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A BIBLIOMETRIC ANALYSIS OF COPPER AND ANTIMICROBIAL COPPER COATINGS

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Abstract

Along with technological development, scientific literature has grown exponentially. Although the data resources are open access, filtering a large amount of information remains a significant problem. This paper aims to search and analyze the specialized literature on copper coatings and antimicrobial copper coatings. The aim was to extract the data from the Web of Science, which is the largest database for scientific literature. We used the VOSviewer software for analyzing the raw data that we collected from the Web of Science Core Collection. Within this domain, visualizing networks of key terms from published works holds significant importance. Thus, the objective of this research is to showcase advancements and research focuses within antimicrobial copper coatings, particularly those employed in high-traffic areas such as public transportation, airports, educational institutions, and notably in hospitals worldwide. This entailed two sets of keyword analyses: one focused on the overarching term describing the copper coatings field, namely "copper coatings," and the other aimed at fulfilling the primary study goal, using the keywords "antimicrobial copper" in the search. When searching for "copper coatings" OR "copper-coat" in all WOS-indexed databases at the time of the inquiry, 9,302 results were found, according to the Web of Science database. When searching for "antimicrobial copper" OR "antimicrob* copper*" in the Web of Science Core Collection, 71 results were found. Through processing these findings with VOSviewer software, distinct clusters of keywords were generated, with their significance filtered based on various criteria as outlined in each map's description. This study presents future avenues for research based on the findings.*

Keywords: *Copper coatings; bibliometric analysis; antimicrobial copper; database; web of science*

Introduction

Copper is a chemical element with atomic number 29, which has a density of 8.96g/cm^3, a melting point of 1085°C, and a pink-orange color. It is known that it is a semi-precious, non-ferrous metal, with good thermal and electrical conductivity, malleable and ductile. Since 1825 it has been used to cover the hulls of ships and it has been proven to extend their life by limiting the effects of algae and seawater corrosion. Currently, body coverings have a wide range of uses, especially in industry due to their unique properties that improve the durability and final performance of the products. Although copper is known for its electrical and thermal conductivity, its use has become more and more extensive, from corrosion protection to the use of its antimicrobial effect to the improvement of the aesthetic appearance of objects. Among the important properties of copper highlighted in Fig. 1, we find excellent electrical and thermal conductivity, good workability in both hot and cold, corrosion resistance, and important antimicrobial properties. Copper coatings also have good adhesion to the substrate - ensuring

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effective protection. Its aesthetic appearance allows it to be used in interior and exterior design. Depending on the application process and composition of the coating, it can provide resistance to wear and abrasion.

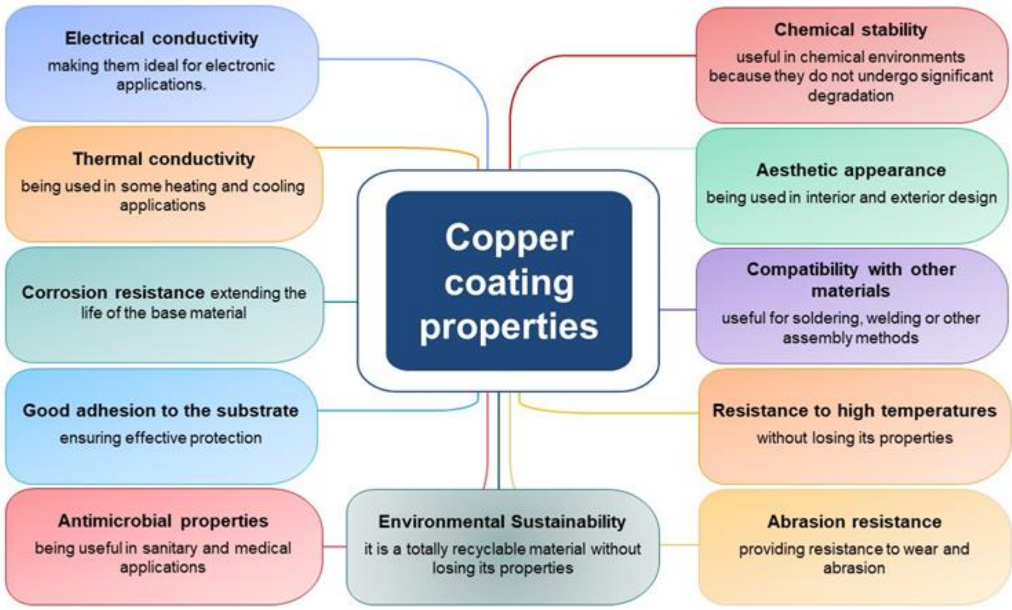

Fig. 1. Copper coating properties

An essential property of copper coatings is their ability to adhere firmly to the surface of the base material, providing effective protection. When it is used in certain applications, copper coatings must withstand high temperatures without losing their properties, having a good resistance to high temperatures. Also, some copper coatings are designed to be compatible with other materials and processes, such as soldering, welding, or other assembly methods, and, due to chemical stability, in certain environments, copper coatings are stable and do not undergo significant degradation.

