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Antimicrobial Effect of Submicron Complex Oxide Particles $CsTeMoO_6$ under Visible Light

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Abstract: The antimicrobial activity of submicron particles of new photocatalytic active complex metal oxide $CsTeMoO_6$ against bacteria *Escherichia coli* and *Staphylococcus aureus* and fungi *Aspergillus niger* and *Penicillium chrysogenum* (spores and vegetative mycelium) was studied. It has been established that $CsTeMoO_6$ has the antimicrobial activity in both under dark and visible light conditions in relation to all test cultures of microorganisms. The most inhibitory effect of $CsTeMoO_6$ was noted for *E. coli*. The light enhanced the antimicrobial effect of the test compound against all cultures of bacteria and fungi, which is associated with the presence of photocatalytic activity of $CsTeMoO_6$. The antifungal activity of $CsTeMoO_6$ increased against spores and vegetative mycelium of fungi under light condition, and this effect increased with an increasing duration of time exposure. The different degree of survival rate of the studied microorganisms in the presence of this compound (under both dark and light) may be associated with the physiological and biochemical characteristics of the used microorganisms, including different mechanisms of resistance against complex metal oxide and reactive oxygen species.

Keywords: complex metal oxide; antimicrobial properties; submicron particles; reactive oxygen species; photocatalysis

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1. Introduction

Various industrial materials and products can be subject to the negative effects of microorganisms, that is, the process of biodamage. The most significant role in such impacts belongs to bacteria and filamentous fungi [1–3]. To protect materials of both natural and synthetic origin, various organic and inorganic compounds with biocidal action are widely used [4–6].

Given the diverse and effective adaptation mechanisms, researchers need to continually improve biosafety performance. Many species of bacterial pathogens have demonstrated the ability to adapt and develop increased resistance to biocidal agents that exert constant selective pressure. A number of studies indicate an increase in the minimum inhibitory concentration of a wide range of currently used biocidal drugs in relation to various microorganisms [7]. The search for new biocidal compositions is associated with additional economic costs during production, and an increase in the concentration of existing protective agents can seriously affect the deterioration of a number of properties of materials, as well as negatively affect the environmental performance of the environment.

A number of studies have shown that nano- and micro-sized particles (ZnO, TiO_2, WO_3, etc.) are capable of suppressing the vital activity of bacteria and micromycetes. These substances are increasingly mentioned in the literature as means of protection against biodamage [8,9]. Many metal oxides increase their antimicrobial effect when exposed to light, which is due to the presence of photocatalytic properties in these compounds. Their antimicrobial activity is based on the ability to form reactive oxygen species

(ROS) under the influence of light, which have an inhibitory effect on the vital activity of microorganisms [10–15].

In addition, ROS are capable of disinfecting water and air, which makes it possible to use metal oxides not only as a means of protection against biological damage, but also as disinfectants [16,17].

It is known that depending on the shape of the particles, their size and concentration, wavelength, the illumination source used and its power, structure and chemical composition, antimicrobial activity may vary.

According to [18–21], nanoparticles of TiO_2, ZnO and other metal oxides exhibit a photocatalytic effect at a wavelength of 100–400 nm, that is, in the ultraviolet region of the spectrum or close to it. In the visible light spectrum, these compounds work poorly and their antimicrobial activity does not increase. The above shows that compounds of this series, which have photocatalytic properties when absorbing light in the visible range, can be very promising. Such compounds may be complex metal oxides, which have been little studied in this context.

In connection with the above, in the present work, we investigated of the antimicrobial activity of a new complex metal oxide ($CsTeMoO_6$) under visible light.

2. Materials and Methods

2.1. Materials Synthesis and Characterization

Complex metal oxide $CsTeMoO_6$ was synthesized at the Research Institute of Chemistry, Lobachevsky State University, Nizhny Novgorod, Russia. Average particle size of 670 nm.

The $CsTeMoO_6$ compound was obtained by the solid-state reaction. Chemically pure cesium nitrate ($CsNO_3$), tellurium oxide (TeO_2) and molybdenum oxide (MoO_3) with a molar ratio of Cs:Te:Mo = 1:1:1 were ground and calcined in a platinum crucible for 2 h in air at 400 °C to remove nitrogen oxides, after which the resulting sample was dispersed again and kept for 10 h at 700 °C. Then, the sample was abruptly cooled to room temperature. Dispersion in a planetary mill for 18 h in ethanol was used to obtain a polycrystalline sample of $CsTeMoO_6$ with a submicron particle size.

The phase composition of the tested materials was determined on the basis of X-ray diffraction analysis. The Shimadzu XRD-6100 diffractometer (Shimadzu, Kyoto, Japan) equipped with a Ni-filter (CuKα, λ = 1.5418 Å) was used in tests. Continuous scan mode of 2Θ was recorded in the range of 10° and 60° at a rate of 1°/min.

Scanning electron microscopy (SEM) imagery of metal oxide particles was performed with a JSM-IT300LV microscope (JEOL, Tokyo, Japan) with an electron-probe diameter of up to 5 nm (operating voltage 20 kV). The study of topographic surfaces was carried out using low-energy secondary and reflective electrons.

The elemental composition of the sample was studied using X-ray microprobe analysis (XMA) with an X-MaxN20 detector (Oxford Instruments, Abingdon, UK) according to Kα (O) and Lα (Cs, Te, Mo) lines.

