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Amino- and Diamino-9,10-Anthracenedione Derivatives: Biofocus and Applied Advantages - A Mini-Review

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Abstract: 9,10-Anthracenediones an important class among quinonic compounds of natural and synthetic origin with many applications. Amino- and diaminosubstituted 9,10-anthracenediones are one of the main classes among 9,10-anthracenedione derivatives. Primarily, they are the key structures for obtaining various dyes. Besides applying their dye properties, aminoanthracenediones became the objects for searching for new biologically active compounds with antibacterial, antifungal, anticancer, antioxidant, antiviral, immunostimulating, and antiprotozoal activity in medical chemistry. However, amino- and diamino-substituted 9,10-anthracenediones are widely used as analytical reagents, color, and metal indicators. Areas of the practical application of these compounds are constantly expanding. This mini-review dedicates the advantages of amino- and diamino-9,10-anthracenediones derivatives as biologically active compounds and substances with applied application in various fields.

Keywords: amino- and diamino-9,10-anthracenedione derivatives; antimicrobial activity; anticancer activity; antiviral activity; antiplatelet activity; applied advantages.

1. Introduction

The widespread application of 9,10-anthcenedione derivatives and the peculiarity of their properties attract the attention of specialists and researchers in various fields of science and technology. This is evidenced by hundreds of publications that are published annually in scientific and patent literature. Amino- and diamino derivatives occupy one of the main places among 9,10-anthracenedione derivatives, and they are key compounds in the synthesis of a number of dyes, pigments, and phosphors. However, in addition to the traditional using of these compounds' coloring properties, they gradually became objects for several studies their pharmacological studies. The discovery of antitumor properties for several diamino-9,10-anthracenediones and introducing the drug Mitoxantrone into medical practice was the impetus. Various amino- and aminohydroxyanthracenediones are also used as analytical reagents for photometric determination of different ions of metals, color reactions, metal indicators in complexometric titration. Many works are devoted to using amino and diaminopohydric 9,10-anthracenediones for the preparation of dyes [1]. There are several reviews on the antitumor activity of 9,10-anthracenedione derivatives [2-5]. However, there is no generalized review of the biological properties and applications of the amino derivatives of 9,10-anthracenedione. Therefore, this mini-review will focus on coverage and compilation of

results on the advantages of amino- and diamino-9,10-anthraquinone derivatives as biologically active compounds and substances with applied application in various fields.

2. Amino-9,10-anthraquinones and their Derivatives as Biologically Active Substances

The ability to bind to the estrogen α-receptor (ERRα) was found for 1(2)-amino- 1, 2, 1,4 (1,5)-diamino- 3, 4 (Fig. 1), 1-amino-2 (4/2,4)-halogen-substituted 9,10-anthracenediones [6]. 2-Aminoanthraquinone 2, due to its ability to inhibit glutathione reductase, has been identified as a potential antimalarial substance [7], as an inhibitor of xanthine oxidase [8] and bacterial collagenase [9], as well as a suppressor of the formation of sulfides by sulfate-reductive bacteria in wastewater [10, 11], wells, process tanks for biomass fermentation [12]. 1,4-Diamino-9,10-anthracendione 3 and its derivatives showed positive results on the fixation of lactate dehydrogenase [13].

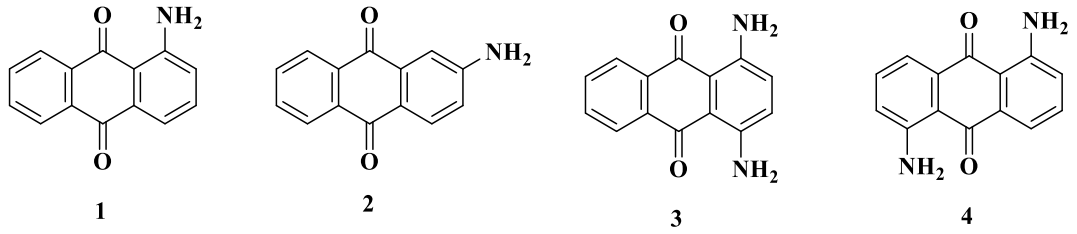

Figure 1. Mono- and diaminoanthraquinones in the biological application.