Extensive research has been conducted on the resilience and anti-corrosive properties of copper and its alloys post-surface coating, particularly about their use in coatings across various industries [1].

Ensuring the firm adhesion of coatings to copper surfaces is pivotal in guaranteeing their durability. Copper coatings have shown to be an effective way to preserve and improve materials, whether they are used in electronics, architecture, or even healthcare (Fig. 2).

Copper coatings are the most used in the electronics industry, being essential both for the manufacture of printed circuit boards and for many other electrical components (for which both the conductive properties of this material - for the transmission of the electrical signal - and its anti-corrosive properties are essential). Cu coatings on semiconductor substrates are essential in the production of advanced electronic devices. It is also used in the transport of electrical energy, through both single-wire and multi-wire cables of various sections and protective materials (sheaths), benefiting both from its excellent conductivity and flexibility properties.

As seen in Fig. 3, there are several techniques for deposition of copper coatings. Using Thermal Spray Techniques, thick copper coatings with good corrosion resistance are created, with an approximate thickness ranging from 20 μm to several millimeters. good adherence, little wear, and affordable [2,3]. After the copper has melted, the molten particles are given kinetic energy via a jet. The molten particles will rapidly solidify to produce a covering at the substrate upon impact.

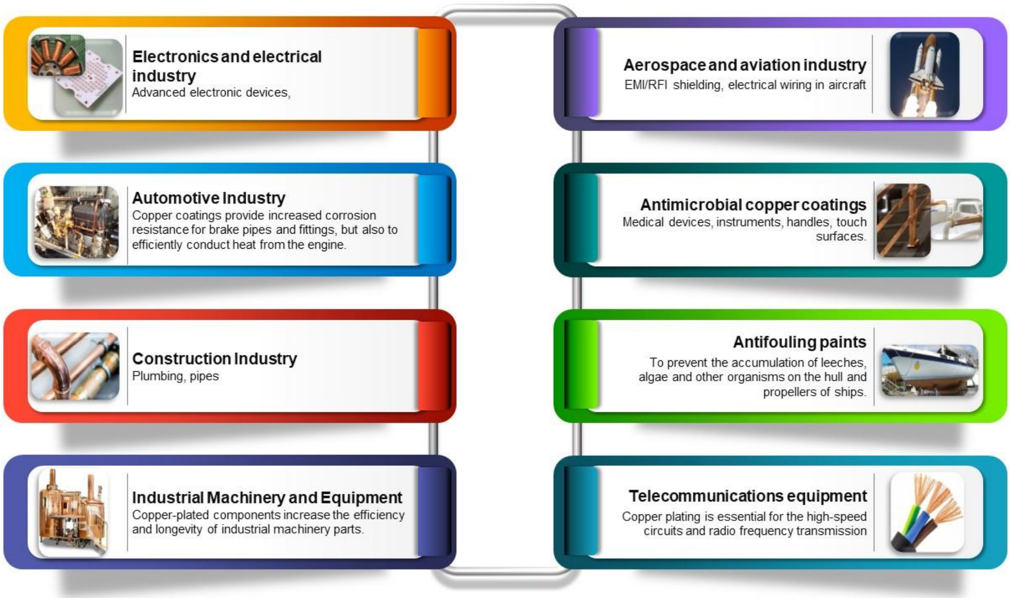

Fig. 2. Industrial applications for copper coatings

The most often used thermal spraying methods include cold, kinetic, flame, wire arc, and plasma spraying. Another technique for applying copper coatings is electrodeposition, which uses the electric energy that flows via a closed electrochemical circuit to deposit metal ions on the cathodic metal substrate. The substrate to be coated is connected as the cathode, and a copper plate serves as the sacrificial anode to maintain a steady density of metal ions in the electrolytic solution.