The volumetric and quantitative particle size distribution of the resulting powder sample was determined by laser-light diffraction using a SALD-2300 analyzer (SHIMADZU, Kyoto, Japan).

2.2. Light Source Characteristics

JAZZWAY PFL-C3 LED spotlights generating light with an intensity of 325.5 W/m^2 (30 W) and 524 W/m^2 (50 W) was used as the light source. The spectrum of these light sources is the same, but they differ in intensity.

2.3. Testing the Antimicrobial Effect of Synthesized Materials on Reference Strains

Test cultures of microorganisms were two species of fungi: active biodestructors of various industrial materials *Aspergillus niger* van Tieghen VKMF-1119 and *Penicillium chrysogenum* Thom VKM F-245 (All-Russian Collection of Microorganisms, IBPM RAS, Pushchino) and two species of bacteria: *E. coli* ATCC 25922 (Gram-negative) and *S. aureus* ATCC 6538 (Gram-positive) (American Type Culture Collection, Manassas, VA, USA). The experiments used fresh cultures of microorganisms prepared from lyophilized strains. Evaluation of the antibacterial activity of various chemical compounds is performed using these bacterial strains.

A suspension of fungal spores (1×10^4 spores/mL) and bacteria (1×10^5 cells/mL for *E. coli* and 1×10^3 cells/mL for *S. aureus*) was prepared in sterile distilled water to assess the antimicrobial activity of the investigated metal oxide compound under visible light. The concentration of $CsTeMoO_6$ was 2 mg/mL.

Experimental variants with test compounds were placed in glass bottles with 10 mL of suspension of microorganisms on orbital shakers at 150 rpm, and some of them were exposed to light, while others were kept in darkness. Both bacterial and fungal spore variants were exposed for 60 min, 120 min and 180 min. Controls were used for experiment variants that did not include the test compound.

The antimicrobial activity of the $CsTeMoO_6$ on bacterial and fungal spores was assessed by the change in the titer of microorganisms after the exposure time, which was determined by the plate method (Koch method) by inoculating 0.1 mL of bacterial suspension on an nutrient agar for the cultivation of microorganisms (FBUN SRCAMB, Obolensk, Russia) and 0.1 mL of suspension of fungi on Czapek's agar (CZA) with the addition of bacteriological agar 20 g/L (Hispanagar s.a., Burgos, Spain) with subsequent accounting of the number of growing colonies of microorganisms. Chemically pure reagents were used to prepare CZA (g/l): $NaNO_3$–2.0; KH_2PO_4–0.74; K_2HPO_4–0.3; KCl–0.5; $MgSO_4 \cdot 7H_2O$–0.5; $FeSO_4 \cdot 7H_2O$–0.01; sucrose–30.0. Petri dishes with cultures of bacteria were placed in a thermostat at 37 ± 2 °C for 24 h, with cultures of fungi at 28 ± 2 °C for 72 h.

The fungal biomass were grown on a liquid (without agar) CZA medium on shakers (150 rpm) in 500 mL Erlenmeyer flask at 27 ± 2 °C for 7 days to assess the antifungal activity against vegetative mycelium. The test compound at a concentration of 2 mg/mL was resuspended in 10 mL sterile CZA with in sterile bottles, and then weighed portions of fungal mycelium (50–100 mg) were added to them. All variants were placed on orbital shakers at 150 rpm for 120 and 240 min under dark and light conditions. Variants with fungal mycelium without the test compound served as controls.

After each time of exposure, the mycelium (in all variants) from the bottles was transferred to Erlenmeyer flasks with 100 mL liquid CZA for growing for another 7 days under the same conditions. At the end of the cultivation period, the inhibitory effect of the studied substances was determined by the increase in mycelium biomass and expressed as a percentage of the control.

2.4. Statistical Analysis

All experimental results were processed using the non-parametric "U" test (Mann–Whitney) with Holm's correction in Microsoft Excel 2021 and OriginPro 2015. The results were obtained in three independent experiments. Each variant in the experiment is presented in five repetitions. Three microbiological samples were taken from each repetition for analysis. Thus, the number of independent experimental repetitions for each variant of the experiment was 15. Experimental data of microbiological studies are presented in the form of diagrams, which indicate the limits of the standard deviation.

3. Results

3.1. Characterization of $CsTeMoO_6$

The $CsTeMoO_6$ compound was obtained and described according to previous works [22,23]. The study of the crystal structure, thermal properties and electronic structure showed that it belongs to the β-pyrochlore structure type and has absorption lines inside the band gap (3.1 eV) at ~2 and 2.6 eV, which corresponds to the visible light range. The obtained electron band diagram for $CsTeMoO_6$ has been confirmed by photocatalytic experiments: absorption of photons with an energy of ~2 eV leads to the formation of electron–hole pairs, which lead to oxidation reactions of organic substances, which is shown by the example of the photodecomposition of methylene blue and methyl orange [22]. Figure 1 shows the powder X-ray diffraction pattern of the resulting $CsTeMoO_6$ sample, which can be indexed in the cubic system with the space group $Fd\bar{3}m$ [23].