Several *N*-[(2-(2-acetamido-6-R-benzothiazol-2-yl]-1-amino-9,10-anthracenediones 5 showed *in vitro* experiments a fairly high antibacterial activity against bacteria *Streptococcuss pneumoniae, Enterococcus faecalis, Salmonella typhi, Acinetobacte sp.* [14]. The compounds with the ability to inhibit the growth of bacterial strains *Staphylococcus aureus, Mycobacterium luteum* and fungi *Candida tenuis, Aspergillus niger* were identified among the amino acid derivatives of 2-[chloro-*N*-(9,10-dioxo-9,10-dihydroanthracen-1-yl)acetamide 6 [15] and 2-dithiocarbamate-*N*-(9,10-dioxo-9,10-dihydroanthracenyl)acetamides 7 [16] (Fig.1). The inhibitory effect on the growth of bacterial strains *Bacillus subtilis, Bacillus cereus, Acinetobacter johnsonii, Sarcina lutea, Xanthomonas oryzae* and fungus *Saccharomyces cerevisiae, Candida lipolytica* was found for the pyridine salt of 2-[chloro-*N*-(9,10-dioxo-9,10-dihydroanthracen-2-yl)acetamide 8 [17].

The compounds with high indices of biological action against strains of gram-positive bacteria *Staphylococcus aureus, Enterococcus faecium, Bacillus subtilis* and the fungus *Candida albicans, Aspirgillus niger, Trichophyton mentagrophytes were* found among β-lactam derivatives 9 obtained based on 2-amino-9,10-dioxoanthracene [18]. The effective anti-staphylococcal agents against strains *Staphylococcus aureus ATCC 25923* та *Staphylococcus aureus ATCC 29213* were found among anthra[1,2-*d*][1,2,3]triazine-4,7,12(3*H*)-triones 10 (Fig. 2) [19]. The compounds with selective antibacterial activity against *Staphylococcus aureus, Escherichia coli, Bacillus subtilis,* and antifungal effect against *Alternaria alternata* were found in a series of oxadiazole and azole derivatives of 9,10-anthracenedione [20].

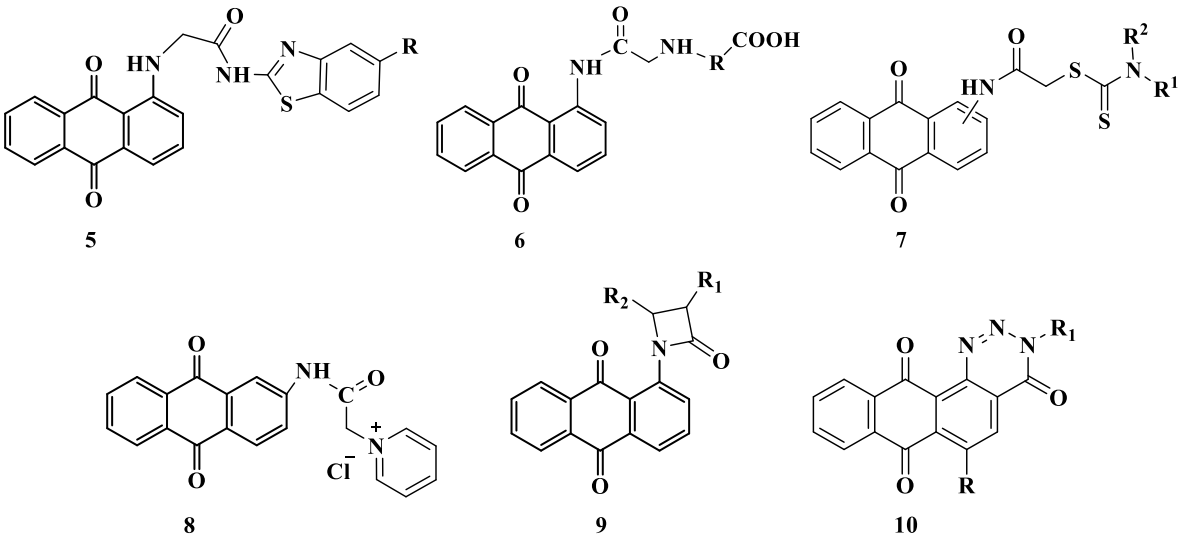

Figure 2. Amino-9,10-anthraquinone derivatives with antimicrobial activity.