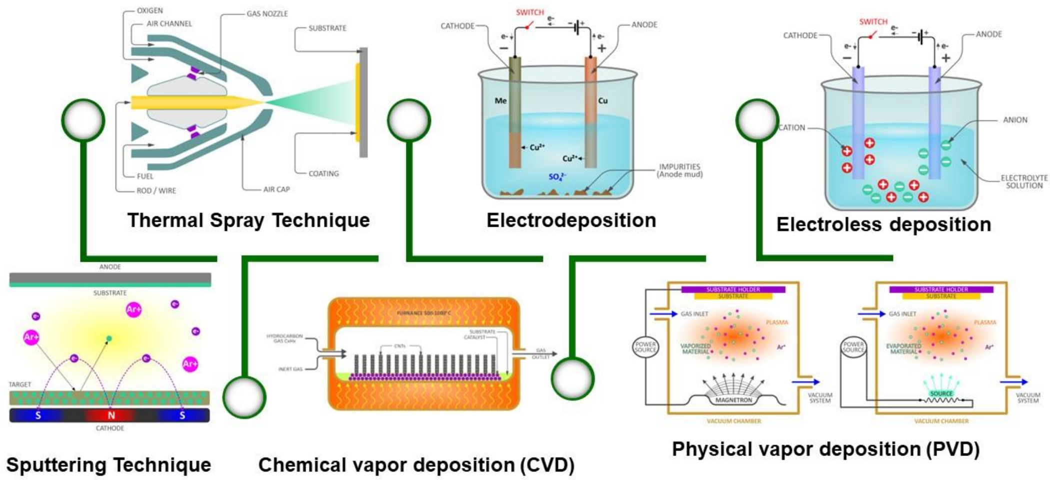

Fig. 3. Copper coatings techniques

Electroless Plating, also known as electroless copper deposition, refers to the chemical process used to deposit a thin layer of copper on a substrate without the use of external electricity based on an auto-catalytic chemical reaction. To activate its surface, the substrate is prepared and immersed in a copper plating bath containing copper ions and a reducing agent [4–6]. Simple cupric salts, such as copper sulfate, chloride, or nitrate, are the source of copper, while formaldehyde or glyoxylic acid are typically employed as reducing agents. The reducing agent reacts at the catalytic sites on the substrate, donating electrons to reduce copper ions to copper metal. The deposited copper acts as a catalyst, continuing the reaction. This self-catalytic process allows the uniform layer of copper to grow on the surface. The plating bath contains complexing agents that regulate the metal deposition rate and other additives that refine the deposit properties. Depending on the temperature, pH, composition of the bath, stirring speed, and the deposited film layer are determined [7]. With the physical vapor deposition (PVD) technique, the coating layer forms on the substrate atom by atom during a thin film deposition process that can produce layers as thin as a few microns. The enhancement of wear and corrosion resistance is the primary purpose of this technique. One can carry out atomic deposition in an electrolytic, gaseous, vacuum, or plasma environment. There are numerous techniques, including as electron beam sputtering, magnetron sputtering, ion plating, and sputtering [8–10]. The two most used PVD techniques for thin film deposition are evaporation and sputtering, often known as cathodic spraying [8]. Chemical vapor deposition (CVD) is the process of using reactive gas to deposit a thin film through a chemical reaction that occurs on or near a heated substrate surface. In an active medium (heat, light, or plasma), gaseous reactants dissociate and/or undergo chemical reactions, and a stable solid product is subsequently formed. Using the sputtering process, ions produced in a light discharge plasma in front of the copper target (cathode) are directed towards it with great force. The cathode is then cleared of atoms, ions, and free electrons. The process of bombardment results in the "spraying" of the target atoms, which are then driven by momentum to deposit as a thin film on the substrate (anode) [11].

The wear and tear experienced by copper coatings, especially in high-traffic zones, has been a focal point of this investigation. Efforts have been directed towards developing coatings with superior abrasion resistance, aiming to preserve their efficacy over extended periods. Copper coatings are required to withstand exposure to chemicals, cleaning agents, and environmental elements without degradation.

The presence of various microbes, bacteria, and microorganisms around people has become a thorny problem due to the infections and diseases they generate or transmit. This problem can be found not only in the sanitary system but also in public spaces, the food industry, the hotel industry, and many other areas as is presented in Fig. 4. Generally, it is a problem in high-traffic areas. Infections associated with the health system (which includes hospitals, polyclinics, clinics, nursing homes, dental offices, and school medical offices), have a considerable impact on people's health throughout the world and implicitly on the associated costs [12]. Healthcare-associated infections (HAIs) have a major influence on public health because they increase the mortality rate and the costs due to the prolongation of patients' hospitalization. Areas with intense traffic like airports, train stations, subways, and bus stations are the most vulnerable because they are contaminated with pathogens and require repeated disinfections, which involve additional costs both in terms of people and related substances [13]. But frequent contact with harsh materials and detergents leaves scrapes and indentations that might host bacteria and viruses [14]. That is why it is useful to ensure surfaces that are not recontaminated for a certain period. Stainless steel is widely used in the manufacture of furniture, hospital equipment, safety bars, surfaces that are frequently touched in elevators, airports, train and bus stations, schools, and universities, food processing equipment, and utensils used in restaurant and hotel kitchens due to their resistance, inertia, appearance, and relatively low production costs. Antimicrobial materials or antimicrobial coatings can help reduce the risk of HAIs. Copper coatings on stainless steel are used in all levels of the medical system from medical implants and surgical devices to IV poles, hospital beds, and push plates due to their antibacterial properties.

Usually, materials with medical applications are used in a wide range of medical applications [15–20]. Numerous strategies and principles have been developed to discover and produce coatings with antibacterial properties [21–23]. There are two main ideas behind commonly used methods to lessen the vulnerability of antimicrobial surfaces to microbial colonization and illness [21]: surface treatment with antibacterial substances and materials with antibacterial properties [24,25]. Frei A. et al. stated that antimicrobial metallic materials should have two main properties:

they should produce ions that are poisonous to viruses and bacteria, and they should have a redox-active surface under ambient conditions [26].