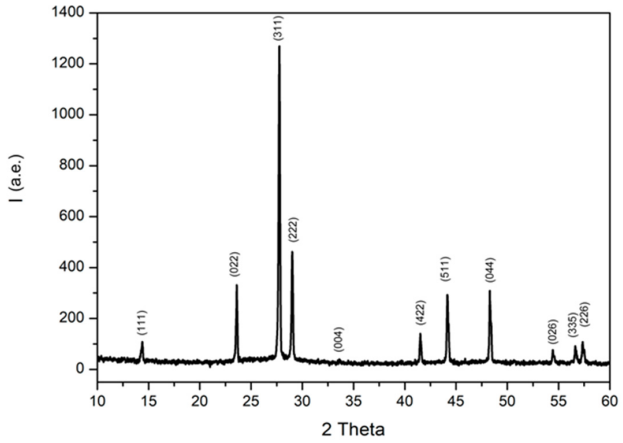


Figure 1. The powder X-ray diffraction pattern of the $CsTeMoO_6$ compound.

A comparison of the obtained X-ray diffraction pattern with the theoretical pattern, calculated from the single crystal X-ray diffraction data for this compound, indicated the monophasic nature of the powder. Impurity phases were not detected within the sensitivity of the method. The results of measuring the elemental composition showed a homogeneous distribution for the Cs, Te, Mo and O elements over the powder and correspond to the stoichiometric ratio of 11.50 at % (Lα, Cs), 10.44 at % (Lα, Te), 11.60 at % (Lα, Mo) and 66.46 at % (Kα, O) within the sensitivity of the method (~0.1 at %). The particle size distribution was calculated according to the Fraunhofer theory (Figure 2).

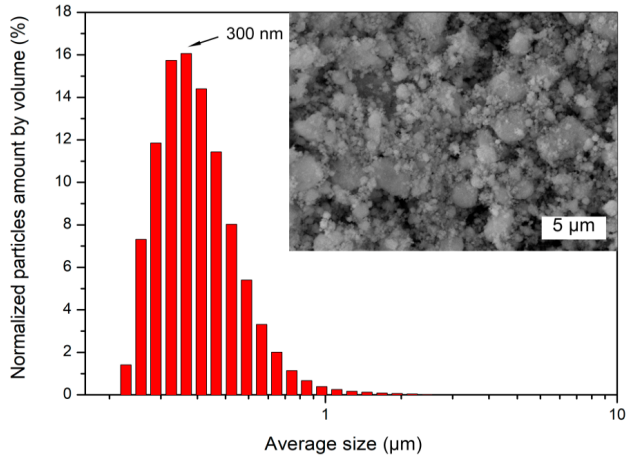


Figure 2. SEM photo and volume distribution of particles by size of $CsTeMoO_6$ powder.

The average particle size of the $CsTeMoO_6$ powder dispersed in a planetary mill was 670 nm, with the maximum particle size distribution at 300 nm (Figure 2). The particle size distribution was unimodal, and the SEM results showed that micron agglomerates of smaller particles predominantly represented the powder.

3.2. Antimicrobial Effect of Synthesized Material on Bacteria

Figure 3 show the results of studies of the antibacterial activity of a new complex metal oxide on the survival of *E. coli* and *S. aureus* bacteria under dark and light conditions.