Derivatives of 9,10-anthracenedione substituted with 1,5-bis(diethylaminoethyl)- and 2,6-bis(dimethylaminoacetylamino) fragments 11 (Fig. 3) showed antiviral activity against *HIV-1* [21, 22]. 1-Amino-4-arylamino-9,10-anthracenediones can act as inhibitors of human cytomegavirus *HCMV* [23, 24], and 1,2- and 1,4-bisadamantanylanthracenediones have been proposed as potential anti-influenza agents due to their ability to stimulate interferon production [25]. The derivatives with antiviral activity against herpes virus *HSV-2* and Ebstein-Barr virus in cell cultures *Raji, B95-8* were found among the dithiocarbamates 9,10-anthracenedione 12, 13 (Fig. 3), obtained by the coupling reaction of the diazonium salts of aminoanthracendiones with dithiocarboxylic acids [26].

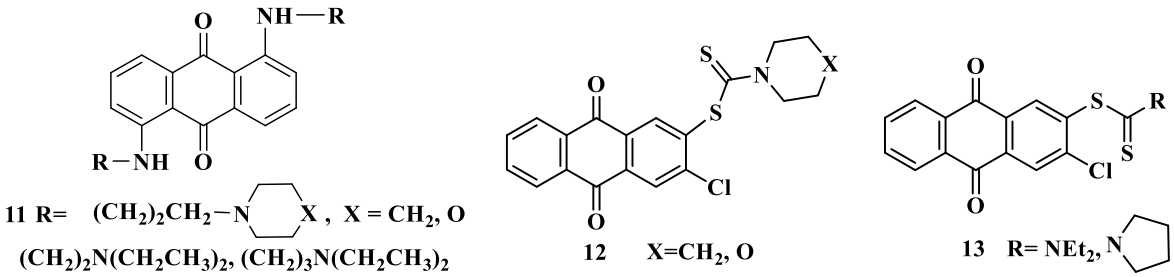

Figure 3. Amino-9,10-anthraquinone derivatives with antiviral action.

In the same series of dithiocarbamate derivatives with 9,10-anthracenediones [27] obtained from 2-amino-9,10-anthracenediones, two compounds 14 and 15 (Fig. 4) with high antiaggregatory activity in *in vitro* experiments with IC_{50} within 15-30 μm for adenosine diphosphate-dependent aggregation and 10-20 μm for arachidonic acid-induced aggregation were found.

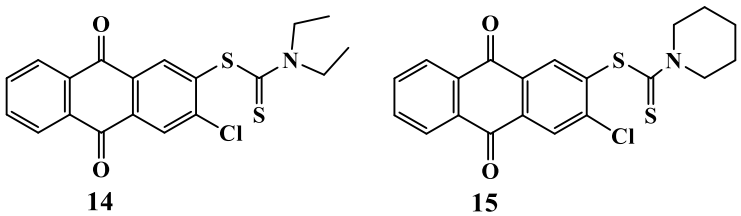

Figure 4. Amino-9,10-anthraquinone derivatives with the antiplatelet action

The indicators of lipid peroxidation and oxidative modification of the protein were determined by studies of several amino acids and 2-iminothiazole derivatives of 9,10-anthracenedione compounds 16-18 with antioxidant effect were found [28]. The hydrazone derivatives of 9,10-anthracenedione 19-21 (Fig. 5) with the highest antioxidant activity were determined by the CUPRAC method [29]. Also, 5-chloro-substituted 1-amino-9,10-anthracenediones 22 with antioxidant effect were detected in work [30] by DPPH and ABTS.

