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INNOVATIVE METAL COMPLEXES AS PHOTOSTABILIZERS TO IMPROVE THE FUNCTIONAL LONGEVITY OF POLYVINYL CHLORIDE

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Abstract: Polyvinyl chloride (PVC) is involved in diverse applications that significantly influence our daily lives; it is industrially synthesized in various configurations in order to fulfill the target application. However, PVC is susceptible to photodegradation, yielding toxic byproducts detrimental to both the environment and human health, which diminishing its utility lifespan. Addressing this concern, current research explores the application of metal complexes as potential photostabilizers to mitigate PVC photodecomposition. The objective of this study is to investigate the effectiveness of formulated metal complexes in preventing photodegradation. Five metals, namely Cu(II), Ni(II), Cd(II), Zn(II), and Sn(II), are included. Examining parameters, such as molecular weight attenuation, the formation of alkene-laden and carbonyl fragments, and the surface morphology alterations of PVC sheets subjected to irradiation are considered. Findings indicate that these metal complexes exhibit remarkable efficacy in decreasing the photodegradation of PVC, thus presenting a promising avenue for extending its functional longevity. The polymer containing Cd(II) complex unit showed the least damage in the PVC surface due to photoirradiation, followed by other complexes.

Keywords: polyvinyl chloride; metal complexes; photodegradation; surface morphology; molecular weight reduction

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1. Introduction

Polymers play a pivotal role in numerous applications owing to their distinctive properties, cost-effectiveness in manufacturing, and adaptability to various forms [1]. Enhancement of polymeric material properties is often achieved through blending and doping techniques [2], resulting in significant modifications to the surface morphology,

structure, physical attributes, and overall performance [3,4]. The efficacious incorporation of metal-containing organic moieties into polymeric materials has been demonstrated to enhance corrosion resistance, durability, and resistance to aging. In particular, the integration of rare-earth oxides-based composite materials showed noticeable

advancements [5]. As a result, many studies have been published using these materials; yet, additional research is imperative to generate higher efficient polymeric composites suitable for diverse applications [6,7].

The impact of the doping procedure is influenced by diverse factors, where miscibility, particle size, phase segregation, dispersion, and crystalline are the most popular ones [8]. Today, PVC has witnessed a growing global demand due to its distinctive properties and cost-effectiveness production. It is extensively used in many industries, including office supplies, building materials, toys, furniture, medical devices, packaging, tubing, film and sheet manufacture, and more. However, PVC's chlorine content constitutes more than half of its weight, rendering it valuable for insulator production [9, 10]. Also, ultraviolet (UV) light, sunshine, and high temperatures can cause photodegradation [11, 12]. This photodegradation induces alterations in both the chemical and physical properties of PVC, resulting in undesirable changes [13]. Notably, the consequences of PVC photodegradation

encompass mass loss, the hindering of volatile products, and the formation of fragments with lower molecular weights [14–18]. To counteract these effects and ensure prolonged utility, additives are incorporated into the PVC during manufacturing. These additives enhance the photostability of PVC while simultaneously improving its transparency, durability, and color over an extended duration [19, 20]. Various additives at low concentrations have been used to enhance the PVC photostability upon UV irradiation [21]. The most common additives include inorganic salts [22], heterocycles [23] and aromatics [24].

We present here a study of the effective use of pre-synthesized metal complexes as a sequence of our previously published studies [19, 20]. These complexes can serve as novel photostabilizers to mitigate PVC photodegradation when exposed to UV light irradiation. Different techniques were used to study the PVC films and findings showed a remarkable improvement in the photo-resistivity.

