

**CHEMISTRY AND TECHNOLOGY OF MEDICINAL COMPOUNDS  
AND BIOLOGICALLY ACTIVE SUBSTANCES**

**ХИМИЯ И ТЕХНОЛОГИЯ ЛЕКАРСТВЕННЫХ ПРЕПАРАТОВ  
И БИОЛОГИЧЕСКИ АКТИВНЫХ СОЕДИНЕНИЙ**

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RESEARCH ARTICLE

## Antibacterial activity of green fabricated silver-doped titanates

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### Abstract

**Objectives.** The study aimed to synthesize the multifunctional materials silver-added titanates via reduction of sol-gel fabricating titanates ( $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$ ) with *Jasminium subtriplinerve* Blume leaf extract.

**Methods.** The physicochemical characteristics of the obtained materials were determined by X-ray diffraction, energy dispersive X-Ray spectroscopy, Raman spectroscopy, Brunauer–Emmett–Teller specific surface area, scanning electron microscopy, and UV–Vis absorption spectroscopy.

**Results.** The results demonstrated good dispersion of silver on the surface of  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  to create photocatalysts with two light-absorbing regions. The obtained materials were applied as antibacterial agents in polluted water. The Ag– $\text{Fe}_2\text{TiO}_5$  (Ag–FTO) samples showed better properties and antibacterial activity than Ag– $\text{NiTiO}_3$  (Ag–NTO) due to the better dispersion of silver nanoparticles on the FTO surface. Besides, the antibacterial results exhibit increased inhibiting activity against gram-negative (–) bacteria as compared with gram-positive (+) bacteria.

**Conclusions.** Nanomaterials  $Fe_2TiO_5$  and  $NiTiO_3$  added Ag were successfully synthesized. These materials showed excellent inhibition against *Bacillus cereus*, *Escherichia coli*, *Pseudomonas aeruginosa*, *Salmonella typhi*, and *Staphylococcus aureus*. Additionally, the Ag- $Fe_2TiO_5$  samples showed much better antibacterial activity than the Ag- $NiTiO_3$  sample.

**Keywords:** Iron(III) titanate, nickel titanate, *Jasminum subtriplinerve* Blume leaf, antibacterial activity

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## НАУЧНАЯ СТАТЬЯ

# Антибактериальная активность биосинтетических титанатов, допированных серебром

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### Аннотация

**Цели.** Синтезировать многофункциональные материалы титанаты с добавлением серебра путем восстановления золь-гелевых производных титанатов ( $Fe_2TiO_5$  и  $NiTiO_3$ ) экстрактом листьев *subtriplinerve* Blume жасмина.

**Методы.** Физико-химические характеристики полученных материалов определяли методами рентгеновской дифракции, энергодисперсионной рентгеновской спектроскопии, спектроскопии комбинационного рассеяния, удельной поверхности Брунауэра – Эммета – Теллера, сканирующей электронной микроскопии и абсорбционная спектроскопия в УФ-видимой области.

**Результаты.** Результаты показывают, что серебро имеет хорошую дисперсию на поверхности  $Fe_2TiO_5$  и  $NiTiO_3$  и создает фотокатализаторы с двумя светопоглощающими областями. Полученные материалы применялись в качестве антибактериальных средств в загрязненных водах. Образцы Ag- $Fe_2TiO_5$  (Ag-FTO) показали лучшие свойства и антибактериальную активность, чем Ag- $NiTiO_3$  (Ag-NTO) за счет лучшего диспергирования наночастиц серебра на поверхности FTO. Кроме того, антибактериальные результаты демонстрируют большую ингибирующую активность в отношении грамотрицательных (-) бактерий, чем в отношении грамположительных (+) бактерий.

**Выводы.** Успешно синтезированы наноматериалы  $Fe_2TiO_5$  и  $NiTiO_3$  с добавлением Ag. Полученные составы показали отличное ингибирование в отношении восковой бациллы (*Bacillus cereus*), кишечной палочки (*Escherichia coli*), синегнойной палочки (*Pseudomonas aeruginosa*), сальмонеллы тифи (*Salmonella typhi*) и золотистого стафилококка (*Staphylococcus aureus*). Кроме того, образцы Ag- $Fe_2TiO_5$  показали гораздо лучшую антибактериальную активность, чем образец Ag- $NiTiO_3$ .

**Ключевые слова:** титанат железа(III), титанат никеля, листья *subtriplinerve* Blume жасмина, антибактериальный

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## INTRODUCTION

The increasing number of industrial production and complex human life activities has seriously affected water quality creating safety concerns. Wastewater contains not only a large number of persistent pollutants but also many dangerous bacteria affecting human health [1, 2]. Therefore, there is an urgent need to overcome these problems, especially in developing countries.

Contemporary treatment methods such as chemical oxidizing agents, chlorination, ultraviolet (UV) radiation, membrane, ozonation, etc., have been considered for water treatment and disinfection purposes [3–5]. Although these methods have demonstrated effectiveness, they also have various disadvantages, such as the creation of toxic byproducts after treatment, high operating costs, and the inability to treat bacteria [3]. The main limitation of catalytic oxidation is the inability to kill bacteria present in wastewater. Semiconductor photocatalytic oxidation is one of the advanced oxidation processes being researched and applied in the wastewater treatment industry.

Although silver (Ag) is well known as a disinfectant against a wide range of bacteria having been widely used in sterilization and bactericidal applications, its practical application is limited due to the ease of oxidation, which can cause aggregation and loss of antimicrobial activity over time. Therefore, Ag requires a supporting substrate to enhance stability of

morphological features and maintain antimicrobial efficacy. Therefore, the research aimed at enhancing the antibacterial ability of photocatalyst materials by modifying them or adding another material with antibacterial properties is required.