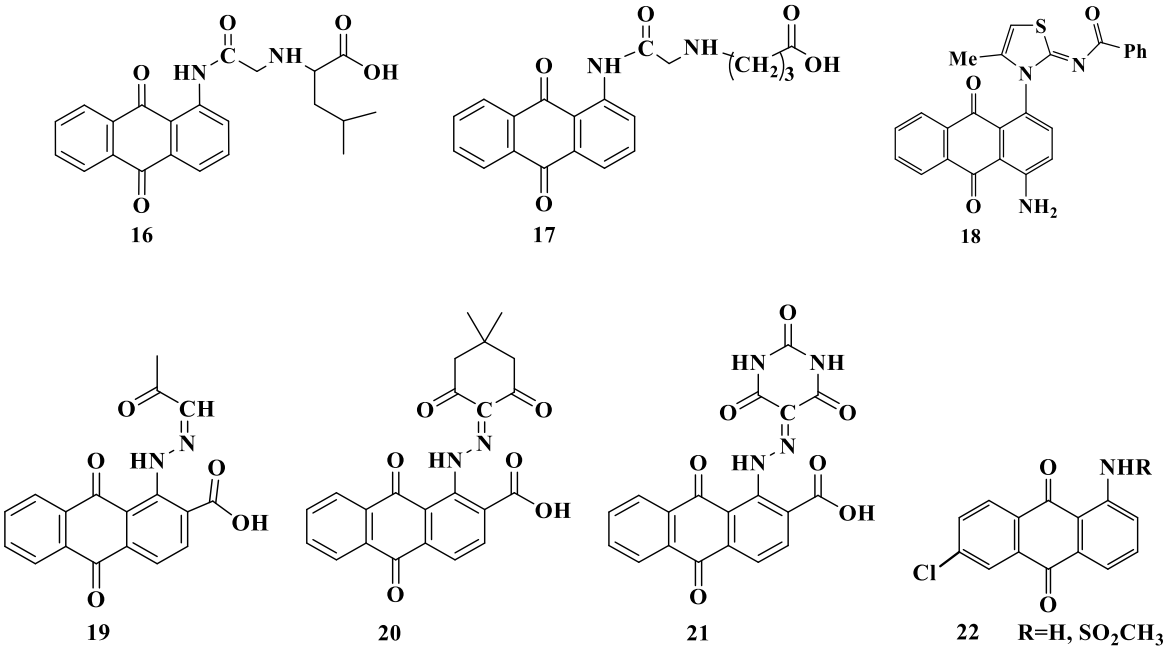

Figure 5. Amino-9,10-anthraquinone derivatives with antioxidant effect.

Schiff bases obtained from 1-amino-9,10-anthracenedione have been proposed as a new type of analgesic substance [31]. The compounds with anti-inflammatory effects were found among derivatives 1(2)-amino-9,10-anthracenediones functionalized with carbocyclic moiety [32-36].

Derivative 23 with activity against *Entamoeba histolytica* was found among the bisamidines of 2,6-diamino-9,10-anthracenedione [37, 38]. Studies of an order of derivatives 24 allowed to classify 6-diacetaminoalkylenamines as agents with action against *Trichomonas vaginalis*, *Trichomonasfetus* та *Entamoeba histolytica* (Fig. 6) [39].

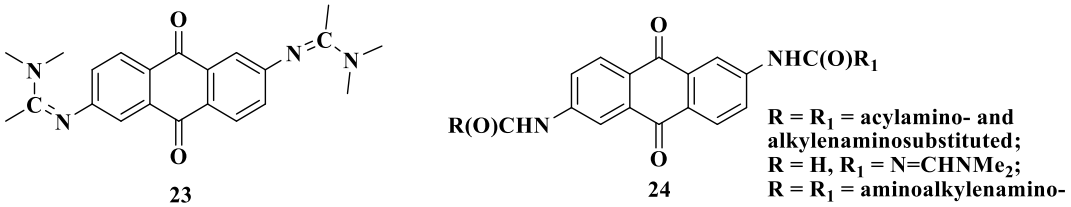

Figure 6. Amino-9,10-anthraquinone derivatives with antiprotozoic effect.