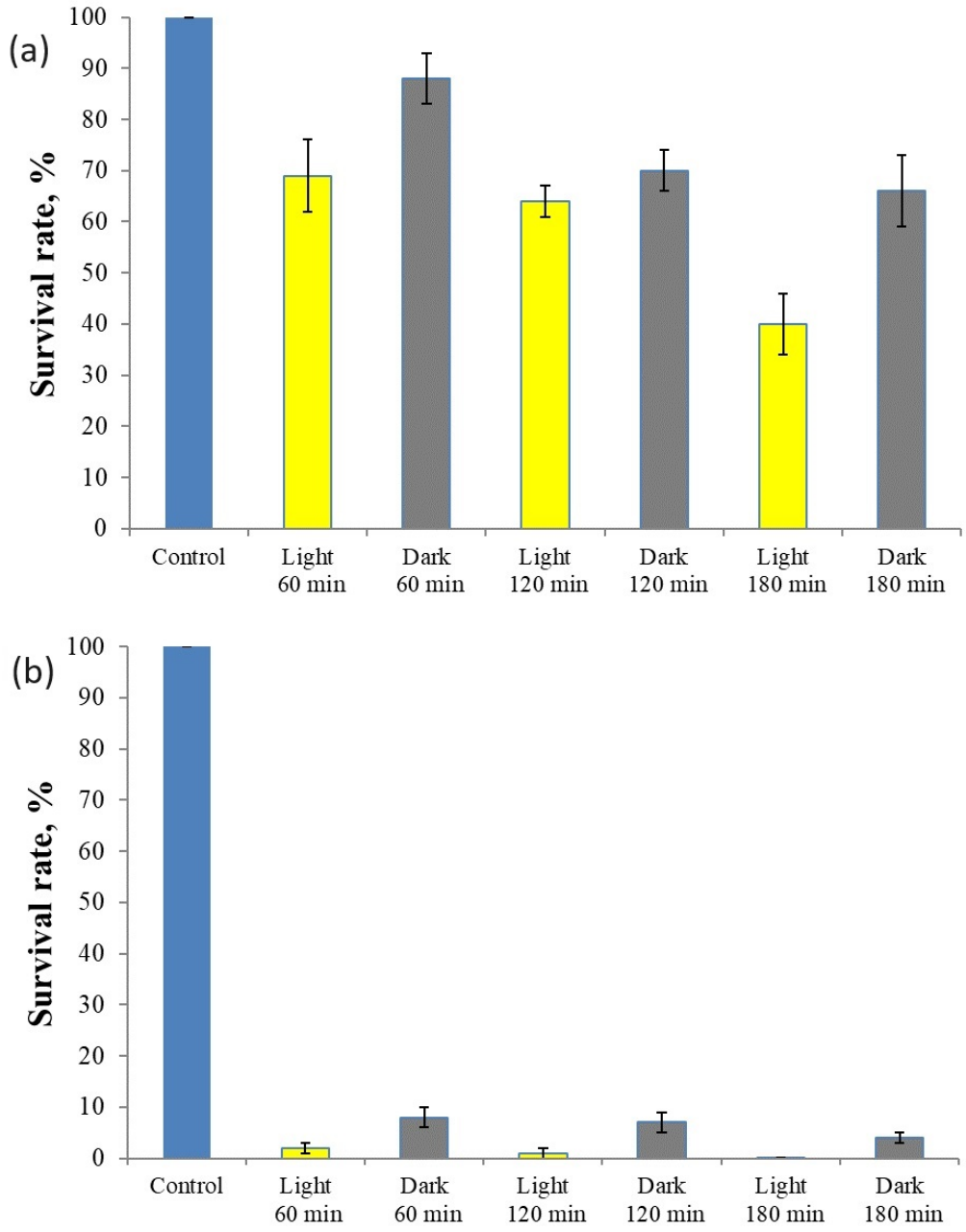


Figure 3. Survival rate of bacteria *S. aureus* (**a**) and *E. coli* (**b**) in the presence of $CsTeMoO_6$ under dark and visible light of a 50 W light source.

It was studied that $CsTeMoO_6$ inhibits the survival rate of all these microorganisms in both conditions (dark and light 50 W). A more pronounced antimicrobial effect was observed against *E. coli*. The action of light on the test substance $CsTeMoO_6$ led to a decrease in the survival of bacteria compared to the variants of the experiment with the introduced $CsTeMoO_6$, which were kept in the dark. It has been observed that the longer the exposure of the tested compound to *E. coli* and *S. aureus*, the higher the degree of inhibition of microbial survival, both in the absence of light and under light exposure conditions.

3.3. Antimicrobial Effect of Synthesized Material on a Fungi

The antifungal activity of $CsTeMoO_6$ against fungi *A. niger* and *P. chrysogenum* to both spores and vegetative mycelium under dark and light conditions was studied (Figures 4 and 5).