The widely used  $TiO_2$  semiconductor catalyst has been commercialized as  $TiO_2$ -P25. However, its practical application is limited by the large bandgap energy (3.2 eV) and high recombination of electron and hole pairs [6, 7]. Therefore, the modification of  $TiO_2$  structure to improve its catalytic efficiency and stability has been receiving much interest from researchers along with the study of new potential semiconductor materials.

Perovskite-like materials possessing suitable properties, such as chemically inert, high mechanical strength, high absorption coefficient, low activation energy, and significant transparency, are being researched and developed for photocatalytic applications [6, 7]. Among them, titanate-based materials have shown their potential with high activity and stability.  $Fe_2TiO_5$  pseudobrookite has received much attention due to its low band gap energy (1.9–2.2 eV), higher energy level minimum conduction band, superior photochemical stability, and low cost, representing a promising photocatalyst product [8–10]. Another titanate,  $NiTiO_3$  perovskite, comprising an n-type semiconductor with an ilmenite-type crystal structure, has attracted considerable attention due to its superior photocatalytic and electro-optical properties and low dielectric constant [11, 12].  $NiTiO_3$  also exhibits an optical absorption

spectrum with band gap energy around 3.0 eV, offering excellent potential for visible light photocatalytic applications [13]. Thus, the Ag-added titanates can be used to create multifunctional composites with excellent photocatalytic antibacterial activity in wastewater. However, there are almost no studies on the addition of Ag to  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  to enhance their antibacterial ability.

In our research, multifunctional Ag-added titanates ( $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$ ) were synthesized by biosynthesis using *Jasminium subtripplinerve Blume* (JS) leaf extract as a reducing agent of  $\text{AgNO}_3$  solution to  $\text{Ag}^0$ , in which  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  are prepared by the sol-gel method. JS is a member of the Oleaceae genera. Compounds such as triterpenoids, oleanolic acid, flavonoids, glucosides, etc. [14–16], which have been identified and reported in its phytochemicals, can be employed as reducing reagents in the process of biosynthesizing silver nanoparticles. The physicochemical properties of the prepared nanoparticles have been investigated. The antibacterial action of prepared nanoparticles was evaluated with the help of bacteria such as gram-negative *Escherichia coli* (*E. coli*), *Pseudomonas aeruginosa* (*P. aeruginosa*), and *Salmonella typhi* (*Salmonella*), and gram-positive *Bacillus cereus* (*B. cereus*) and *Staphylococcus aureus* (*S. aureus*), which are typically present in wastewater.

## MATERIALS AND METHODS

Pseudobrookite  $\text{Fe}_2\text{TiO}_5$  and perovskite  $\text{NiTiO}_3$  were prepared by sol-gel method according to the procedure described in detail in the works [17, 18], respectively. First, 4.04 g of  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  (99.9%, Merck, Germany) or  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  (99.9%, Merck, Germany) and 2.1 g of  $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$  (99.9%, Merck, Germany) were dissolved with 5 mL of  $\text{C}_2\text{H}_5\text{OH}$  (99%, Merck, Germany). Next, 3 mL of  $\text{Ti}(\text{OC}_3\text{H}_7)_4$  (99.7%, Merck, Germany) was added drop by drop. The synthetic gel was dried at 60°C for 24 h and follows by heating at 700°C for 2 h to obtain a  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  samples.

Ag-doped  $\text{Fe}_2\text{TiO}_5$  (Ag-FTO) and Ag-doped  $\text{NiTiO}_3$  (Ag-NTO) catalysts were synthesized using JS leaf extract as a reducing agent of  $\text{AgNO}_3$  solution to  $\text{Ag}^0$ . JS leaf was collected from Ho Chi Minh City in Vietnam. After washing and shredding, JS leaf is dried at a temperature of 60°C for 4 h. Next, 50 g of the JS leaf were mixed with 1000 mL of deionized water and heated to 80°C for 2 h under stirring. Finally, the JS leaf extract

was filtered and preserved at 4°C for further experiments. Silver nitrate ( $\text{AgNO}_3$ , >99.8%, Merck, Germany) was purchased. The optimized conditions for the Ag nanoparticles (AgNPs) synthesis using JS extract as a reducing agent were determined based on our previous publication [16], involving the presence of the light illumination, a synthesis time of 150 min, a volume ratio of  $\text{AgNO}_3$  solution/JS extract of 18:2, a  $\text{AgNO}_3$  concentration of 1.0 mM and a stirring rate of 300 rpm at temperature of 20°C. To synthesize Ag-FTO and Ag-NTO,  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  powders were initially added together in a determined proportion with 1 mM  $\text{AgNO}_3$  precursor. The samples are denoted as  $x\text{Ag-FTO}$  and  $x\text{Ag-NTO}$ , where  $x$  represents the silver content added to titanates.

The physicochemical characteristics were studied including X-ray diffraction (XRD), Raman spectroscopy, scanning electron microscopy (SEM), Brunauer–Emmett–Teller adsorption (BET), energy dispersive X-ray spectroscopy (EDS), and UV–Vis absorption spectra techniques. The method of implementation is as detailed in our previous study [19].

The obtained samples have been tested for antibacterial activity against gram-negative *E. coli*, *P. aeruginosa*, and *Salmonella*, and gram-positive *B. cereus* and *S. aureus* by the minimum inhibitory concentration (MIC). These methods have been presented in our previous studies [20].

