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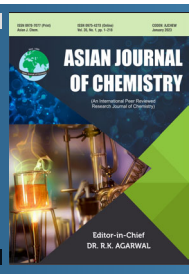

REVIEW

A Comprehensive Review for Ruthenium(II) Complexes in Photodynamic Therapy

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Photodynamic therapy (PDT) has emerged as a promising therapeutic approach for the treatment of various diseases, including cancer. In recent years, ruthenium(II) complexes have garnered significant interests as photsensitizers in PDT due to their unique photophysical properties and versatile coordination chemistry. The current developments in the use of Ru(II) complexes as photosensitizers in PDT are thoroughly examined in this review article. The design techniques used to increase their photodynamic effectiveness, increase selectivity and reduce off-target impacts are discussed. The formation of reactive oxygen species (ROS) and the ways in which they influence biological processes are also being explored. Additionally, the Ru(II) complexes with dual functioning, targeting abilities and uses in various disease models are highlighted. Prospects and challenges in this area such as water solubility, biocompatibility and translational issues are discussed. The goal of this review article is to provide a thorough overview of the potential of Ru(II) complexes in PDT and to inspire additional investigation in this fascinating field.

Keywords: Photodynamic therapy, Ruthenium(II) Complexes, Photosensitizers, Localized cytotoxity.

INTRODUCTION

Novel therapeutic strategy known as photodynamic therapy (PDT) has received a lot of attention recently due to its potential to treat a variety of ailments [1-4]. It includes causing localized cytotoxicity and tissue damage in selected locations using photosensitizing chemicals, light at particular wavelengths and molecular oxygen [5-7]. PDT is a non-invasive, extremely selective and potentially curative therapy option for a number of illnesses, including cancer, bacterial infections and specific dermatological problems [8-10]. In photodynamic treatment (PDT), photosensitizers are essential because they are the elements that trigger the healing process [11,12]. They are substances or molecules with the capacity to take in light energy and transmit it to nearby molecules, producing reactive oxygen species (ROS) [13-16]. These ROS especially singlet oxygen ($^1O_2$) are extremely reactive and can cause cytotoxicity and tissue damage in the targeted location [17-21]. The formation of reactive oxygen species (ROS) and the ways in which they influence biological processes are also being explored [22,23]. Ideal photosensitizers should have qualities such as high absorption coefficients in the relevant wavelength range, great photostability and preferential accumulation in target tissues [24-26].

Ruthenium(II) complexes have shown promise as photosensitizers in photodynamic treatment (PDT), due to a number of unique benefits that make them especially well-suited for this use [27-29]. Using Ru(II) complexes as photosensitizers in PDT has a number of benefits, including wide absorption spectrum, long excited state lifetimes and photophysical properties that are highly tunable, *i.e.* photophysical characteristics of ruthenium(II) complexes, such as the absorption and emission wavelengths, can be adjusted through careful ligand design [30-33]. Furthermore, complexes of ruthenium(II) are known to posses the exceptional chemical and photochemical stabilities [34-36]. They can tolerate continuous light exposure without suffering any significant deterioration. Ruthenium(II) complexes with dual functionality that combine PDT with other treatment modalities can also be created [37-40]. These complexes may have inherent cytotoxicity or additional properties that support their overall therapeutic actions such as DNA intercalation or enzyme inhibition [41,42]. As PDT treatments become

more effective and adaptable, ruthenium(II) complexes are being explored and getting modified in this area of study due to their unique features.

Design strategies for Ru(II) complexes in PDT

Cooordination chemistry of Ru(II) complexes: The ligands and coordination environment around the ruthenium centre can be carefully designed to optimize the characteristics of ruthenium(II) complexes for PDT [43-46]. It is possible to choose ligands based on their capacity to extend emission and absorption wavelengths, manage excited state durations and alter the complex's redox potential [47-51]. Polypyridyl and carboxylate ligands are frequently utilized in ruthenium(II) complexes. Examples of these are bipyridine and terpyridine [52-54].