Besides the types mentioned above of pharmacological effects, special attention deserves derivatives 1(2)-amino-9,10-anthracenediones as an antineoplastic compound with a broad spectrum of activity against various types of cancer [3]. Mitoxantrone 25 and ametantrone 26 (Fig. 7) can be called the "progenitors" of the antitumor alkylamino-substituted 9,10-anthracenediones. In particular, mitoxantrone is one of the most widely used and effective chemotherapeutic agents used in breast cancer treatment. It is also used to treat lymphoma,

acute leukemia, chronic myelogenous leukemia, hepatocellular carcinoma [39]. In 2000, the drug was approved by the agency FDA (USA), has shown promising results in the treatment of multiple sclerosis [2] and acute disseminated encephalomyelitis [40]. Its analog pixantrone 27 is a licensed anticancer drug in Europe and is used as a monotherapeutic agent to treat aggressive resistant cases of non-Hodgkin's B-cell lymphoma (Fig. 7) [41].

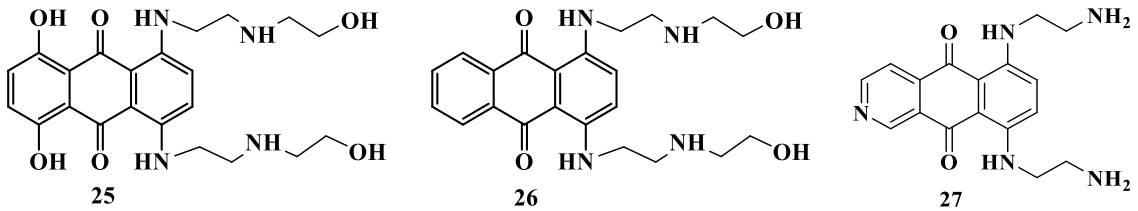

Figure 7. Amino-9,10-anthraquinone derivatives as anticancer drugs.

The discovery of several antitumor substances that are at different stages of preclinical testing was the result of successful structural modification of the following aminoanthracendiones: 1-aminoethylamino-, 1,4(1,5)-bis-(2-aminoethylamino)-, bis(oxyalkylamino)-, 1,5-bis/(*N*,*N*-dialkylamino)alkylamino/-9,10-anthracenediones and complexes of 1,4-bis(alkylamino)-5,8-dioxoanthracene-9,10-dione with metals [42-45]. In particular, such aminoanthracendiones *NSC-639365* 28, *M-18* 29, and *Banoxantron* 30 (Fig. 8) are in different phases of clinical trials [46].

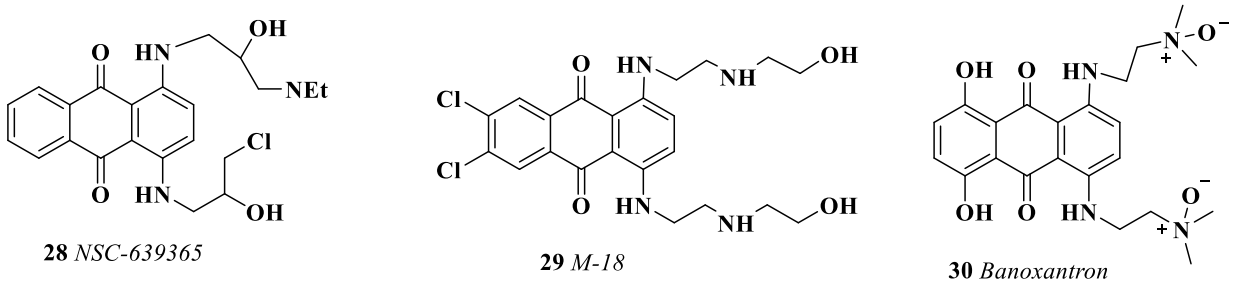

Figure 8. Amino-9,10-anthraquinone derivatives in clinical investigations

Analysis of the data of the structure-activity dependence in a series of asymmetric derivatives of 1,2-diaminoanthracenedionivs allowed obtaining compounds of a new chemotype 31-34 with high cytotoxicity concerning the majority of *NCI-60* cancer cell lines, which were patented (Fig. 9) [47].