## RESULTS AND DISCUSSION

The XRD pattern (Fig. 1) of the Ag-FTO samples shows mainly diffraction peaks of the pseudobrookite  $\text{Fe}_2\text{TiO}_5$  at  $2\theta = 18.2^\circ, 25.7^\circ, 27.6^\circ, 32.7^\circ, 37.5^\circ, 40.6^\circ, 46.3^\circ, 49.0^\circ, 52.5^\circ,$  and  $60.1^\circ$  (JCPDS<sup>1</sup> card No. 41-1432). In addition, a few other characteristic peaks of rutile with low intensity at  $2\theta = 27.3^\circ, 36.1^\circ,$  and  $41.5^\circ$  (JCPDS card No. 21-1276) were observed. There was practically no appearance of characteristic peaks of anatase (JCPDS card No. 21-1272). Some silver peaks with low intensity were also observed for Ag-FTO samples at  $2\theta = 37.4^\circ, 39.0^\circ,$  and  $41.1^\circ$  (JCPDS card No. 04-0783). Diffraction peaks of  $\text{Ag}^0$  species were observed more clearly in the 1.0Ag-NTO sample, where Ag concentration was loaded up to 1.0 wt %. The phase composition of the Ag-FTO sample completely coincides with that of the pure  $\text{Fe}_2\text{TiO}_5$  sample [17], indicating that Ag does not change the phase structure of

<sup>1</sup> JCPDS (Joint Committee on Powder Diffraction Standards)—International Center for Diffraction Data.

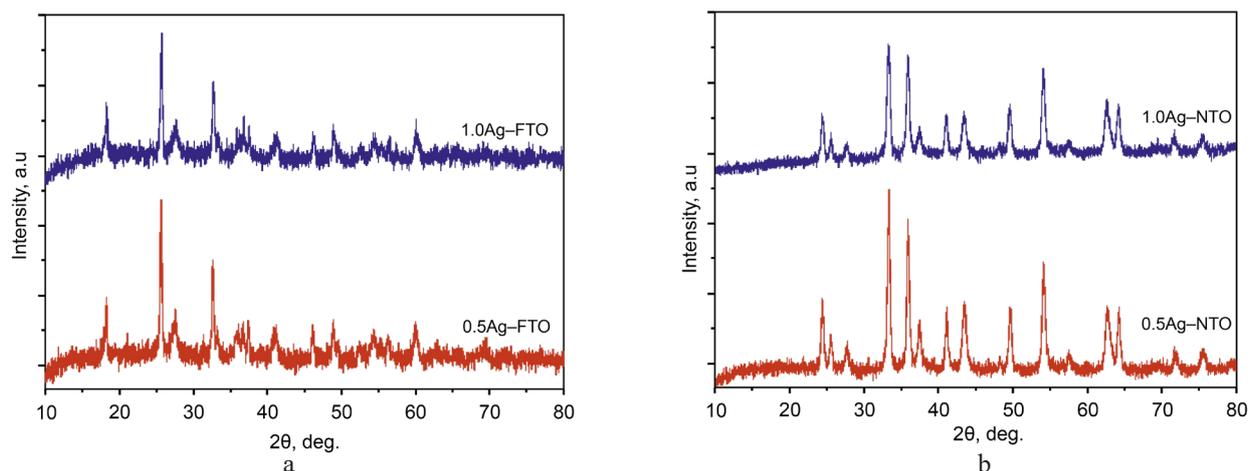


Fig. 1. XRD patterns of Ag-FTO (a) and Ag-NTO (b) catalysts.

pseudobrookite. However, the presence of Ag reduced the crystal size of  $\text{Fe}_2\text{TiO}_5$  from 31.6 nm to 18.6 nm as calculated based on the Scherrer equation. For the Ag-NTO samples, the peaks of  $\text{NiTiO}_3$  were observed at  $2\theta = 23.9^\circ; 32.8^\circ; 35.4^\circ; 53.4^\circ; 61.9^\circ; \text{ and } 63.5^\circ$  (JCPDS 75-3757). The XRD spectra of these samples also demonstrated the presence of anatase crystalline phase at  $2\theta = 25.4^\circ$  and rutile crystalline phase at  $2\theta = 27.6^\circ$ , as well as some characteristic peaks of Ag at  $2\theta = 37.4,$

$39.0,$  and  $41.1^\circ$ . The average crystal size of  $\text{NiTiO}_3$  is 17–18 nm, which were approximately the same for the Ag-FTO samples. From the XRD diagram, the crystal size of  $\text{Ag}^0$  is estimated to be a few nm.

The elemental compositions of the 0.5Ag-FTO and 0.5Ag-NTO samples were determined by EDS analysis. EDS images show that both silver-modified samples have a uniform distribution of Ag crystals on the  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  surfaces (Fig. 2). In which, the Ag distribution is more