2. Materials and Methods

2.1. General

The reagents and chemicals were purchased from Merck of Gillingham, UK. However, the material used was PVC obtained from Petkim Petrokimya in Istanbul, Turkey, with a molecular weight (M_v) of around 180,000. The Fourier Transform Infrared (FT-IR) of spectra range $4000\text{--}400\text{ cm}^{-1}$ was recorded on a Jasco FT/IR-4200 spectrometer (Tokyo, Japan). The PVC sheets were exposed to irradiation at $25\text{ }^\circ\text{C}$ utilizing a Q-Panel tester from Homestead, FL, USA, with a UV light with the highest wavelength and light intensity of 365 nm and $6.2 \times 10^{-9}\text{ Einstein dm}^{-3}\text{ s}^{-1}$. The sheets were set vertically, 10 cm away from the light source and in line with the UV lamps, while the tester stood on either side of the light with two fluorescent lamps that emit ultraviolet light (UV-B 365, 40 watts). The plastic sheets were rotated regularly to provide even irradiation from all directions. Ostwald U-Tube viscometers (Ambala, Haryana, India) were used to estimate viscosity. Using a FEI Inspect

S50 microscope (Czech Republic), the surface morphology of the irradiated sheets was investigated.

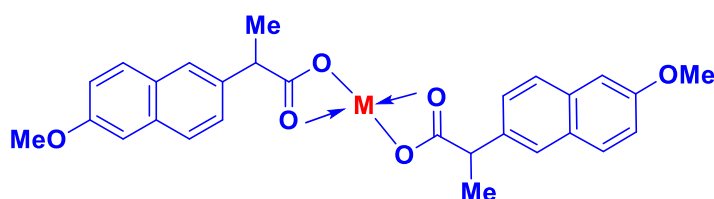
2.2. Synthesis of metal complexes

Metal complexes involving Cu(II), Ni(II), Cd(II), Zn(II), and Sn(II) metals with 2-(6-methoxynaphthalen-2-yl) propanoate were synthesized using the previously established method [19,20], where the structure is illustrated in Scheme 1. The preparation of metallic complexes with 2-(6-methoxynaphthalen-2-yl) propanoate, conducted in a 1:2 molar ratio, resulted in the formation of the respective complexes [19, 20].

2.3. PVC Films Preparation

Mixing 5 g of PVC with 25 mg of metal complexes in 100 ml of tetrahydrofuran (THF) allowed the mixture to swirl for two hours. The resulting uniform solution was subsequently doped onto a glass tablet that had 15 cavities measuring around $45\text{ }\mu\text{m}$ in thickness. The tablet was left to dry in air for 24 hours at $25\text{ }^\circ\text{C}$. After that, to make sure that all traces of THF

were completely removed, the films went through another drying process for 8 hours in a vacuum oven set at 40 °C.



M= Ni(II), Cu(II), Zn(II), Cd(II) and Sn(II)

Scheme 1. Synthesis of metal complexes.

2.4. Monitoring the photodegradation of PVC films by FTIR Spectrophotometry

Photodegradation of PVC leads to the formation of tiny plastic pieces with carbonyl (C=O) and alkene (C=C) groups. The main reason for this breakdown is dehydrochlorination, which is the process of removing hydrochloride from the plastic chains [21]. At different irradiation periods, FTIR spectra of the PVC sheets that were exposed to

radiation were recorded. Using the C-H bond as a reference peak at 1328 cm⁻¹ for comparison, the strength of the absorption bands corresponding to the C=O and C=C groups were at 1714 cm⁻¹ and 1618 cm⁻¹, respectively. By plugging the values of the reference peak (A_{C-H}) and the absorbance of the functional group (A_{C=O} or A_{C=C}) into Equation (1), we were able to estimate the functional group indexes (I_s, I_{C=O}, or I_{C=C}).

$$I_s = \frac{A_s}{A_r} \quad (1)$$

2.5. Average Molecular Weight (M_v)

We examined the inherent viscosity, abbreviated as [η], of the PVC sheets after dissolving them in THF after irradiation. The

M_v of the irradiated films was determined using the Mark-Houwink equation [22] as shown in Equation (2).

$$[\eta] = 1.63 \times 10^{-2} M_v^{0.766} \quad (2)$$

3. Results and discussion

3.1. FTIR Spectrophotometry

The initiation of radical species, such as chloride and carbon radicals, occurs through PVC photooxidation induced by photoirradiation in an oxygen-rich environment.