Fig. 4. Use of antimicrobial copper coatings

As the bibliometric analysis shows, only in the last 25 years have researchers started to analyse its antibacterial properties. It is now known that about 80% of infectious diseases are transferred through touch contact [27]. The Centres for Disease Control and Prevention (CDC) records estimate that infections in US hospitals affect 2 million people annually, resulting in approximately 100,000 deaths annually [28].

Some microorganisms, bacteria, and viruses can survive on surfaces for as long as one to five months. These include *Mycobacterium tuberculosis, Clostridium difficile, Enterococcus spp., Staphylococcus aureus* (including MRSA), *Streptococcus pyogenes, Acinetobacter spp., Escherichia coli, Klebsiella spp., Pseudomonas aeruginosa, Serratia marcescens* or *Shigella spp., Candida albicans, Torulopsis glabrata*, astrovirus, HAV, poliovirus, or rota virus. Other microorganisms, bacteria, and viruses that can survive for a few hours to two weeks include *Candida parapsilosis, Bordetella pertussis, Haemophilus influenzae, Proteus vulgaris* or *Vibrio cholerae*, corona, coxsackie, flu, SARS or rhinoceros' virus, and herpes viruses like CMV or HSV types 1 and 2 [14,30,31].

According to a 2016 study by Schmidt et al., adding copper surfaces to items that had previously been covered in wood, plastic, stainless steel, or other materials lowered the microbial load by 83% and kept it there for 21 months. [29]. Similarly, after 30 hours, the microbial load of some tables with copper-impregnated trays was 81% lower than that of normal materials, according to a 2017 study by Coppin et al. [30]. It was demonstrated in a different study by Schmidt et al. that stethoscopes impregnated with copper showed up to 91% lower CFU/cm^2 than stethoscopes without copper [31].

As it can be seen in Fig. 5, copper's antimicrobial properties stem from its ability to interact with microbial cells and disrupt their cellular processes [32]. Can also inhibit the reproduction of bacteria and can lead to their rapid destruction by affecting the integrity of the cell membrane, by disrupting metabolic processes or by generating reactive oxygen species (ROS) in bacterial cells, when it is applied to various surfaces susceptible to contamination[33]. One of the mechanisms by which copper coatings irreversibly affect bacteria is related to the fact that it interferes with the metabolic processes of bacteria, such as cellular respiration and DNA synthesis, preventing them from multiplying and developing. Reactive oxygen species in bacterial cells are unstable molecules that can generate damage by oxidizing cellular components (proteins, lipids, DNA), oxidation that affects the functioning of the cell and can cause the death of the bacteria [34,35]. Furthermore, too much copper causes bacteria's membrane integrity to deteriorate[36,37].

Because of this damaged integrity, vital nutrients for the cells—like potassium and glutamate—leak out, causing the cells to dry out and eventually die. Copper can also inhibit the ability of microorganisms to reproduce, which contributes to its effectiveness in preventing microbial contamination[38]. It can effectively kill or inhibit the growth of bacteria, viruses, fungi, and even some protozoa. Therefore, the surfaces covered with copper, through its antimicrobial properties, can lead to the rapid destruction of bacteria and the creation of an environment.

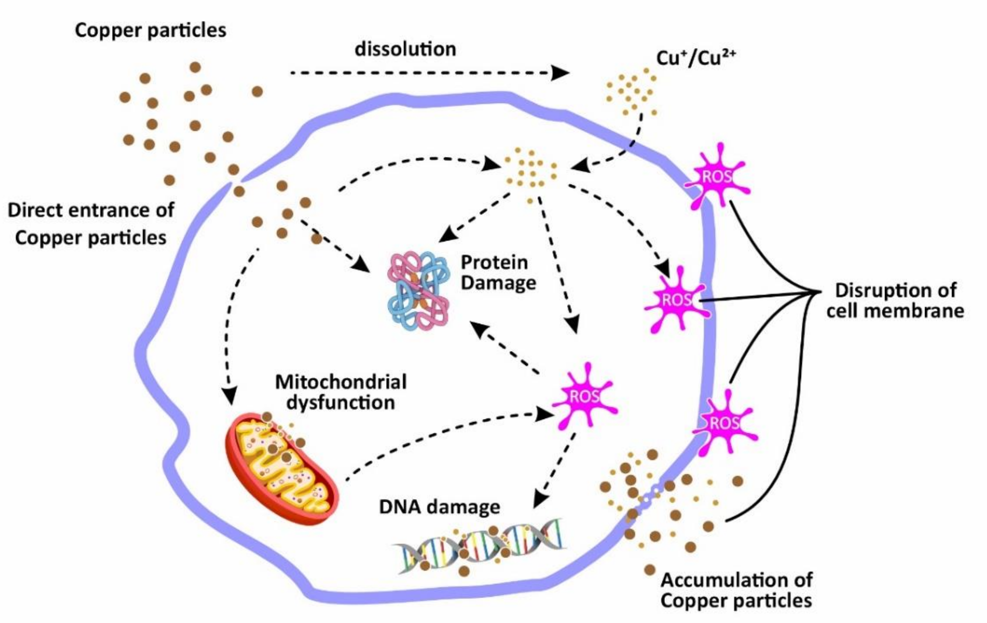

Fig. 5. Antibacterial mechanism of copper coatings.
Adapted from [33]. In the initial reference Fig. is licensed under CC BY

Studies have delved into the chemical stability of coatings across diverse environmental conditions. Konieczny J. And Co. mentioned that there are several mechanisms the increased presence of copper in cells induces oxidative stress, which results in the generation of hydrogen peroxide, copper actively participating in what is known as the Fenton reaction, a chemical process that causes oxidative damage to cells [1,39–41].