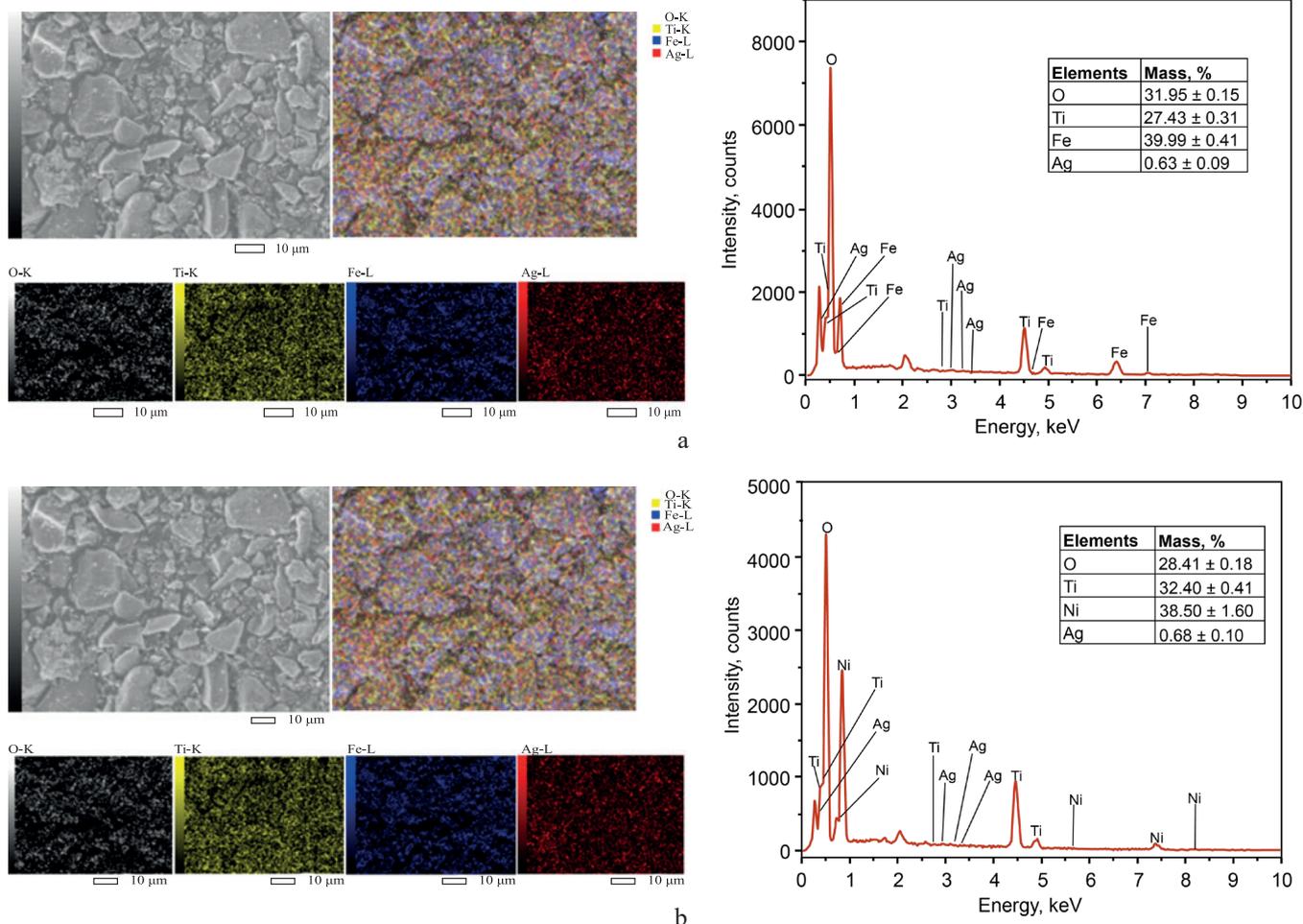


Fig. 2. EDS analysis of 0.5Ag-FTO (a) and 0.5Ag-NTO (b) samples.

dominant for 0.1Ag–FTO sample than that of 0.1Ag–NTO sample. Meanwhile, the EDS spectrum of the 0.1Ag–FTO sample shows the characteristic peaks of Fe, Ti, O, and Ag. The mass ratio of these elements corresponds to 39.99:27.43:31.95:0.63, which is nearly equivalent to the theoretical mass ratio of 0.1Ag–FTO (46.43:19.09:33.17:0.50). The EDS spectrum of sample 0.1Ag–NTO also shows characteristic peaks of Ni, Ti, O, and Ag with mass ratios 38.50:32.40:28.41:0.68, respectively. This ratio is also quite close to the theoretical mass ratio of 0.1Ag–NTO (37.78:30.82:30.90:0.50). Comparing 0.1Ag–FTO and 0.1Ag–NTO samples, the 0.1Ag–FTO sample seems to have a more similar experimental mass ratio of elements and theoretical mass ratio than that of 0.1Ag–NTO, demonstrating the higher purity of this sample. In addition, some unassigned peaks on the EDS spectra of both samples were characteristic peaks of the carbon tape material covering the samples. From the EDS and XRD results, it can be concluded that the silver was formed and uniformly dispersed on the surface as well as inside the structure of  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$ .

The Raman spectrum of Ag–FTO samples (Fig. 3) shows the appearance of absorption peaks characteristic for  $\text{Fe}_2\text{TiO}_5$  structure at  $144\text{ cm}^{-1}$ ,  $199\text{ cm}^{-1}$ ,  $222\text{ cm}^{-1}$ ,  $292\text{ cm}^{-1}$ ,  $327\text{ cm}^{-1}$ ,  $394\text{ cm}^{-1}$ ,  $661\text{ cm}^{-1}$ ,  $787\text{ cm}^{-1}$ , and  $1294\text{ cm}^{-1}$  [21]. Here, the characteristic Ag oscillations at  $199$ ,  $327$ , and  $787\text{ cm}^{-1}$  correspond to the strain vibration in the inner plane of the O–Ti bond, the symmetric stretching vibration of the O–Fe bond, and the symmetrical bending vibrations of the O–Ti bond. Meanwhile, the  $B_{1g}$  oscillations at  $144\text{ cm}^{-1}$ ,  $222\text{ cm}^{-1}$ , and  $661\text{ cm}^{-1}$  are formed by the out-of-plane rotation of the O–Ti bond, bending vibrations in the plane of the O–Fe bond, and external