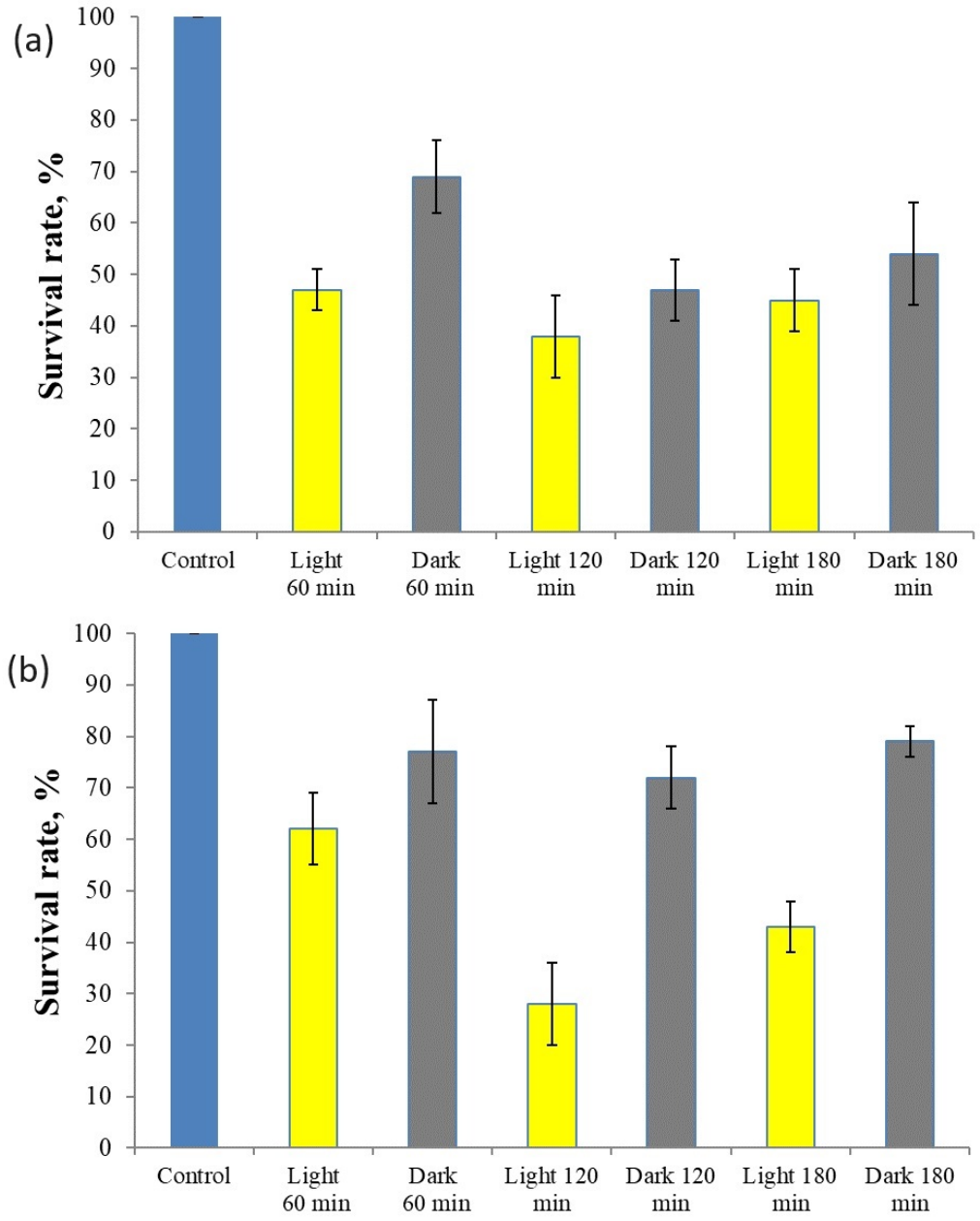


Figure 4. Survival rate of *A. niger* (**a**) and *P. chrysogenum* (**b**) spores in the presence of $CsTeMoO_6$ under dark and light 50 W.

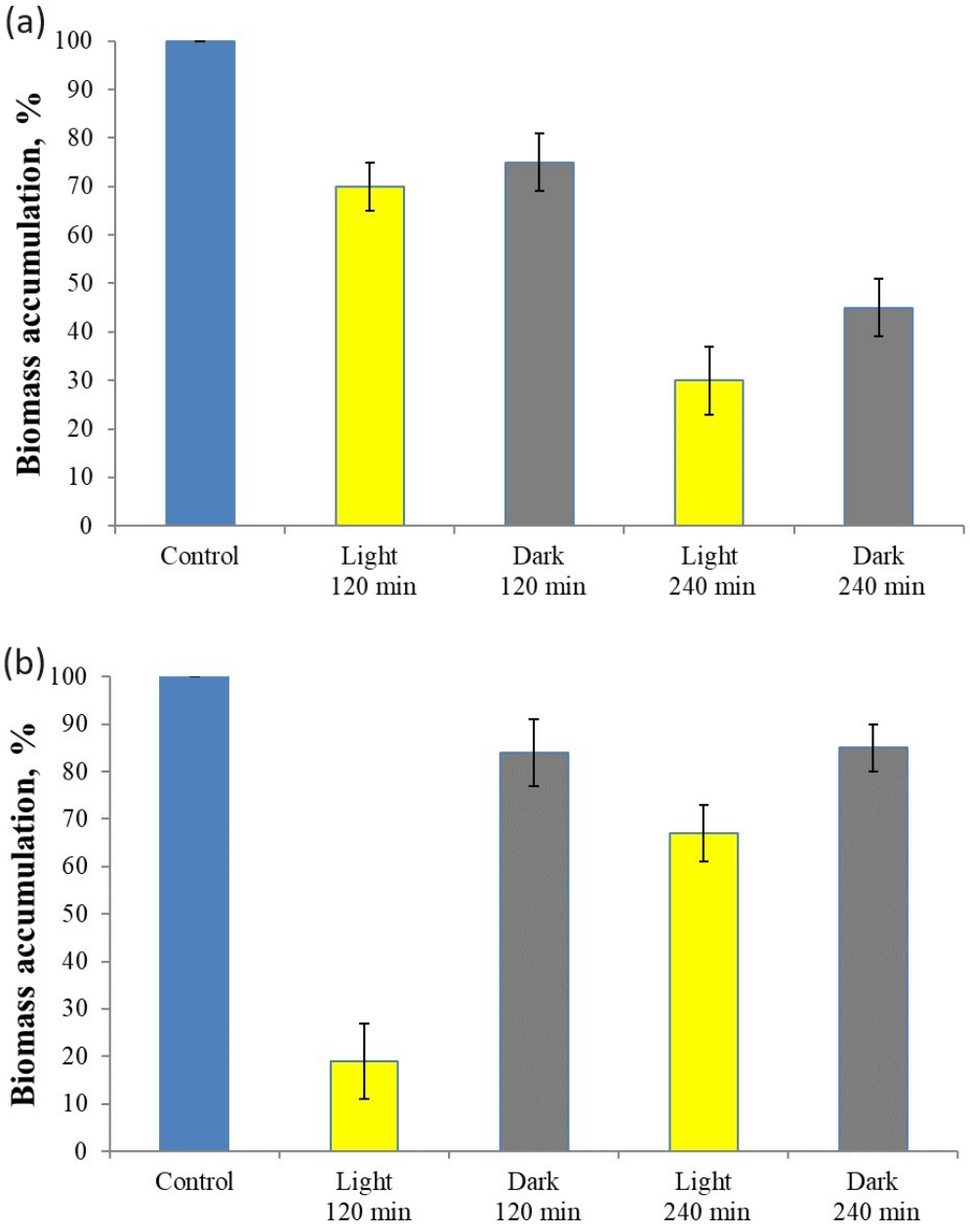


Figure 5. Reduction in biomass of *A. niger* (**a**) and *P. chrysogenum* (**b**) on a medium supplemented with $CsTeMoO_6$ in darkness and under the influence of 50 W light source.