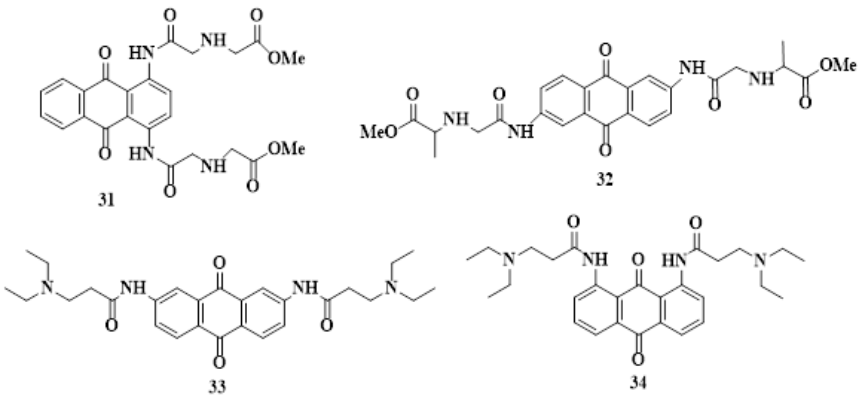

Figure 9. Amino-9,10-anthraquinone derivatives with anticancer activity.

Antitumor compounds with immunostimulatory action were found among 3-amino-1,2-alkoxy derivatives 35 [48], and for 1,8-diamino-3-methyl-9,10-anthracenedione and their *N*-alkyl/aryl derivatives 36 radioprotective properties were found [49], which is important for radiation therapy (Fig. 10).

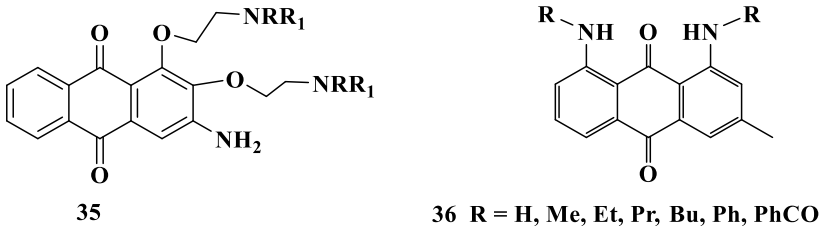

Figure 10. Amino-9,10-anthraquinone derivatives with anticancer, immunostimulating, and radioprotective properties.

Generalized advances in the development of antitumor substances based on aminoanthracendiones in recent decades are presented in two reviews [2, 3].

The amino derivative of 9,10-anthracenedione with a 1,3,5-triazine moiety under the trade name *Reactive Blue 2* 37 (Fig. 11) has been found useful as a ligand in biochemical processes experimental pharmacology, and the development of new biologically active substances [2]. 9,10-Dioxoanthracenylamide derivative of ethacrynic acid 38 (Fig. 11) can act as an antagonist of Wnt/β-catenin signaling and cell survival *CLL* [50].

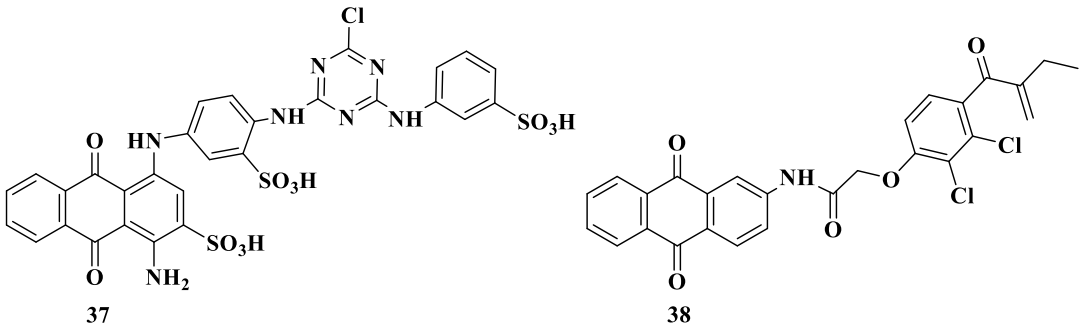

Figure 11. Amino-9,10-anthraquinone derivatives in biochemical processes.