bending vibrations of the Ti–O bond, respectively. Moreover, the characteristic peaks of  $\text{Fe}_2\text{O}_3$  are not significantly present in the Raman spectrum. Therefore, this result proves that the obtained  $\text{Fe}_2\text{TiO}_5$  has high crystallinity with a stable phase structure. Meanwhile, for Ag–NTO samples, the Raman vibrational modes for  $\text{NiTiO}_3$  are located at  $192\text{ cm}^{-1}$  ( $A_g$ ),  $229\text{ cm}^{-1}$  ( $B_g$ ),  $246\text{ cm}^{-1}$  ( $A_g$ ),  $291\text{ cm}^{-1}$  ( $B_g$ ),  $345\text{ cm}^{-1}$  ( $B_g$ ),  $394\text{ cm}^{-1}$  ( $A_g$ ),  $465\text{ cm}^{-1}$  ( $B_g$ ),  $484\text{ cm}^{-1}$  ( $A_g$ ),  $613\text{ cm}^{-1}$  ( $B_g$ ), and  $709\text{ cm}^{-1}$  ( $B_g$ ). The characteristic  $A_g$  and  $B_g$  oscillations correspond to the strain vibration in the inner plane of the O–Ti bond, symmetrical bending vibrations of the O–Ti bond, and the symmetric stretching vibration of the O–Ni bond. Since the Raman spectrum shows almost no characteristic oscillations of Ag for both Ag–FTO and Ag–NTO samples, it can be concluded that silver is present in low concentrations and well dispersed on the surface of  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$ .

SEM images (Fig. 4) show that the Ag–FTO and Ag–NTO materials exist in the form of small quasi-spherical particles and large bulk. For both Ag–FTO and Ag–NTO samples, samples containing 0.5% Ag exhibited less agglomeration, smaller clumps, and better dispersion of brightly colored spherical Ag particles. This means that there will be better porosity for bacteria location and conversion.

Figure 5a shows the  $\text{N}_2$  adsorption-desorption isotherms of 0.5Ag–FTO and 0.5Ag–NTO samples. According to the IUPAC nomenclature [22], the shape of the nitrogen isotherm of two samples could be considered to be a type IV isotherm, which is typical for mesoporous materials. Here, the width of the hysteresis loop of the Ag–FTO sample is much higher than that of the Ag–NTO sample, demonstrating the better adsorption capacity of this material for bacteria. The curve of

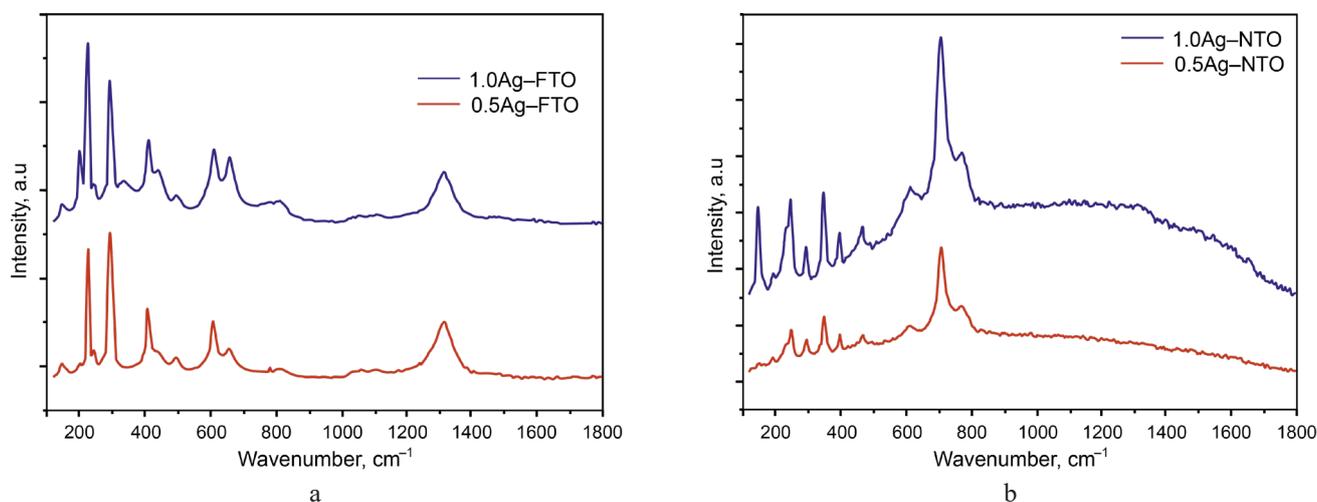


Fig. 3. Raman spectra of Ag–FTO (a) and Ag–NTO (b) catalysts.

