, and the reaction mixture was refluxed for 3 h. A precipitate formed on cooling, which was collected by means of filtration, washed with ethanol and dried under vacuum. The solid obtained was then re-crystallized with an ethanol/benzene solvent mixture, resulting in dark green-colored crystals.

### 2.3. Spectroscopic analysis

IR spectra were carried out at the Regional Center for Mycology and Biotechnology, Al-Azhar University, Cairo, Egypt. It was recorded in Mac FTIR spectrometer (KBr technique). <sup>1</sup>HNMR spectra (DMSO-d<sub>6</sub>) were recorded on a Varian Mercury-300 NMR spectrometer at Micro-analytical Centre, Cairo University, Egypt. Mass spectroscopy was measured at the Regional Center for Mycology and Biotechnology, Al-Azhar University.

### 2.4. Coating composition

The coating compositions were designed by means of incorporating the prepared coumarin derivatives I–III in the ratio of 0.5, 1.0 and

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