Moreover, copper in higher amounts leads to decreased membrane integrity of microorganisms, which results in the leakage of essential cellular nutrients such as potassium and glutamate, leading to cellular dehydration and subsequent cell death [42–44]. Although copper is essential for many protein functions, too much copper can bind to proteins that don't need it to function correctly, which causes protein functions to atrophy [45,46].

Copper coatings may have ecotoxicological effects since the element is used in antifouling paints, which stop organisms from colonizing the surface of marine boats. Because scientists fear that places with a high density of sailing vessels may see heightened quantities of copper, the toxicity of copper to aquatic creatures has been the subject of recent research. There are European countries where the use of copper on pleasure vessels in inland fresh waters has been banned. In aquatic toxicology there is a concern related to the influence of copper on marine fauna because it can be toxic at concentrations relevant to the environment and because it is an essential micronutrient [47].

Usually, we find copper in nature, but also as part of a significant number of enzymes, which makes it essential for any organism. At the same time, in too large quantities it becomes toxic. As can be seen in the presented study, the importance of using copper and copper coatings is increasing. High concentrations of copper can also be found in mining areas, especially when it appears together with nickel or zinc [48,49].

Methodology of Research

The current study uses the bibliometric evaluation technique to analyze published literature in the field of copper coatings with antimicrobial properties used for high-touch surfaces. This technique allows for the creation of visual presentations through quantitative analysis of the published literature.

The most popular data source for analyzing scientific papers across all study domains is the Science Citation Index, which was derived from the Web of Science Core Collection of Clarivate Analytics. Currently in use are the programs Gephi [50], Leximancer [51], and the VOSviewer [52,53]. Bibliometric analysis began to be used more and more in many fields [50,54–56] through the distribution models of the articles but also due to the ease of handling the volume of research published on a certain topic.

Using this technique, one may confirm the trend in each research field and draw attention to the distinct links between the keywords unique to each field of study. Since the Web of Science (WoS) database is the most selective in terms of publisher ranking and indexing, the analysis was conducted on studies that were indexed in it. Nees Jan VanEck and Ludo Waltman of Leiden University (Netherlands) developed the VOSviewer software (version 1.6.19), which was used to process the data transmitted from the WoS databases on April 4, 2024.

If in our search by the title and the topic of the articles we add copper coatings without quotation marks (" "), then the search engine will also add the articles in which they are not included, and a significantly larger number of articles will result (147,940). The reason why we added the keywords "copper coat*",”Cu coat*”, and ”copper surface*”, is to select in our searches including the variants of the word coating, coatings, or any of the variants. We also requested that all results be selected only from articles and reviews written between 1975 and 2024 (the maximum range). The number of articles published between 1975 and 2024 with the keywords "copper coatings" OR "copper coating*" OR ”Cu coating*” in the tile is 9,302 of which 8,015 articles, 1,552 proceeding papers, 49 early access, 103 review articles, 74 meeting abstracts, 45 letters, 43 notes, 16 editorial materials, 12 book chapters, 7 News items, 4 Corrections, 2 data papers, 2 retracted publications, 1 corrections addition, 1 discussion, and 1 retraction.

We also searched by the keywords of the articles "copper coatings" OR "copper coating*" OR ”Cu coating*”, in the same period and we added 71 articles and reviews to our database, of which 63 articles, 4 proceeding papers, 8 review articles and 1 book chapter.

We applied the same algorithm for ”antim* copper*” OR ”antimicrob* Cu” OR "anti* copper surf*" OR "antibact* copper coat*", OR "antib* copper coat*" respectively we searched for these keywords only in articles and reviews written in the same period. If we had only searched for the keywords antimicrobial copper (without the quotes), several 4472 results would have appeared. After the correct selection of the algorithm, only 162 articles and reviews were written of which 137 articles, 13 proceeding papers, 2 early access, 9 review articles, 4 meeting abstracts, 2 notes 1 editorial material, 1 letter, and 1 book chapter, shown in the graphs below according to the journal and the years in which they were published all written between 1975-2024. The data were retrieved from the databases in a tab-delimited format (TAB), considering two sets of data: one analysis was performed for the most general term that describes the copper coatings field, i.e., "copper coatings"; the second analysis was performed to achieve the main objective of the study, and the terms "antimicrobial copper" were used in the search. According to the WoS database, at the interrogation moment, the search for the term "copper coatings" or "copper-coat*" shows 9,302 results from all databases indexed in WOS, while the search for the term “antimicrobial copper” shows 162 results. Different keywords were created by processing these findings with the VOSviewer software, and their applicability was screened based on various factors described in each map's description. The search by the title of the article gives just one result. A total of 652 authors contributed to the topics during this period.