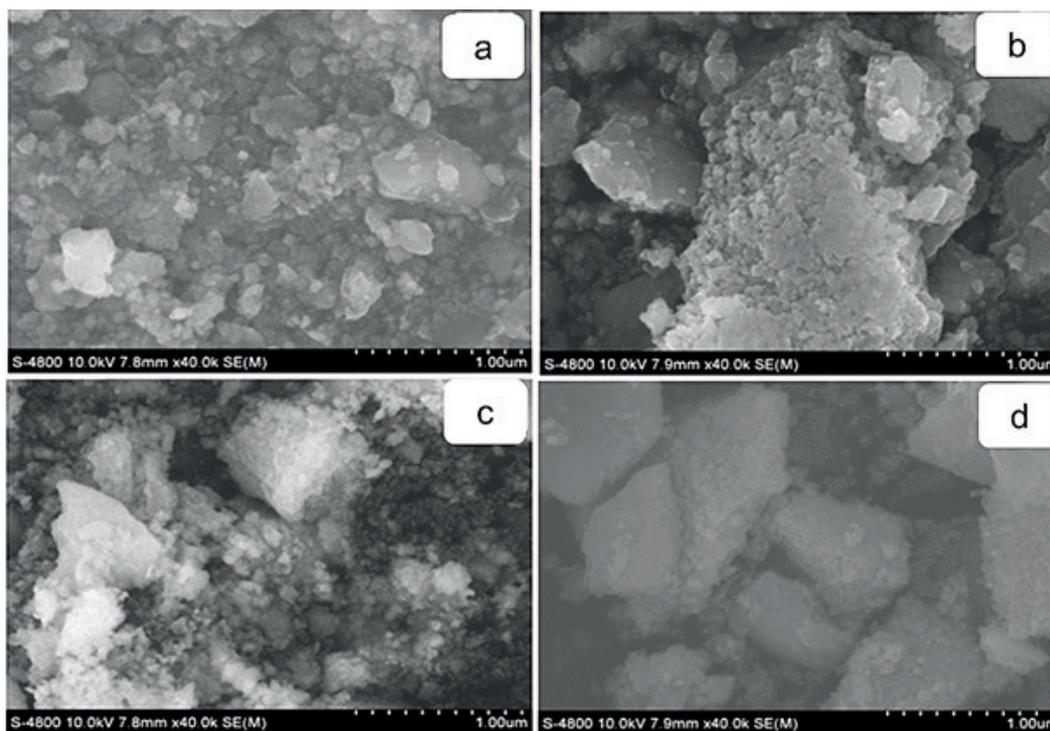


Fig. 4. SEM images of samples: 0.5Ag-FTO (a), 1.0Ag-FTO (b), 0.5Ag-NTO (c), 1.0Ag-NTO (d).

the pore size distribution of the 0.5Ag-FTO and 0.5Ag-NTO samples according to their diameters additionally allowed the maximum diameter densities of 24.0 Å and 23.4 Å, respectively, to be determined (Fig. 5b). These were used to calculate the BET data for 0.5Ag-FTO and 0.5Ag-NTO samples. For the 0.5Ag-FTO sample, the specific surface area of the examined sample was found to be 20.2 m<sup>2</sup>/g, while the total pore volume was found to be 0.025 cm<sup>3</sup>/g. Meanwhile, these values reach 13.8 m<sup>2</sup>/g and 0.017 cm<sup>3</sup>/g for the 0.1Ag-NTO sample, respectively. These results demonstrate that Ag-FTO has better adsorption capacity than the Ag-NTO.

The absorbed wavelength and the band gap energy of the catalysts were determined by UV-Vis spectroscopy and the Tauc plot. The obtained results show that there is almost no difference in absorption wavelength and band gap between Ag-added samples. Specifically, for the Ag-FTO samples (Fig. 6a), it can be seen that the absorbed light band and band gap energy form two different regions: the first region has an absorption wavelength range in the range of 630–633 nm corresponding to a band gap energy of 1.96–1.97 eV. The second region has an absorption wavelength range of 493–496 nm corresponding to a band gap energy of 2.50–2.52 eV.

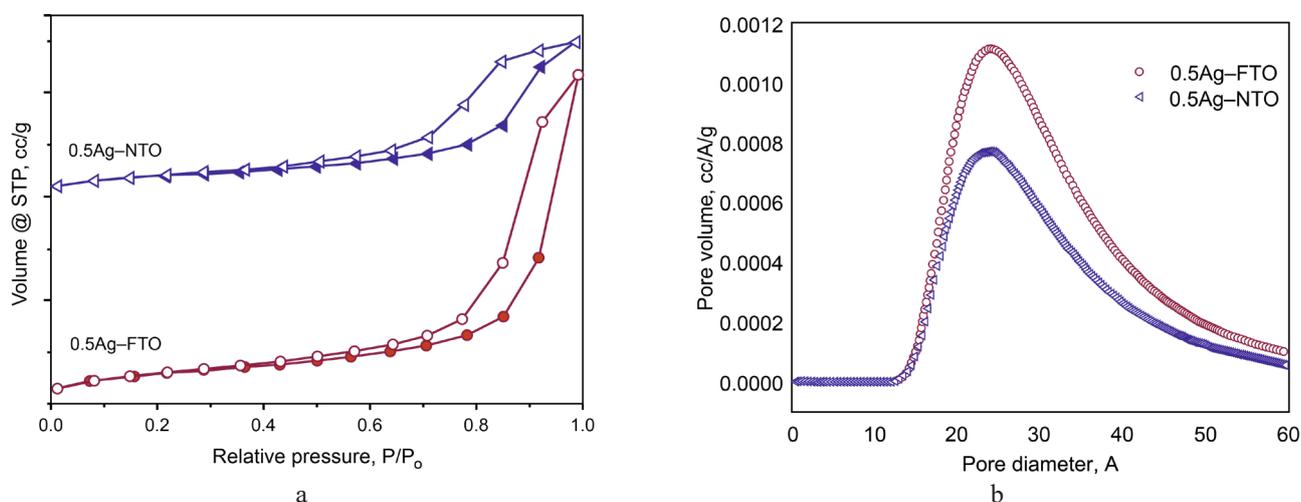


Fig. 5. N<sub>2</sub> adsorption-desorption isotherms (a) and pore size distribution (b) of 0.5Ag-FTO and 0.5Ag-NTO samples.