These radicals play a critical role in the detrimental degradation of PVC, leading to the formation of volatile byproducts, notably HCl. Consequently, C=O moieties (including chloroketones and ketones) and C=C groups within unsaturated chains are generated in the residual PVC [23]. To evaluate the impact of irradiation on PVC, Fourier-transform infrared

(FTIR) spectroscopy was employed. Throughout the irradiation process, bands corresponding to the vibrations of the C=O at 1714 cm⁻¹ and C=C at 1618 cm⁻¹ groups were observed to increase in intensity. Intensification of these functional groups was compared to a reference peak (C-H bond; 1328 cm⁻¹), which remained relatively unaffected by the procedure [23]. Figure 1 illustrates changes in the vibration band intensities of C=O and C=C observed in the FTIR spectra of pure PVC as a result of irradiation.

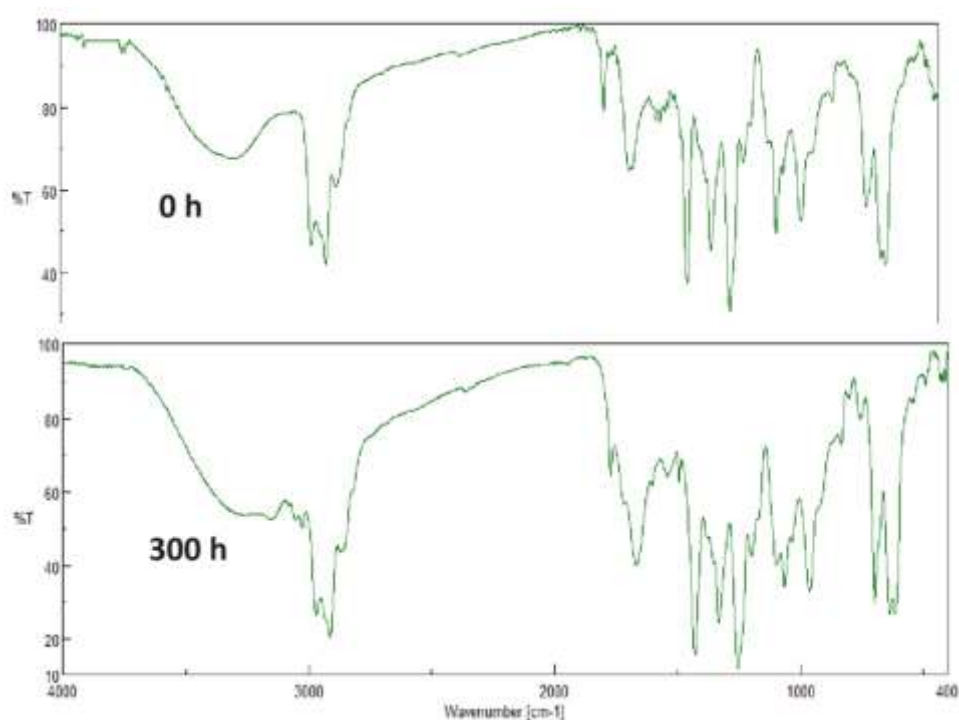


Figure 1. FTIR spectra of PVC films: comparison between non-irradiated and irradiated states.

3.2. $I_{C=O}$ and $I_{C=C}$ of the polymeric films

Following irradiation, the functional group indices $I_{C=O}$ and $I_{C=C}$ were calculated using Equation (1) and plotted against the irradiation time (Figures 2 and 3, respectively). Both $I_{C=O}$ and $I_{C=C}$ showed an upward trend

during the irradiation process, with the pristine PVC case showing the most significant changes. Importantly, the increases in $I_{C=O}$ and $I_{C=C}$ were relatively smaller for PVC blends containing metal complexes compared to the blank film.