As can be seen from the previously presented bibliometric data, copper coatings are intensively used since 1975 (since information is available in the researched database) respectively 9302 articles, while their applicability in the medical area, where antimicrobial properties are needed, is less researched, 162 articles were found.

Results and Discussion

Copper Coatings

Fig. 6 shows the co-occurrence of keywords for the search "copper coatings" shown. The fractional counting approach is used to identify about 22684 keywords. Only 413 of the results fulfill the criterion when the results are filtered based on a minimum of 25 occurrences of a term. The highest scoring term was copper, which was followed by adsorption and behavior. Copper, adsorption, behavior, films, microstructure, surface, corrosion, Cu, performance, fabrication, oxidation, water, deposition, electrodeposition, copper surfaces, nanoparticles, growth, surfaces, mild steel, mechanism, coatings, mechanical properties, wettability, aluminum, spectroscopy, copper surface, temperature, benzotriazole, inhibition, adhesion, alloy, graphene, derivatives, thin-films, steel are the terms that have been identified, listed in descending order of occurrence.

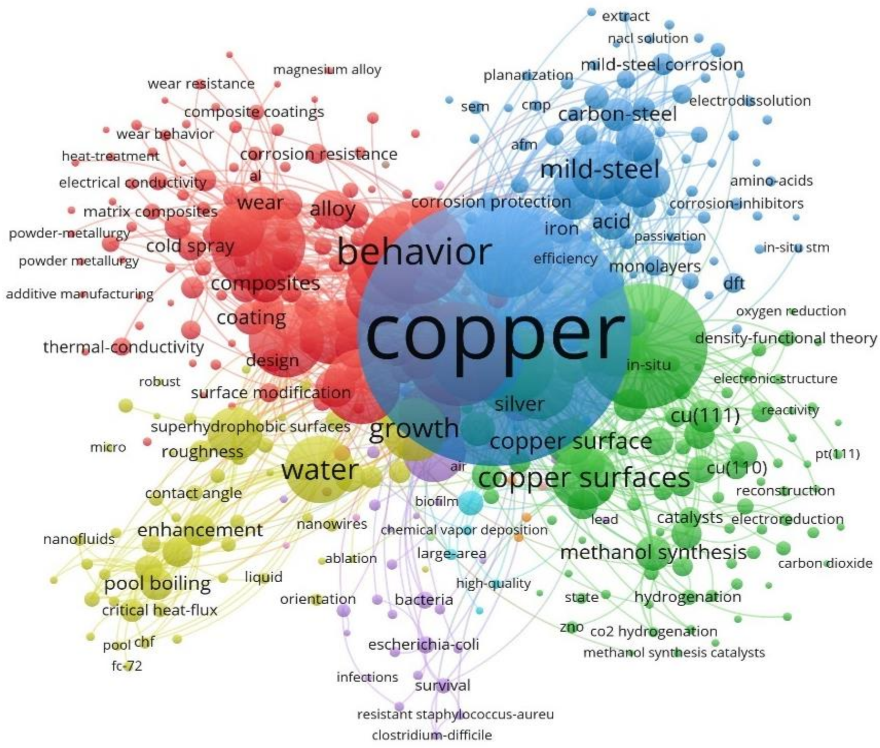

Fig. 6. Co-occurrence of keywords for the term "copper coatings" in the Web of Science database

By the co-authorship analysis visualization using the fractional counting method, the software recognizes 31,377 writers, 7 of whom match the requirement of having at least 20 documents. Additionally, it appears that the map displays 4 clusters connected to the authors' international distribution and the absence of publications from these research groups. Eight of the clusters only have one author, with the largest cluster meeting the requirements having four writers.

In the next Fig., the timeline in which the authors published articles on the given topic is presented in color. Light colors mean the period closer to the present time and dark colors mean the period closer to the beginning of the analyzed interval.