In the case of the action of the compound on fungal spores, the antifungal activity was observed both in darkness and under light exposure and increased under light exposure, which, apparently, was associated with the presence of the photocatalytic activity of the $CsTeMoO_6$ (Figure 4).

A decrease in fungal biomass accumulation was observed under the action of the test compound in dark and light conditions in the case of experiments with vegetative mycelium (Figure 5).

The fungicidal effect of the compound increased under the action of light compared to the experiment in the dark like in the case of bacterial studies. It should be noted that in the case of *A. niger*, the inhibitory effect both in the dark and in the light increased with increasing exposure time. The inhibitory effect on the survival rate of vegetative mycelium decreased under the action of light with increasing exposure time in variants with *P. chrysogenum*. Thus, presumably it can be said that the effect of exposure duration on antimicrobial activity was ambiguous: when studying the effect of the compound on bacteria and vegetative mycelium of *A. niger*, there was an increase in the biocidal effect with increasing exposure time, and in the case of fungal spores and mycelial cells of *P. chrysogenum*, a decrease in or absence of statistically significant changes.

4. Discussion

This section presents data enabling us to postulate and elucidate the potential mechanisms of the antimicrobial activity of the compound under investigation, as well as the uncertainty in demonstrating the antimicrobial effect based on the metal's nature and on the anatomical–morphological and physio-biochemical characteristics of the microorganism group under investigation.

Previously [24], the antimicrobial properties of submicron particles of WO_3 and $RbTe_{1.5}W_{0.5}O_6$ were studied. It has been shown that the survival rate of bacteria and fungi decreased under dark and light conditions. Moreover, under the conditions of using light sources in the visible spectrum of different powers, there was an observed improvement in the antimicrobial ability of these compounds. It has been shown that reducing the particle size of $RbTe_{1.5}W_{0.5}O_6$ enhances its antimicrobial effect. The duration of exposure also influenced the degree of vital activity of bacteria and fungi.

Regarding the mechanisms of antimicrobial action on the metabolism of microorganisms, the literature provides the following information. Nano- and submicron particles of metal oxides can cause the death of microorganisms, and they exhibit an antimicrobial effect without light exposure [22,23,25–28].

Nano- and submicron particles of oxides can not only lead to the destruction of the cell wall and membrane of microorganisms due to electrostatic interactions, but they can also penetrate inside the cell and damage organelles (mitochondria and ribosomes) and, causing condensation and margination of chromatin, lead to the apoptotic death of the cell [29]. Also, nanoparticles can permeate through the cell membrane of microorganisms and bind to transport proteins to disrupt the operation of proton pumps [30], and they can inactivate phosphorus- and sulfur-containing enzymes and DNA. Nanoparticles play an important role in inhibiting ATP and reducing the number of copies of the bacterial 16S rDNA gene [31].

The toxicity of nano- and submicron metal oxides particles against fungi and bacteria increases under light. This means that there is the formation of reactive oxygen species (ROS) as a result of photocatalytic activity that can inhibit the vital activity of microorganisms [32,33].

According to [34], complex oxide $CsTeMoO_6$ has a wide band gap of ~3.1 eV, which corresponds to the UV range. However, combining the results of reflection and transmission spectroscopy allows us to conclude that the compound inside the band has two impurity-defect levels with energies of 2 and 2.6 eV [34,35]. Due to these levels, the $CsTeMoO_6$ compound can absorb visible light with the formation of electron–hole pairs on its surface, which lead to oxidation reactions and organic compounds, which we have shown using the example of methylene blue and methyl orange dyes [34] (Figure 6).