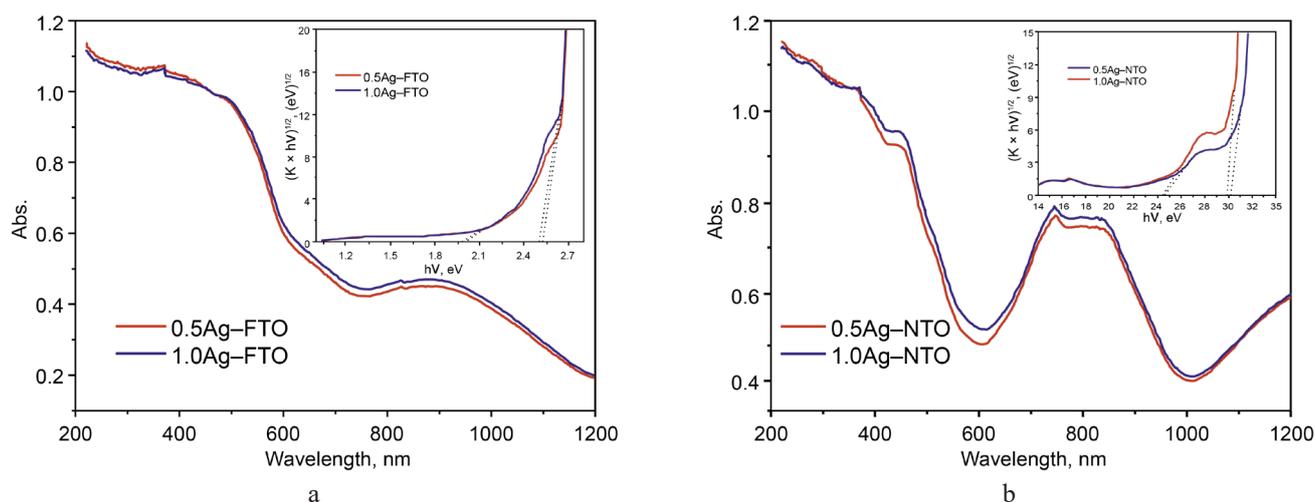


Fig. 6. UV-Vis diffuse reflectance spectra and Tauc plot of the Ag-FTO (a) and Ag-NTO (b) samples.

According to previous studies [8–10],  $\text{Fe}_2\text{TiO}_5$  has a band gap energy of 1.9–2.1 eV (corresponding to an absorption wavelength of about 590–650 nm), which is consistent with the first band data; meanwhile, silver has an absorption wavelength in the range of 400–500 nm (band gap energy in the range of 2.4–3.1 eV) [23], which is consistent with the second band parameters. The appearance of two absorption wavelength regions and band gap energies is similar for Ag-NTO samples (Fig. 6b). In which, the first region corresponds to 411–414 nm and 3.0–3.02 eV; and the second region is 504–406 nm and 2.45–2.46 eV, respectively. According to author [13],  $\text{NiTiO}_3$  has a band gap energy of about 3.0 eV, which is consistent with the first band results of the catalysts, while the second is also compatible with the absorption region of silver. It can be concluded that the combination of silver with  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  leads to the appearance of a smaller frequency band, which increases flexibility in the light absorption of the material. It can be found that the Ag-FTO materials with bandgap absorb light and bandgap better than the Ag-NTO samples. This is an essential property of photocatalyst materials, which can lead to their improved activity.

The antibacterial properties of the samples were determined by their minimum inhibitory concentration (MIC). Five types of bacterial including *E. coli*, *P. aeruginosa*, *Salmonella*, and *S. aureus* were used in this study. The exponential phase of bacteria was observed to be delayed in the presence of samples; this phenomenon was more obvious with the rise of Ag-FTO and Ag-NTO concentrations (Fig. 7). The samples were able to inhibit the exponential stage of both gram-negative and gram-positive bacteria. The MIC against *B. cereus*, *E. coli*, *P. aeruginosa*, *Salmonella*, and *S. aureus* of Ag-FTO and Ag-NTO samples are described

in Table. Here it can be seen that the 0.5Ag-FTO were able to completely inhibit the growth of *B. cereus*, *E. coli*, *P. aeruginosa*, *Salmonella*, and *S. aureus* at a similar MIC of 1.25, 0.31, 1.25, 1.25, and 2.50  $\mu\text{g}/\text{mL}$ , respectively. The almost doubled MIC value when increasing the silver content up to 1.0% for the  $\text{Fe}_2\text{TiO}_5$  samples (1.0Ag-FTO) demonstrates their enhanced antimicrobial activity with the addition of silver. Meanwhile, the antibacterial ability of 0.5Ag-NTO proved to be lower than that of 0.5Ag-FTO with MIC values of 5.00, 2.50, 2.50, 2.50, and 5.00  $\mu\text{g}/\text{mL}$  respectively. A similar phenomenon was observed for the 1.0Ag-FTO sample with an improvement in antibacterial activity with increasing Ag concentration. It is clear that the Ag-FTO samples exhibit much higher antibacterial activity than Ag-NTO due to the better dispersion of silver nanoparticles on the  $\text{Fe}_2\text{TiO}_5$  surface (shown by their physicochemical properties). The synthesized samples in this study were shown to be more effective against the negative bacterial (*E. coli*, *Salmonella*, and *P. aeruginosa*) as compared to the positive bacterial (*B. cereus* and *S. aureus*). These results were the agreement with previous reports [24, 25] when assuming that the gram-negative bacteria had thinner cell walls than the gram-positive bacteria; thus, the antibacterial materials can more easily penetrate and inhibit gram-negative bacteria.