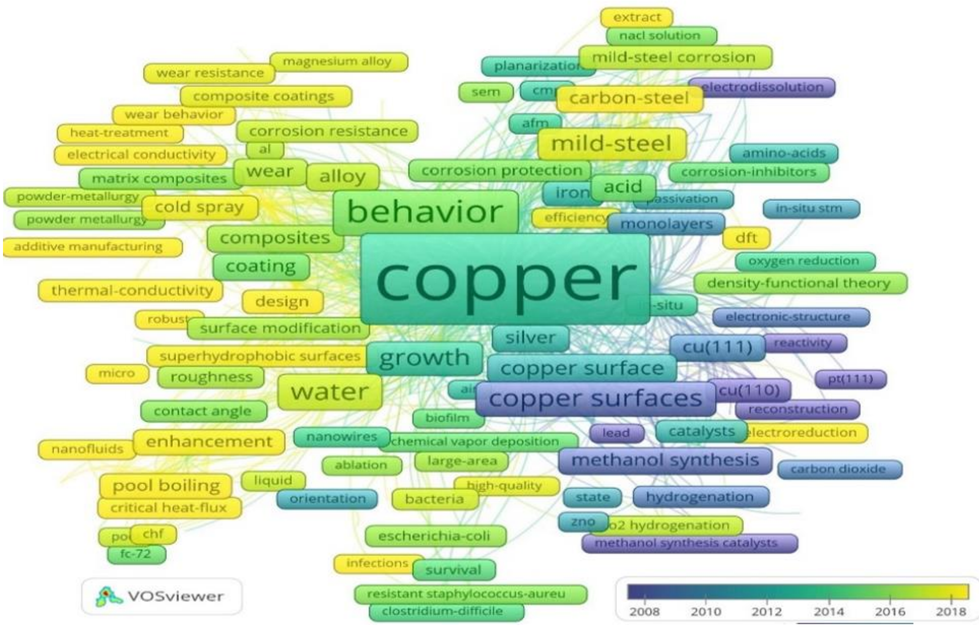

Fig. 7. Co-occurrence of keywords for the term "copper coatings" in the Web of Science database based on the articles' publication dates

53 of the 114 distinct nations identified by the examination of the geographical distribution of authors with publications in this discipline had at least 25 articles (Fig. 8).

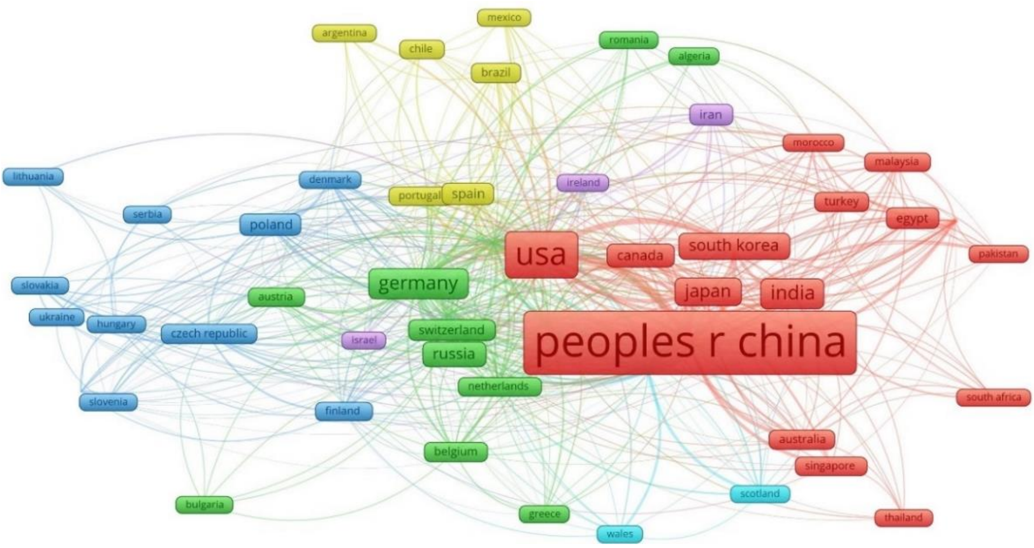

Fig. 8. Visualization of countries affiliated with a minimum of 25 publications in the period 1975 – 2024

The United States of America has the most citations (62,020 citations for 1,481 documents), followed by Germany (15,952 citations for 563 documents) and the People's Republic of China (53,952 citations for 2,494 documents). With 9365 citations for 352 documents, France holds the fourth position, and the United Kingdom is rated fifth with 12694 citations for 342 documents.

Antimicrobial Copper Coatings

The visualization of keyword occurrence of the search ”antim* copper*” OR ”antimicrob* Cu” OR "anti* copper surf*" OR "antibact* copper coat*", OR "antib* copper coat*" is presented in Fig. 9.

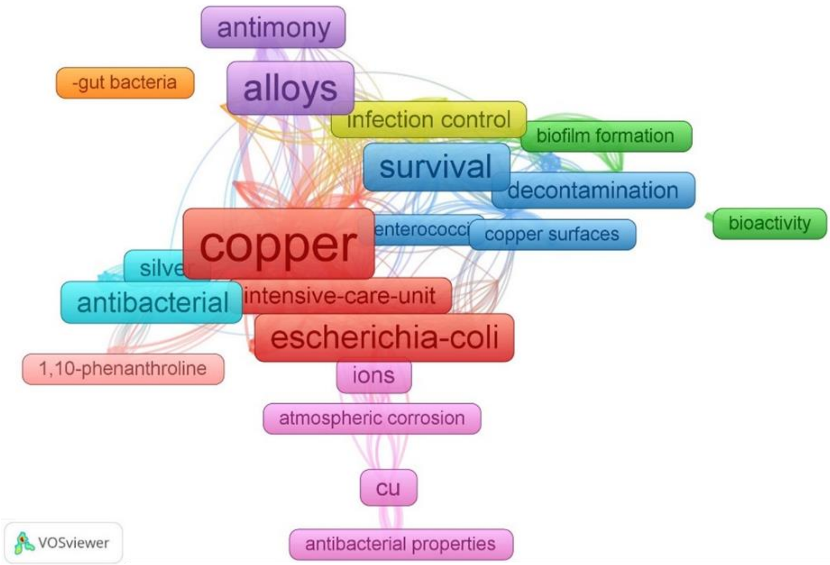

Fig. 9. Visualization of keywords co-occurrence for the terms “antimicrobial copper” in the WoS database