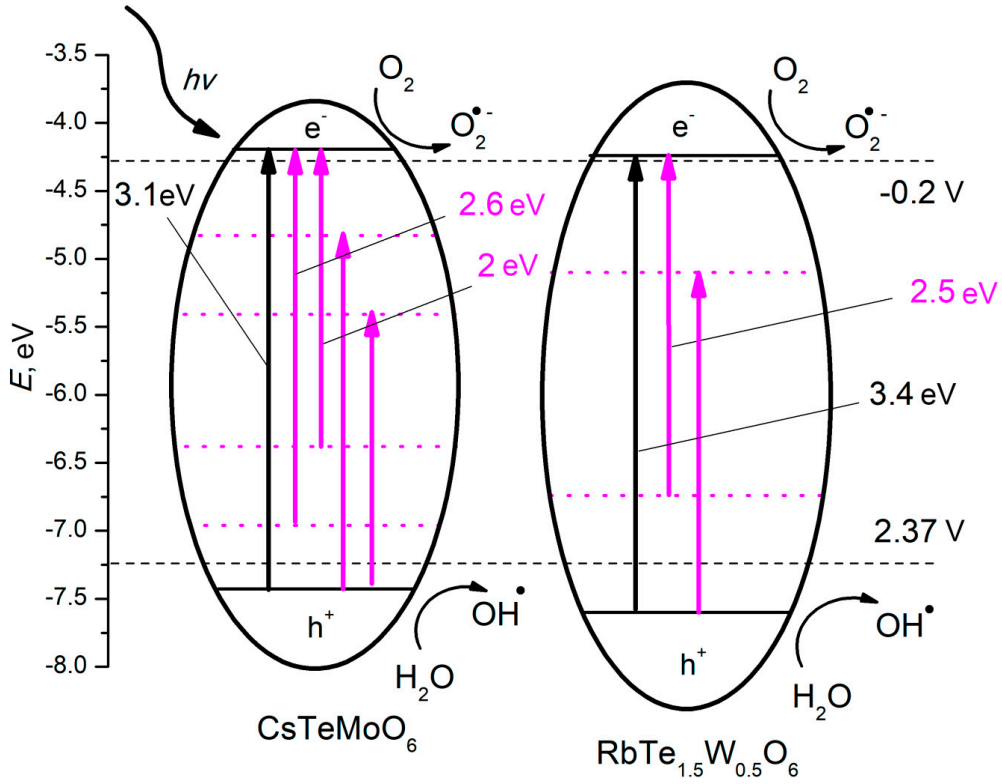


Figure 6. Scheme regarding the responsible conduction levels and the edge of the valence band of compounds $CsTeMoO_6$ and $RbTe_{1.5}W_{0.5}O_6$.

Figure 6 illustrates the mutual arrangement of the electronic levels of the $CsTeMoO_6$ and $RbTe_{1.5}W_{0.5}O_6$ compounds relative to the main redox potentials of water $\bullet O_2^-/O_2$ (−0.2 V) and $\bullet OH/H_2O$ (2.37 V) [36]. Both complex oxides can form simultaneously hydroxyl and superoxide radicals upon absorption of visible light. Possible chemical reactions on the surface of $CsTeMoO_6$ under visible light will be similar to $RbTe_{1.5}W_{0.5}O_6$ and they can be written as:

$$CsTeMoO_6 + h\nu \rightarrow CsTeMoO_6\ (h^+ + e^-) \quad (1)$$

$$H_2O + h^+ \rightarrow H^+ + \bullet OH \quad (2)$$

$$O_2 + e^- \rightarrow \bullet O_2^- \quad (3)$$

$$\bullet O_2^- + H^+ \rightarrow \bullet HO_2 \quad (4)$$

$$2HO_2\bullet \rightarrow H_2O_2 + O_2 \quad (5)$$

$$\text{biomolecules} + e^-/h^+/\bullet O_2^-/\bullet OH/H_2O_2 \rightarrow \text{oxidation.} \quad (6)$$

Thus, the oxidation of organic compounds can occur both directly through the interaction with the formed electrons and holes (1), and with the help of the formed hydroxyl and superoxide radicals ((2) and (3)). Moreover, secondary reactions between radicals are possible with the formation of hydrogen peroxide according to reaction ((4) and (5)), which also acts as a strong oxidizing agent.

According to [37,38], an active process of recombination of the resulting electron–hole pairs take place on the surface of the $RbTe_{1.5}W_{0.5}O_6$ photocatalyst, which reduces its oxidative capacity. However, studies on the mechanism of the photocatalytic process in the case of $CsTeMoO_6$ have shown an insignificant contribution of recombination; that is, the main part of the formed electrons and holes enters oxidation reactions [35].

Comparing the antimicrobial activity of complex oxides $CsTeMoO_6$ with $RbTe_{1.5}W_{0.5}O_6$ under the same conditions (50 W light source; surface flux density 524 W/m^2 and concentration of complex oxides 2 mg/mL) studied earlier, one can note the following: $RbTe_{1.5}W_{0.5}O_6$ had a much stronger inhibitory effect on bacteria *E. coli* and *S. aureus*, as well as on the vegetative mycelium of the fungus *P. chrysogenum* compared to cesium oxide both in the dark and in the light, despite the similar electronic structure and close particle size. This may be due to the different reactivity of electron–hole pairs and different efficiency of these reactions (2)–(5).

In [39], it was shown that the degree of oxidation of various organic bonds depends, among other things, on the nature of active radicals and particles. Thus, according to [35,38], in dye oxidation processes by complex metal oxides, the main contribution in the case of $RbTe_{1.5}W_{0.5}O_6$ to the decomposition reactions is made by hydroxyl radicals, while for $CsTeMoO_6$, these are superoxide radicals, and •OH are formed to a lesser extent. Apparently, hydroxyl radicals are most actively involved in the oxidation of bacteria and vegetative mycelium biomolecules compared to $\bullet O_2^-$, which interact more efficiently with simpler organic molecules. In this regard, $RbTe_{1.5}W_{0.5}O_6$ powder shows better antimicrobial activity than $CsTeMoO_6$, which is more suitable for the oxidation of organic dyes.

In addition, different types of fungi have different needs for water and nutrients, so adsorbed water and some impurities on the surface of complex oxides could both inhibit and stimulate their growth.

As we have already noted, the antimicrobial effect of nano- and submicron-sized metal oxide particles may depend on the nature of the metals. Thus, it can be hypothesized that different metals are capable of inhibiting certain metabolic targets in microorganisms under both light and dark conditions. On the other hand, the difference in the degree of survival of microorganisms under the action of $CsTeMoO_6$ under dark and light conditions may depend on the physiological and biochemical features in the studied cultures, namely, the presence of possibly different mechanisms of resistance to this complex metal oxide.