Ligand design and modification strategies: The ability of ligands to absorb light can be improved by including π-conjugated systems into Ru(II) complexes [55-57]. The redox potential of Ru(II) complexes can be considerably altered by adding or subtracting electrons from the ligands [58-60]. Reactive oxygen species (ROS) can be produced more effectively and easily when the redox potential is lowered by electron withdrawing groups like cyano (-CN) or nitro (-NO_2) groups [61-64]. Hydrophilic groups, including carboxylate (-COOH) or sulfonate (-SO_3H) groups, are added to the complexes to increase their water solubility to make it easier to deliver and distribute them in aqueous environments [65,66]. The addition of peptides, antibodies or particular receptor-binding moieties as ligands can facilitate the selective formation of the complexes in cancer cells or particular disease-related biomarkers [67,68]. The addi-tion of multifunctional ligands broadens the possible uses of Ru(II) complexes in PDT and enables complementary therap-eutic strategies including DNA binding, enzyme inhibition or imaging capabilities [69-71].

Photophysical properties of Ru(II) complexes: The effectiveness of Ru(II) complexes as photosensitizers in photodynamic treatment (PDT) is greatly influenced by their photophysical properties, particularly their absorption and emission properties. The visible and near-infrared (NIR) wavelength ranges are often characterized by significant absorption by Ru(II) complexes [72,73]. The absorption bands can be red- or blue-shifted by ligands with prolonged conjugation or electron-donating/withdrawing groups, respectively [74-76].

ROS generation pathways: A crucial mechanism in the photodynamic treatment (PDT) process, which is mediated by Ru(II) complexes, is the formation of reactive oxygen species (ROS) [77-80]. These complexes can go through photochemical processes upon light activation, producing a variety of ROS species. The production of radical species can come from the excited-state Ru(II) complex transferring an electron to a neighbouring substrate or biomolecule [81-84]. The superoxide anion radical ($O_2^{\bullet-}$) can then be produced by these radical species when they interact with molecular oxygen (O_2) [85-88] or in an alternate pathway singlet oxygen ($^1O_2$) is created when the excited-state Ru(II) complex directly transfers its energy to molecular oxygen (O_2) [89-91]. The excited complex transferring its energy to ground-state molecular oxygen through an energy transfer mechanism produces the extremely reactive singlet oxygen [92-94].

Dual-functionality and targeting approaches

Ru(II) complexes with inherent cytotxicity: Ruthenium(II) complexes are flexible cancer treatment agents because they can serve as both photosensitizers in photodynamic therapy (PDT) and have inherent cytotoxicity [95-98]. Ru(II) complexes can target numerous cellular elements and active sites to produce cytotoxic effects through a variety of methods. Through intercalation or groove binding, Ru(II) complexes can attach to DNA and cause damage as well as obstruct DNA replication and transcription [69,99,100]. Specific proteins involved in vital biological pathways can also interact with certain Ru(II) complexes, causing inhibition or dysregulation [101-103]. Due to the inherent cytotoxicity of Ru(II) complexes and the photosensitizer effects of these compounds, PDT treatment outcomes may be improved [104-106].

Fig. 1 represents some of the most studied Ru(II) photosensitizers [107-111]. The cellular absorption efficiency, intracellular localization and even toxicity mechanism of Ru(II) complexes can be adjusted by modifying their complex characteristics such as hydrophilicity, charge distribution or steric hindrance. Compound (**a**) with a net charge of +2 aggregated in mitochondria and promoted necrotic cellular death in PDT, whereas compound (**b**) with a total charge of -4 concentrated outside mitochondria, induced cell death by apoptosis under irradiation [107]. Irradiation causes ligand ejection from strained complexes (**c** and **d**), which are then covalently attached to DNA [108]. The long-lived (π-π*) state in complex (**e**) may result in effective photo-induced DNA breakage in a relatively short amount of time [109]. Complex (**f**) was able to effectively cleave DNA due to the high NIR absorption band [110]. Highly charged Ru(II) complexes (**g**) [111] initially entered lysosomes and then pierced the nucleus, triggering cell death.