Fig. 9 shows the co-occurrence of terms for the search term "antimicrobial copper" shown. 302 keywords are found by using the fractional counting approach. Only five of the results fulfill the criterion after the results are filtered based on a minimum of five keyword occurrences. Copper had the greatest score, followed by toxicity and bacteria. According to the frequency of recurrence, the terms that have been detected are as follows: copper, toxicity, bacteria, *Escherichia coli*, as well as inactivation, survival, alloys, and antimicrobial copper. Furthermore, the size of each point in the network visualization is determined by the frequency of each keyword in the data that was studied.

Evidently, the terms that are most highlighted are The following is a list of terms that have been recognized, arranged in descending order of occurrence: The terms "*copper*", "*toxicity*", "*bacteria*", "*Escherichia-coli*", "*inactivation*", "*survival*", "*alloys*", and "*antimicrobial copper*" are listed in order of decreasing order of correlation with the characteristics of high-touch surfaces and the interest of researchers in this field. Thus, it can be stated that the antimicrobial copper coatings will influence the effect over some of the most important factors that determine healthcare-associated infections by destroying them. Furthermore, there appears to be a lot of novelty in the articles on this subject, and the majority of the study has been done on copper coatings, deposition techniques, coating characteristics, and adherence. Also, the effect produced by antimicrobial copper coatings was mostly evaluated through toxicity, survival, and inactivation parameters (as seen in Fig. 9). The frequency of these phrases in the titles or abstracts of the reviewed papers is shown by the size of the frame from the keyword occurrence visualization. The binary counting method (which takes into account a term's presence or absence rather than its occurrence inside a document) was used to construct the map based on text data.

Additionally, the software created three clusters out of the gathered data. There are eleven items in the first cluster (shown by the red links) among which we find copper, Escherichia coli, intensive care unit, bacteria, antibacterial, and survival. The five components in the second cluster (the green connections) are biofilm formation, bioactivity, alloys, decontamination and bacteria. One of the clusters is related to the problems that occur due to the infections and the main solution – antimicrobial copper, the second includes terms specific to the effect of the copper used for the coatings, and the third one is mostly related to comparative studies made by the researchers.

Limitations

The bibliometric analysis has several limitations, despite the fact that this study adds to the literature by highlighting the developments in the field of copper coatings, particularly in the case of antimicrobial copper coatings. These restrictions have to do with the authors' use of the WoS indexing and inhomogeneity when characterizing the copper coatings. The database's limitations are connected to the discrepancy between the published literature's publication date and indexing date. Furthermore, a large number of journals are not included in the WoS database's index. The words "coatings for copper products," "copper surface," "antipathogenic copper coatings," and so on that researchers have used for the term "copper coatings" are among the restrictions linked to the inhomogeneity of the literature. Given these limitations, the study's description of the present status of the literature on antimicrobial copper coatings may not be entirely accurate. On the other hand, the quality of the database and the large quantity of examined literature provide it relevance.

Conclusions

The VOSviewer software, which enables a quantitative analysis of the published literature, is used in the current study's bibliometric analysis of copper coatings and antimicrobial copper coatings. Using this method, the studies listed in the Web of Science database were assessed in order to identify the most important researchers and current developments in the field of copper coatings.

The entire database for the two keywords—"copper coatings" and "antimicrobial copper"—was searched using 31,511 authors and 22,755 keywords related to the body of current literature. Furthermore, 114 different countries' institutions are connected to these co-authors. With the exponential evolution of technology, various industries are constantly looking for innovative solutions to improve the performance of their products. As this bibliometric study proves, copper coatings have attracted the attention of researchers and implicitly of manufacturers in recent years. Copper coatings (classic or tinted antimicrobial) offer a multitude of benefits, among which we only mention extraordinarily good conductivity, outstanding corrosion resistance, and of course important antimicrobial properties. Among the areas that will gain great interest, we mention nanostructured copper coatings (created by depositing Cu nanoparticles on the surface that offer better adhesion than classic coatings and implicitly a much-improved durability and corrosion resistance through the control of the layer thickness).

Another direction of research is the hybrid coatings of copper with other materials (for example graphene), the resulting products showing improved conductivity, increased resistance, and reduced friction. In the aerospace industry, they have proved interesting, especially by reducing the weight of their various components without changing the properties of the individual components. Another interesting direction is the development of ecological copper coatings, researchers are looking for new coating methods without involving dangerous or energy-consuming chemical substances (such as electroless deposition which is an interesting technique that allows the deposition of copper coatings without the use of chemicals harmful and does not require external energy sources).

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