One of the mechanisms of the antimicrobial action of biocidal compounds is their influence on the cell wall and cytoplasmic membrane, resulting in the disruption of permeability processes through these structures. Therefore, the specific structure of the cytoplasmic membrane and cell wall of microorganisms plays an important role [40]. It is known that the structure and composition of the fungal cell wall differ from that of bacteria. The structure of the cytoplasmic membrane of bacteria also differs from that of fungi (the latter containing ergosterols in the membrane, which can affect its permeability) [41–44]. Furthermore, the chemical composition and structure of the cell walls of Gram-positive bacteria (*S. aureus*) are markedly distinct from those of Gram-negative bacteria (*E. coli*) [45]. It is known that the structure of peptidoglycans in E. coli and *S. aureus* differs in the nature of interpeptide bridges [46].

Metal oxides and their modifications destroy biological membranes, which leads to the death of living organisms. The membrane effect of these compounds, for example, in relation to bacteria such as *Escherichia coli*, *Staphylococcus aureus*, *Pseudomonas aeruginosa* and *Bacillus subtilis*, is mainly associated with a change in the charge of the bacterial cell wall [10].

There are also disparities in the structure of the cell wall between the vegetative mycelium and fungal spores, with the latter having a more robust, two-layered structure. Additionally, in species of the *Penicillium genus*, the spores' cell wall exhibits additional cutinization. It is recognized that the chemical composition of the mycelial cell wall differs between pp. Penicillium and pp. *Aspergillus*. The anatomical and morphological characteristics of the microorganisms under consideration, along with the diverse mechanisms of the inhibitory action of metal oxides on microbial metabolism, enable us to partially elucidate potential differences in the antimicrobial efficacy of the compound under investigation. This applies to its impact on various biological entities under both light and dark conditions.

Furthermore, it is known that microorganisms may exhibit unequal susceptibility to toxic compounds due to the presence of enzymes that facilitate the conversion of biocides into non-toxic forms. These enzyme systems may vary in their activity, impacting the microorganisms resistance to antimicrobial agent [47]. The elucidation of the specific mechanisms of the inhibitory effect of this compound lies outside the scope of this article and requires additional special research.

5. Conclusions

Thus, the studied complex oxide $CsTeMoO_6$ has an antimicrobial effect both in the dark and under light exposure to *E. coli* and *S. aureus*, as well as to spores and vegetative mycelium of *P. chrysogenum* and *A. niger*. The greatest inhibitory effect of submicron $CsTeMoO_6$ particles was noted against *E. coli*.

The antimicrobial effect of the test compound $CsTeMoO_6$ against bacteria and fungi increases under visible light source power 50 W and surface radiation flux density 524 W/m^2, which should be explained by its photocatalytic activity.

The duration of exposure had an ambiguous effect on the antimicrobial effect $CsTeMoO_6$. In some cases, the survival rate of microorganisms was decreased with an increased in the time of exposure in under light and dark conditions, but in some cases, it did not change or, on the contrary, it even increased.

Comparing the antimicrobial effect of $CsTeMoO_6$ and $RbTe_{1.5}W_{0.5}O_6$ compounds against of the same studied microorganisms, it can be noted that the latter one has stronger antimicrobial effect. The degree of suppression of the vital activity of individual cultures by these compounds is also different, which confirms the validity of the assertion that the antimicrobial effect of nano- and submicron particles of metal oxides with photocatalytic activity depends on the nature of the metals in these compounds.

It was shown that the oxidation of biomolecules by $CsTeMoO_6$ under the action of visible light can lead to the death of living organisms and can occur due to the formation of both electron–hole pairs and ROS.

The variability in the extent of inhibition observed in our experiments could also be associated with the presence of anatomical, morphological, physiological, and biochemical characteristics within the studied groups of microorganisms. These factors could potentially influence the penetration processes of the compound into cells through the cell wall and cytoplasmic membrane.

The obtained results expand theoretical concepts in the field of photobiology, namely, in terms of studying the antimicrobial effect and the mechanisms of its manifestation by nano- and submicron particles of complex metal oxides with photocatalytic activity. It is very important to compare the effect of submicron metal oxide particles of different chemical structure on various groups of microorganisms, as well as to reveal the dependence of antimicrobial activity on the size, shape of particles, the power of radiation sources, and the characteristics of their wave spectrum.

The photocatalytic activity of complex metal oxides makes it possible to selectively regulate their antimicrobial activity against certain types of microorganisms. It facilitates the use of such compounds as antimicrobial agents in various industries, in medicine and in veterinary medicine. Moreover, these chemicals can be used for wastewater treatment.

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Institutional Review Board Statement: Ethical review and approval were waived for this study, due to There were no human or animal experiments involved. The cultures of microorganisms from the study were used only for research in the laboratory. After the experiment, all cultures of microorganisms were sterilized to prevent their release into the environment. We have not used the cultures for commercial purposes or shared them with third parties or other institutions.

Informed Consent Statement: Informed consent was obtained from all subjects involved in the study.

Data Availability Statement: The data presented in this study are available upon request from the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest.

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