Also, 1-amino- 1 and 2-amino-9,10-anthracenediones 2 are convenient promoters in the biotransformation of biphenol *F* with the participation of *Pseudomonas sp. HS-2* [51]. 1-Amino-4-chloroanthracenedione, 1,4-diaminoanthracenedione, 1,5-diaminoanthracenedione, 1-amino-2-methylanthracenedione, 1-amino-2-carboxyanthracenedione, 1-amino-2-chloroanthracenedione [52], and 1-amino-4-chloroanthracenedione [52, 53] were used as objects for the investigation of the biotransformation ability of *Saccharopolyspora erythraea* and *Saccharothrix espanaensis*, respectively.

3. Applied Advantages of Amino-9,10-anthraquinones and their Derivatives

Aminoanthracendiones and their derivatives occupy one of the main places in obtaining several dyes [1], which are widely used in various sectors of the industries.

A new fluorescent paper with enhanced fluorescent photoinduced electron transfer for Mn^{2+}, Cr^{3+} та F^- was proposed based on 1-amino-9,10-anthracenedione structurally combined with calix[4]arene [54].

2-Amino-9,10-anthracendione has been successfully used to produce a highly efficient supercapacitor electrode based on chemically modified graphene hydrogel [55] and in the production of ordered ensembles of organic and biological molecules on Au-bonded surfaces [56].

Graphene-anthracenedione composite surfaces with grafted plastic based on 1-amino-2-bromo-4-hydroxy- and 2-amino-9,10-anthracenediones, have been patented as important agents in wastewater treatment polluted with azo dyes and nitrates [57]. A convenient method of dechlorination and biodegradation of 2-chloroanilines under anaerobic conditions was proposed using 2-amino-9,10-anthracenedione-graphene oxide composite [58]. 1,4-Bis(*p*-tolylamino)anthracene-9,10-dione was useful in obtaining a polymer for ophthalmic cosmetic purposes [59].

1-Amino-, 1-(methylamino)- and 1-(benzamido)-9,10-anthracenediones exhibited the properties of cationic photoinitiators of polymerization of epoxy monomers and divinyl ether under the influence of LEDs [60]. 1-Amino-4-hydroxy-, 1,4-diamino- and 1,5-diamino-9,10-anthracenediones have been successfully used in multicolor photoinitiator systems [61].

The low-cost, safe flow-through battery system with stable charging and discharging cycles based on 1(2)-amine derivatives of 9,10-anthracendione has been patented [62]. 1,4-Bis((2-(2-(2-methoxyethoxy)ethoxy)ethyl)amino)anthracene-9,10-dione for use in symmetric all-organic redox flow batteries that can store energy on a large scale without relying on critical raw materials [63].

The method of obtaining a conjugated redox poly-*N*-pyrrolylantracenedione polymer to obtain an aqueous all-organic hybrid-flow battery based on 9,10-dioxoanthracenyl-2-pyrrole obtained from 2-amino-9,10-anthracenedione [64] was carried out [65].

Also, photoinitiating systems for the free radical photopolymerization of various acrylate monomers and fast 3D printing under visible light were developed using 1-amino-4-hydroxyanthraquinone, 1,4-diaminoanthraquinone, and 1,5-diaminoanthraquinone [66].

The supramolecular interaction of β-cyclodextrins and *N*-alkylaminoanthrcenedione was used to obtain self-assembly fluorescent vesicles as a new kind of fluorescence staining material for living cells, for example, the human breast cancer cells *MCF-7* and mice mononuclear macrophages *REW-264.7* [67]. The obtained experimental data allowed to propose the plausible mechanism of the self-assembly fluorescent vesicles forming [67].

N'-(9,10-Dioxo-9,10-dihydroanthracen-2-yl)-1-glycylpyrrolidine-2-carbohydrazide 39 was used as a new compound with fluorogenic properties for demonstration dipeptidyl peptidase IV(DPP IV) in the detection of the enzyme using the histochemical study (Fig. 12). [68].

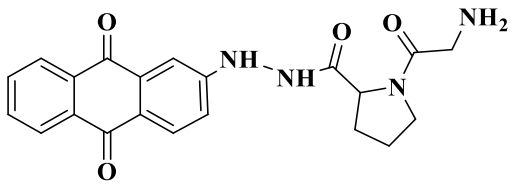

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Figure 12. *N'*-(9,10-Dioxo-9,10-dihydroanthracen-2-yl)-1-glycylpyrrolidine-2-carbohydrazide as fluorogenic compound for the histochemical DPP IV.