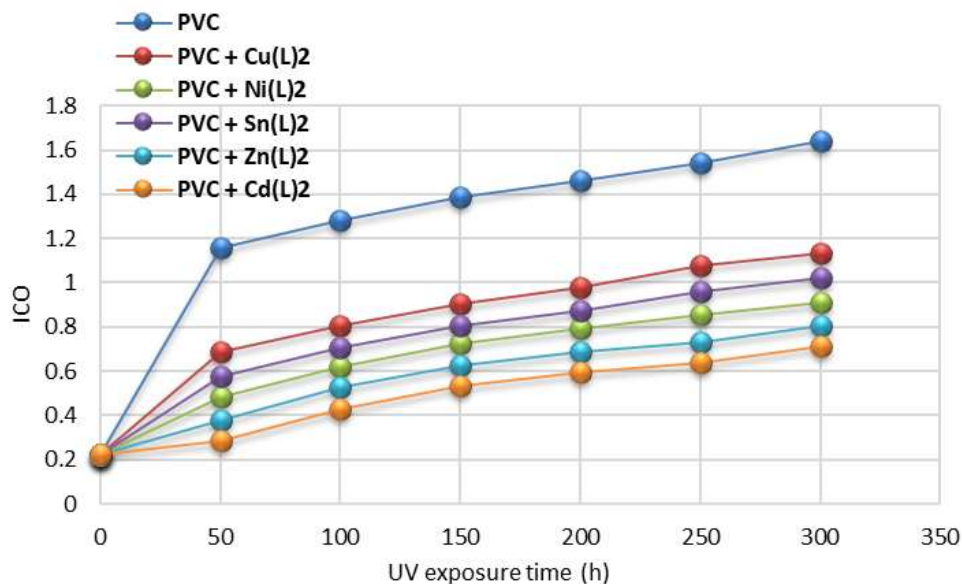


Figure 2. Alterations in the $I_{C=O}$ Indicator of PVC Films.

3.3 Average M_v

The photoirradiation of PVC induces the cleavage of small plastic fragments, leading to a reduction in the M_v . The primary factors

contributing to this decline in M_v are the chain scission and cross-linking of polymeric chains. The intrinsic viscosity $[\eta]$ is highly influenced by M_v , consequently anticipating a reduction in

the solution viscosity of irradiated PVC films [24].

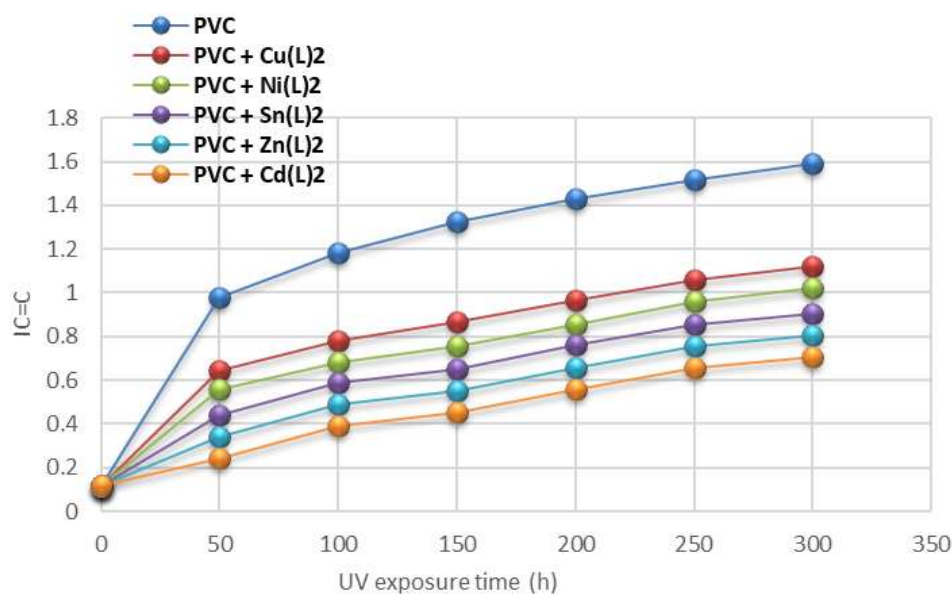


Figure 3. Variations in the $I_{C=C}$ Indicator of PVC Films.

To assess the impact of irradiation on M_v , PVC films irradiated for varying durations were dissolved in THF, and their viscosities were estimated. The M_v was calculated via Equation (2) and plotted versus irradiation time (50–300 h; Figure 4). The presence of insoluble residues indicated branching and cross-linking during the irradiation process. The M_v exhibited a sharp and continuous decrease with increasing

irradiation time. Notably, all metal complexes consistently mitigated the reduction in M_v . The metal complexes demonstrated a remarkable ability to alleviate the depression in M_v of PVC, providing a high degree of stabilization. Among them, the Cd(II) complex emerged as the most effective PVC photostabilizer compared to others.