Combination therapy and synergistic effect: Combination therapy has become a viable approach in cancer therapy because of its use of numerous treatment methods concurrently or sequentially [112-114]. Ru(II) complexes can be used in conjunction with standard chemotherapy medications or radiation therapy to increase treatment effectiveness, which results in increased cancer cell killing and reduces the requirement of radiation doses, in turn minimizing radiation dose-related harm to healthy tissues [115-117]. By leveraging the body's immune system to specifically target and kill cancer cells, immunotherapy has transformed the way that cancer is treated [118,119]. To improve immune responses against cancer, Ru(II) complexes can be coupled with immunotherapeutic drugs such as immune checkpoint inhibitors or cancer vaccines [120-122]. Ru(II) complexes can be used in conjunction with photothermal therapy (PTT), a mode of treatment that uses light-absorbing substances to turn light into heat, which causes localized hyperthermia and the death of cancer cells [123-125]. Ru(II) complexes can generate heat upon light activation, when added to photothermal agents like nanoparticles or nanomaterials [126-128], which boosts the thermal effects on cancer cells. Ru(II) complexes can have synergistic effects when com-

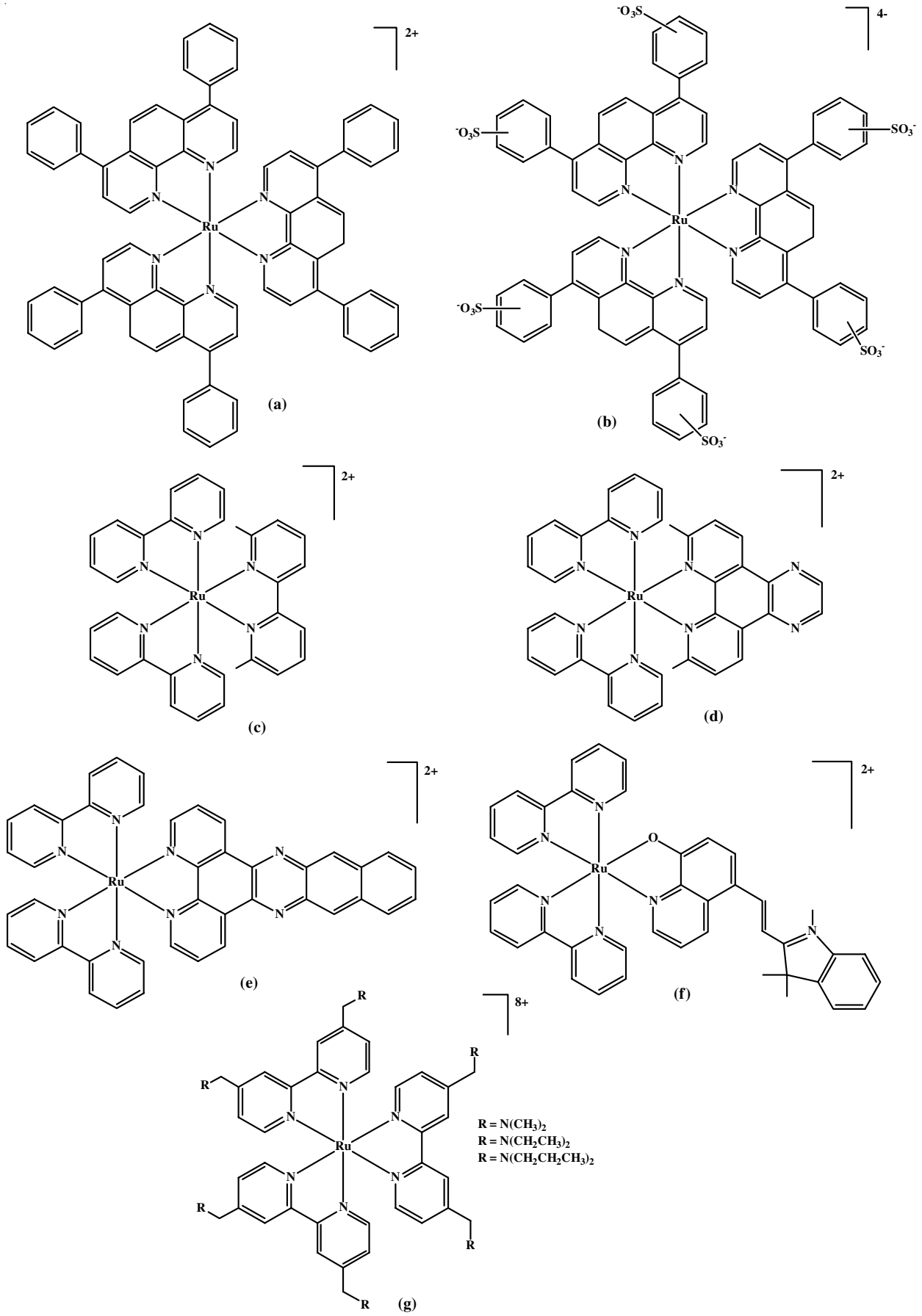

Fig. 1. Chemical structures of selected Ru(II) photosensitizers, (a,b) [107]; (c,d) [108]; (e) [109]; (f) [110]; (g) [111]

bined with PTT, combining the photothermal and cytotoxic capabilities for improved tumour ablation [129-132].