Amino- and diamino-9,10-anthracenedione derivatives 40-42 (Fig. 13) were utilized in an aromatic composition as dye components with aldehyde perfume component [69].

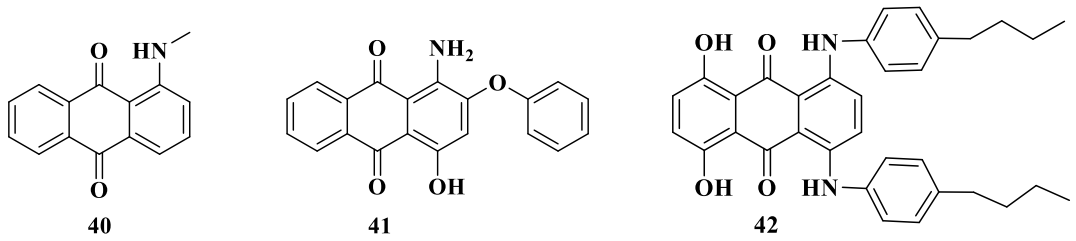

Figure 13. Amino- and diamino-9,10-anthracenedione derivatives as colorant component in an aromatic composition.

Moreover, amino derivatives of 9,10-anthracenedione 41, 43-46 (Fig. 14) found their application as a component of the dimethrin-containing carrier in the obtaining of the mosquito-repellent fibers with long-acting properties [70].

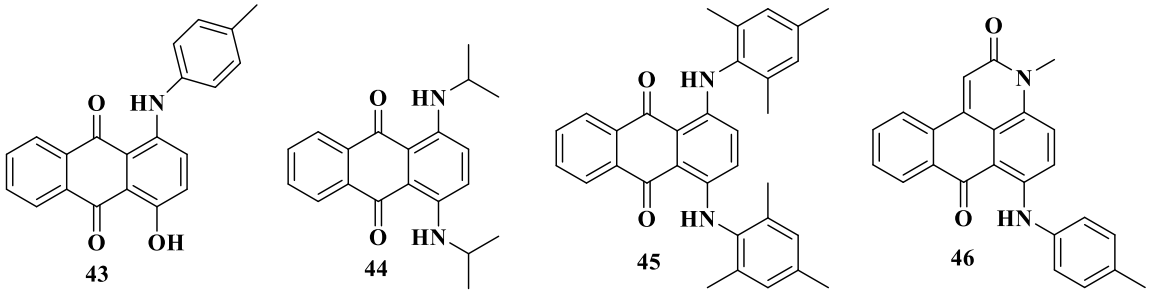

Figure 14. Amino- and diamino-9,10-antracenediones as components for preparation of mosquito-repellent fibers.

4. Conclusions

Amino- and diamino-9,10-anthracenediones and their derivatives are compounds with strong synthetic, pharmacological and practical potential, which do not lose their relevance to this day. They occupy one of the main places in obtaining several dyes, various reagents for analytical needs, organic chemosensors, and catalysts. Amino derivatives of 9,10-dioxoanthracene have enriched the arsenal of biologically active substances, including compounds with antitumor, antiprotozoal, antiviral, antimicrobial, anti-inflammatory action. Some of them are known as antitumor drugs (mitoxantrone, ametantrone), which are widely used in world chemotherapeutic practice, and several derivatives are in the II and III phases of clinical trials. It should be noted that 1(2)-amino- and 1,4(1,5)-diamino-9,10-dioxoanthracenes are key substrates in obtaining almost all 9,10-anthracenedione dyes and pigments, as well as bioactive derivatives, in particular with antimicrobial and antitumor activity. Therefore, directed structural modification of amino groups of the 9,10-anthracenedione ring by various pharmacophore fragments remains the most interesting and promising direction among the production and study of new derivatives 9,10-anthracenedione, which is determined by the needs of both medical chemistries and applied using these class of compounds.

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This research has no acknowledgment.

Conflicts of Interest

The authors declare no conflict of interest.

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