As compared to some similar earlier reported materials, the obtained materials exhibit superior antibacterial ability. Specifically, silver nanoparticles (AgNPs) synthesized using *Acalypha indica* leaf extract had an MIC of 10  $\mu\text{g}/\text{mL}$  against *E. coli* and *Vibrio cholera* (*V. cholera*) [26], while  $\text{LaAlO}_3$  perovskite nanoparticles had an MIC of 63  $\mu\text{g}/\text{mL}$  against gram-negative bacteria *P. aeruginosa* [27] and

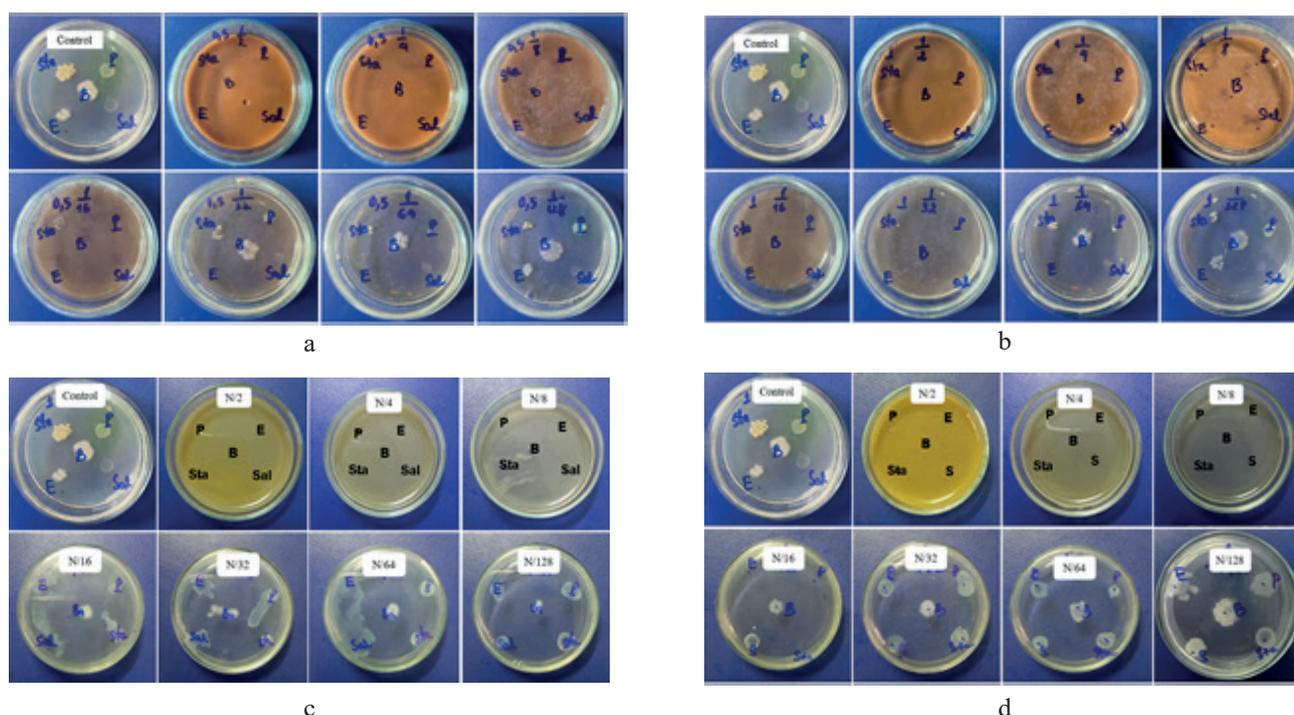


Fig. 7. Minimum inhibitory concentrations of 0.5Ag-FTO (a), 1.0Ag-FTO (b), 0.5Ag-NTO (c), 1.0Ag-NTO (d) samples against five bacteria.

Table. Minimum inhibitory concentrations (MIC) on samples against five bacteria

Bacteria	MIC (µg/mL)			
	0.5Ag-FTO	1.0Ag-FTO	0.5Ag-NTO	1.0Ag-NTO
<i>Bacillus cereus</i>	1.25	0.63	5.00	2.50
<i>Escherichia coli</i>	0.31	0.31	2.50	1.25
<i>Pseudomonas aeruginosa</i>	1.25	0.63	2.50	1.25
<i>Salmonella topfi</i>	1.25	0.63	2.50	1.25
<i>Staphylococcus aureus</i>	2.50	1.25	5.00	2.50

the modified perovskite material  $\text{LaCo}_{0.4}\text{Fe}_{0.6}\text{O}_3$  showed an MIC of 63 µg/mL against two bacteria *S. aureus* and *P. aeruginosa* [28]. Studies on perovskite materials and silver nanoparticles have shown effective bactericidal ability against both gram-negative and gram-positive bacteria. Perovskite-like materials combined with silver not only release metal ions  $\text{Fe}^{3+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Ti}^{+4}$  but also  $\text{Ag}^+$  ions having a positive charge, causing electrostatic attraction with negative charge of bacterial cell membrane. As a consequence, ionic particles easily adhere to cell walls and membranes, changing the membrane structure to destroy the bacterial cell [29, 30].

### CONCLUSIONS

In summary, Ag-added  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  nanomaterials were successfully synthesized by

biosynthesis combined with the sol-gel method. Well-distributed silver nanoparticles on  $\text{Fe}_2\text{TiO}_5$  and  $\text{NiTiO}_3$  surfaces on a nanoscale and with a low band gap were observed for the samples. The antibacterial efficacy of Ag-FTO and Ag-NTO samples has been demonstrated through excellent inhibition against *B. cereus*, *E. coli*, *P. aeruginosa*, *Salmonella*, and *S. aureus* with the low MIC minimum inhibitory concentration due to the excellent dispersion of Ag on the surface of the perovskite. At the same time, the Ag-FTO samples also showed much better antibacterial activity than the Ag-NTO samples as a result of the excellent dispersion of Ag on the  $\text{Fe}_2\text{TiO}_5$  surface. Hence, according to the findings of this research, the use of Ag-FTO nanomaterials may suggest potential applications in disinfection and sterilization applications in future wastewater treatment approaches.

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**Authors' contribution**

All authors equally contributed to the research work.

**Вклад авторов**

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