Bacterial infections and antibacterial PDT: Through a method known as antibacterial photodynamic therapy (PDT), Ru(II) complexes have demonstrated remarkable potential in the treatment of bacterial infections in addition to their uses in cancer therapy [133-136]. Antibiotic resistance has become a critical challenge and novel medicines are required to combat these illnesses [137-139]. One approach that looks very promising towards that aim is antimicrobial PDT (aPDT). Numerous studies have shown that various Ru(II) polypyridyl complexes are effective against Gram-positive and Gram-negative bacteria [140-142]. It has also been observed that bacterial biofilms provide bacteria with a safe environment and increase their resistance to antibiotics and as such, these present a substantial issue in the treatment of bacterial illnesses [143-146]. Combining Ru(II) complexes with PDT has been shown to be effective in eliminating biofilms [147-150]. The complexes' ROS can pierce and destroy the biofilm matrix, making the bacteria more susceptible to therapy [151-154].

Inflammatory disorders and immune modulation: Ru(II) complexes have recently attracted attention due to their potential use in the treatment of inflammatory diseases and immunological regulation [155-157]. Numerous immune cells, including macrophages, dendritic cells and lymphocytes have been found to be impacted by these complexes [158,159]. Pro-inflammatory cytokines including interleukin-1β, tumour necrosis factor-α (TNF-α) and interleukin-6 (IL-6) can all be inhibited by them. Inflammation can be decreased by these complexes because they have the ability to modify the activity of nuclear factor-κB (NF-κB) and mitogen-activated protein kinases (MAPKs), two important regulators of inflammatory responses [160-163].

Challenges and opportunities

Water solubility and water compatibility: Many Ru(II) complexes have low water solubility, which can limit their ability to reach target tissues [164-167]. Poor water solubility can cause aggregation, decreased bioavailability and ineffective cellular uptake. The complexes' water solubility can be increased by incorporating hydrophilic functional groups into the ligands [168-171]. Water solubility difficulties can also be solved by incorporating Ru(II) complexes into nanoparticles or by means of other delivery methods [172-174]. The complexes can be encapsulated by nanoparticles, which improve their stability, solubility and biodistribution [175,176].

Biocompatibility: A key factor to consider when using Ru(II) complexes in biomedical applications is the biocompatibility of these. Some complexes may turn out to be harmful to healthy cells or may be prone to trigger unwanted immunological responses [177-179]. It is critical to ensure that the complexes are well-tolerated by the body and do not cause unnecessary damage. As discussed earlier, potential toxicity of these complexes can be reduced by modifying the ligands which can influence the complexes' interactions with biological molecules and enhance their selectivity for target cells [180-182]. Surface polymerization or ligand functionalization of nanoparticles can improve biocompatibility and ease targeted distribution [183-186].

Conclusion

To summarize, ruthenium(II) complexes allow for multimodal applications due to their unique photophysical characteristics and diversified coordination chemistry and it leads to their appeal as a tool in the field of biological science. These complexes have the ability to deal with difficult challenges in a plethora of diseases which ranges from photodynamic therapy to antibacterial treatment, immunological modulation and much more. Though there are roadblocks to overcome with respect to water solubility and biocompatibility, the advancement in chemical innovation in tandem with multidisciplinary cooperation and cutting-edge technology certainly paves the way for a promising road ahead. With the advancement in this field, the conjugated efforts of scientists, chemists, biologists, clinicians and engineers will play a pivotal role in unlocking the true potential of Ru(II) complexes, ultimately leading to a stage where these compounds will redefine the domain of front-line medicine, opening up a new horizon of optimism and lead to a significantly improved quality of life for patients across the globe.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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