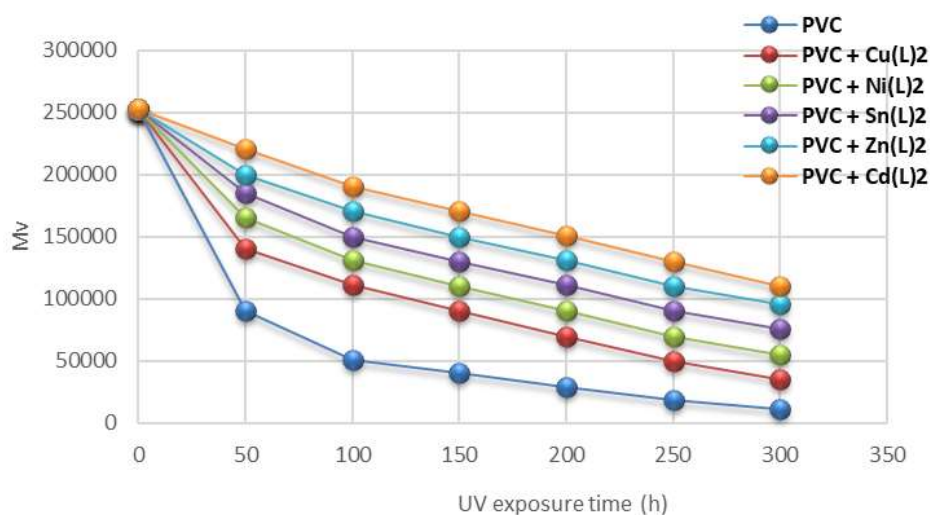


Figure 4. Fluctuations in the M_v Parameter for PVC Films.

3.4. Scanning Electron Microscopy (SEM)

SEM serves as a powerful technique for examining the surface morphology of polymers,

offering insights into their internal structures [26]. Electron beams enable the capture of precise, magnified images of the PVC surface.

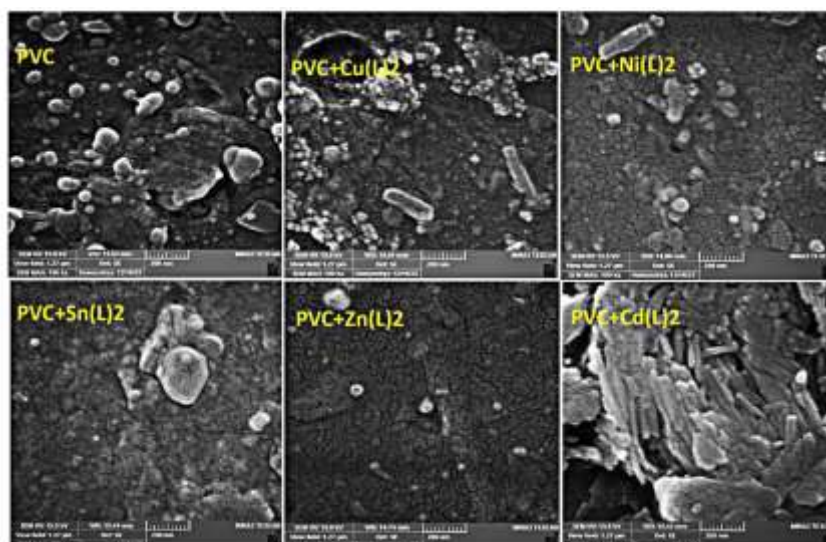
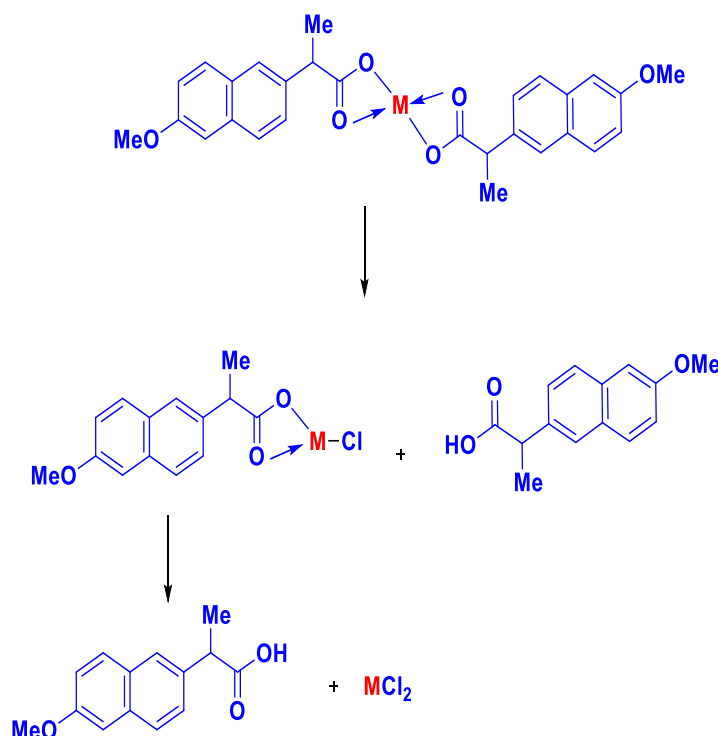


Figure 5. SEM images for PVC films

The introduction of metal complexes into the PVC induced an overall alteration in particle size and their ununiform distribution on the surface after 300 hours of UV irradiation. SEM images of the PVC films revealed a notable rough surface post-irradiation, indicating a noticeable alteration in PVC surface

morphology. Images of PVC blended with various metal complexes illustrated that irradiation had only a minimal impact on the compact texture, which is clarified in Figure 5. The films containing additives exhibited high resistance to irradiation, demonstrating excellent chemical stability in the blends.



Scheme 2. Proposed Mechanism for Photostabilization of $M(L)_2$ Complexes as HCl Scavengers.

3.5. Proposed mechanisms for PVC photostabilization

Several proposed mechanisms for the photostabilization of polyvinyl chloride (PVC) have been elucidated in the literature. The efficacy of Bis[2-(6-methoxynaphthalen-2-yl)propanoate] complexes as stabilizers for PVC sheets can be evaluated by analyzing changes in concentration indices, as supported by extensive documented data. These complexes act by forming covalent carboxylates, which in turn stabilize PVC through mechanisms reliant upon the strong Lewis acidic properties of the associated metals. Furthermore, these stabilizers exhibit the additional function of scavenging hydrogen chloride (HCl) and facilitating the replacement of allylic chlorine atoms with carboxylate groups [25]. Typically classified as secondary stabilizers, they demonstrate

exceptional long-term stability, as depicted in Scheme 2.

Metal carboxylates serve as effective substitutes for allylic chlorine in PVC stabilization, as suggested by infrared spectroscopic analyses indicating their interaction with PVC molecules at the main particle morphology [25]. This pivotal role in the stabilization process designates these complexes as primary stabilizers.

3.6. The quantum (Φ_{cs}) yield of PVC films' chain scission

The quantum yield of chain scission (Φ_{cs}) serves as an alternative method for quantifying degradation processes. The quantum yield for chain scission in modified PVC films can be calculated using the equation provided in [26]. The corresponding Φ_{cs} values for these polymers are presented in Table 1.

Table 1. Quantum yield (Φ_{cs}) for the chain scission for 40 μm thick PVC sheets after 300 h irradiation time before and after filling the complexes

Additive	The quantum yield of main chain scission (Φ_{cs})
PVC	5.14E-05
Zn(L) ₂	1.46E-07
Sn(L) ₂	1.35E-07
Ni(L) ₂	2.88E-07
Cu(L) ₂	3.71E-07
Cd(L) ₂	1.28E-07

The relatively low values of Φ_{cs} can be attributed to the phenomenon of energy absorption at a single site followed by dispersion of electronic excitation across numerous bonds within the extensive PVC molecule. This dispersion mechanism diminishes the probability of individual bond breakage or may result in the dissipation of absorbed energy through non-reactive pathways. It is well-established that elevated temperatures often correlate with increased Φ_{cs} values,

particularly in proximity to the glass transition temperature (T_g) for amorphous polymers and the melting point for crystalline ones [26]. In the context of this study, PVC film undergoes photolysis at temperatures ranging between 25 and 40 °C, which notably falls below the T_g of PVC (80 °C). Consequently, it is not anticipated that temperature will exert a significant influence on Φ_{cs} values in this experimental setup.

4. Conclusions

In the present study, the photostabilization of PVC sheets employing Bis[2-(6-methoxynaphthalen-2-yl)propanoate] complexes was investigated. These additives exhibited notable efficacy as photostabilizers for PVC films, primarily functioning through effective

HCl scavenging mechanisms. Notably, among these additives, the cadmium complexes emerged as particularly efficient in the photostabilization process, as evidenced by the observed enhancement in photostability and the elucidated underlying mechanism. This finding

underscores the potential utility of cadmium stabilizers in PVC applications. complexes as viable candidates for commercial

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Conflict of Interest

The authors declare there is no known conflict for this work.

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POLİVİNİLKLORİDİN FONKSİYONAL DAYANIQLILİĞİNİ ARTIRMAQ ÜÇÜN İNNOVATİV METAL KOMPLEKSLƏRİNİN FOTOSTABİLİZATOR KİMİ TƏTBİQİ

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Xülasə: Polivinilxlorid (PVX) gündəlik həyatımıza əhəmiyyətli dərəcədə təsir edən müxtəlif tətbiqlərə malikdir və sənaye üsulu ilə müxtəlif konfigurasiyalarda sintez olunur. Bununla belə, PVX fotodeqradasiyaya uğrayaraq həm ətraf mühitə, həm də insan sağlamlığına zərərli əlavə məhsullar əmələ gətirir və bununla da onun yararlılığını azaldır. Bunları nəzərə alaraq, işdə PVX-in fotodeqradasiyasını azaltmaq məqsədilə metal komplekslərin potensial fotostabilizatorlar kimi tətbiqi araşdırılmışdır. Fotodeqradasiyanın qarşısının alınmasında polimerin molekulyar kütləsinin azalması, alken yüklü, karbonil fraqmentlərinin formalaşması və şüalanmaya məruz qalan PVX təbəqələrinin səth morfoloqiyası kimi parametrlərin araşdırılmasında metal komplekslərinin effektivliyi tədqiq edilmişdir. Müəyyən edilmişdir ki, bu innovativ metal kompleksləri polivinilxloridin fotodeqradasiyasını azaltmaqda effektivlik nümayiş etdirir və beləliklə, onun funksional uzunömürlüklüyünü artırmaq üçün perspektivli ola bilər.

Açar sözləri: polivinilxlorid; metal kompleksləri; fotodeqradasiya; səth morfoloqiyası; molekulyar çəkinin azalması

ИННОВАЦИОННЫЕ МЕТАЛЛОКОМПЛЕКСЫ КАК ФОТОСТАБИЛИЗАТОРЫ ДЛЯ ПОВЫШЕНИЯ ФУНКЦИОНАЛЬНОЙ ДОЛГОВЕЧНОСТИ ПОЛИВИНИЛХЛОРИДА

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Абстракт: Поливинилхлорид (ПВХ) находит разнообразное применение, существенно влияющее на нашу повседневную жизнь, и промышленно синтезируется в различных формах. Однако ПВХ подвергается фоторазложению, образуя токсичные побочные продукты, вредные как для окружающей среды, так и для здоровья человека, тем самым сокращая срок его службы. Чтобы решить эту проблему, в настоящей работе изучено применение металлокомплексов в качестве потенциальных фотостабилизаторов для смягчения фоторазложения ПВХ. Это исследование углубляется в эффективность недавно разработанных комплексов металлов в смягчении фотодегradации, изучая такие параметры, как ослабление молекулярной массы, образование алкен-нагруженных и карбонильных фрагментов, а также изменения морфологии поверхности листов ПВХ, подвергнутых облучению. Результаты исследований показывают, что эти инновационные металлокомплексы демонстрируют эффективность в уменьшении фотодегradации поливинилхлорида, тем самым представляя многообещающий путь к продлению его функциональной долговечности.

Ключевые слова: поливинилхлорид; металлокомплексы; фотодегradация; морфология поверхности; снижение молекулярной массы