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New antimicrobial systems based on zeolites with RE = La, Gd functional ions

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INTRODUCTION & AIM

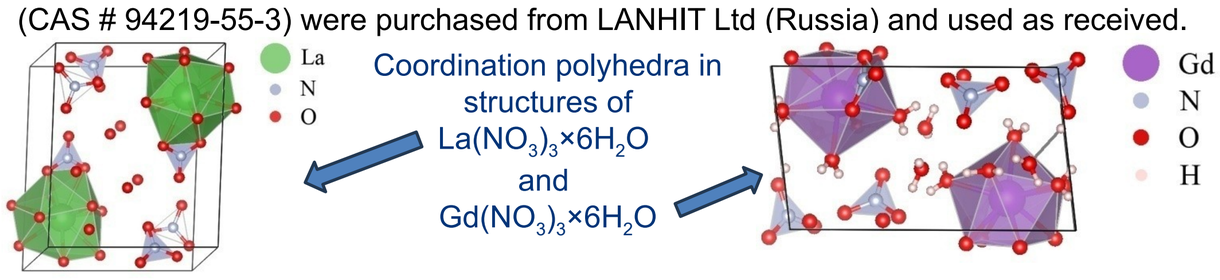
The antibiotic resistance necessitates the transition to fundamentally new drugs, in particular, *RE*$(NO_3)_3\times$ *x*H_2O which have good antimicrobial properties [*Kuz'micheva G.M. et all. Crystallography Reports. 2020. V. 65. P. 922-932*]. To reduce the content of the active substance (*RE* ions) in the preparation while maintaining its functional effect, auxiliary components are added, which additionally introduce aesthetic and/or new functions. These components include zeolites: white powdery substances, biocompatible, inexpensive, with a large specific surface area.

THE PURPOSE OF THIS WORK is to create new composites based on two types of zeolites (MFI and BEA) with different silicate modules and functional particles *RE*$(NO_3)_3\times6H_2O$ (*RE*=La,Gd).

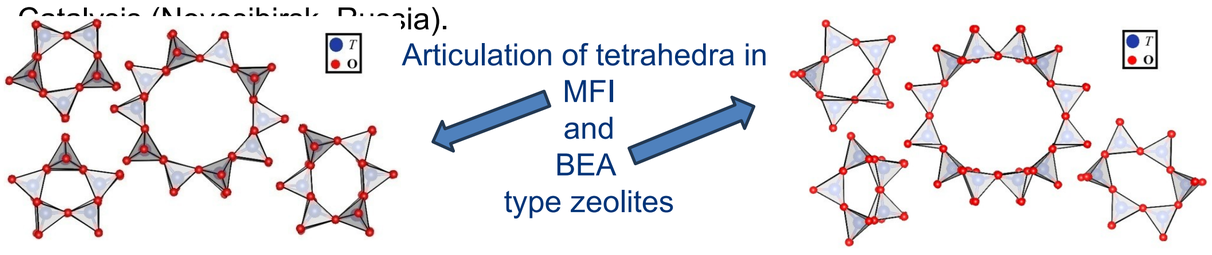
MATERIALS & METHODS

❑ MATERIALS:

***RE*$(NO_3)_3\times x H_2O$ (*RE* = La, Gd) salts** with 99.9% La (x=6) (CAS # 10277-43-7), Gd (CAS # 94219-55-3) were purchased from LANHIT Ltd (Russia) and used as received.



MFI type zeolites of compositions $(H_x)[(Fe^{3+}_xSi^{4+}_{12-x})O_{24}]\times wA$ (Si/Fe = 25, 68) and $[(Ti^{4+}_xSi^{4+}_{12-x})O_{24}]\times wA$ (Si/Ti = 47, 60) **and BEA type zeolites** of composition $(H_x)[(Al^{3+}_xSi^{4+}_{12-x})O_{24}]\times wA$ (Si/Al = 12, 150) were synthesized at Boreskov Institute of Catalysis (Novosibirsk, Russia).



❑ SINTHESIS:

Composites in the system "zeolite (MFI type with Si/Fe = 25, 68 and with Si/Ti = 30, 47) or BEA type (Si/Al = 12, 150))-*RE*$(NO_3)_3\times6H_2O$ (*RE* = La, Gd)" were synthesized using the **cold impregnation method**: solid-phase mixing of the components (1:1.2), grinding (~4 min), annealing (250°C, 1 hour).

❑ METHODS:

X-Ray Powder Diffraction: HZG- diffractometer, CuKα_1, λ = 1.54051 Å, graphite flat monochromator, sample rotation, continuous shooting: pulse acquisition time 5 seconds, step 0.05°, angle range 2θ = 2°-50°.

Scanning Electron Microscopy: scanning electron microscope MINISEM A5100.

Disk diffusion method: antimicrobial activity of the composites was assessed against bacteria (*Staphylococcus aureus, Escherichia coli, Pseudomonas aeruginosa, Acinetobacter baumannii, Klebsiella pneumoniae*) and fungi (*Candida albicans, Candida glabrata, Candida parapsilosis*). The results were assessed by the size of the growth inhibition zone (D, mm) using a ruler.

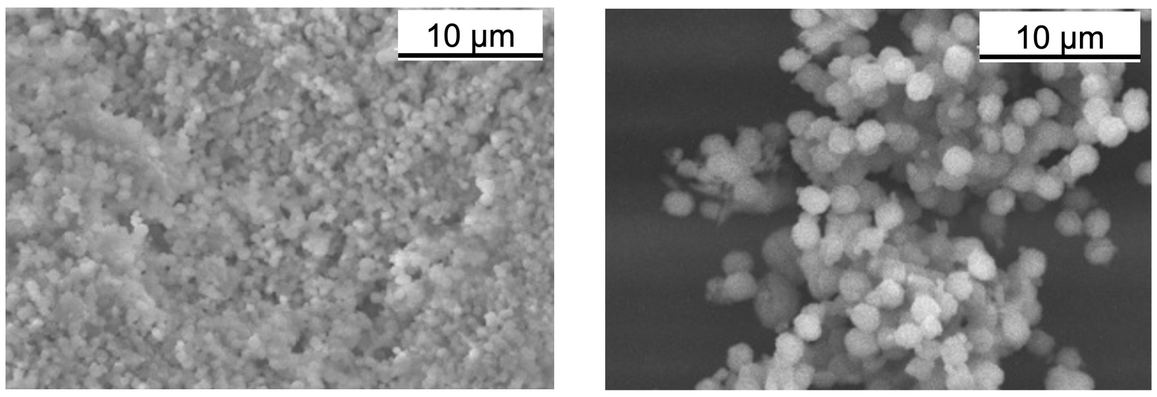
RESULTS & DISCUSSION

Characteristic X-Ray patterns:

An amorphous component (blue arrow) was found in the diffraction patterns of composites in the system **"BEA zeolites–*RE*$(NO_3)_3\times6H_2O$ (*RE* = La, Gd) salts"** in contrast to the diffraction patterns of composites **"MFI zeolites–*RE*$(NO_3)_3\times6H_2O$ (*RE* = La, Gd) salts"**, where amorphous components were not detected.

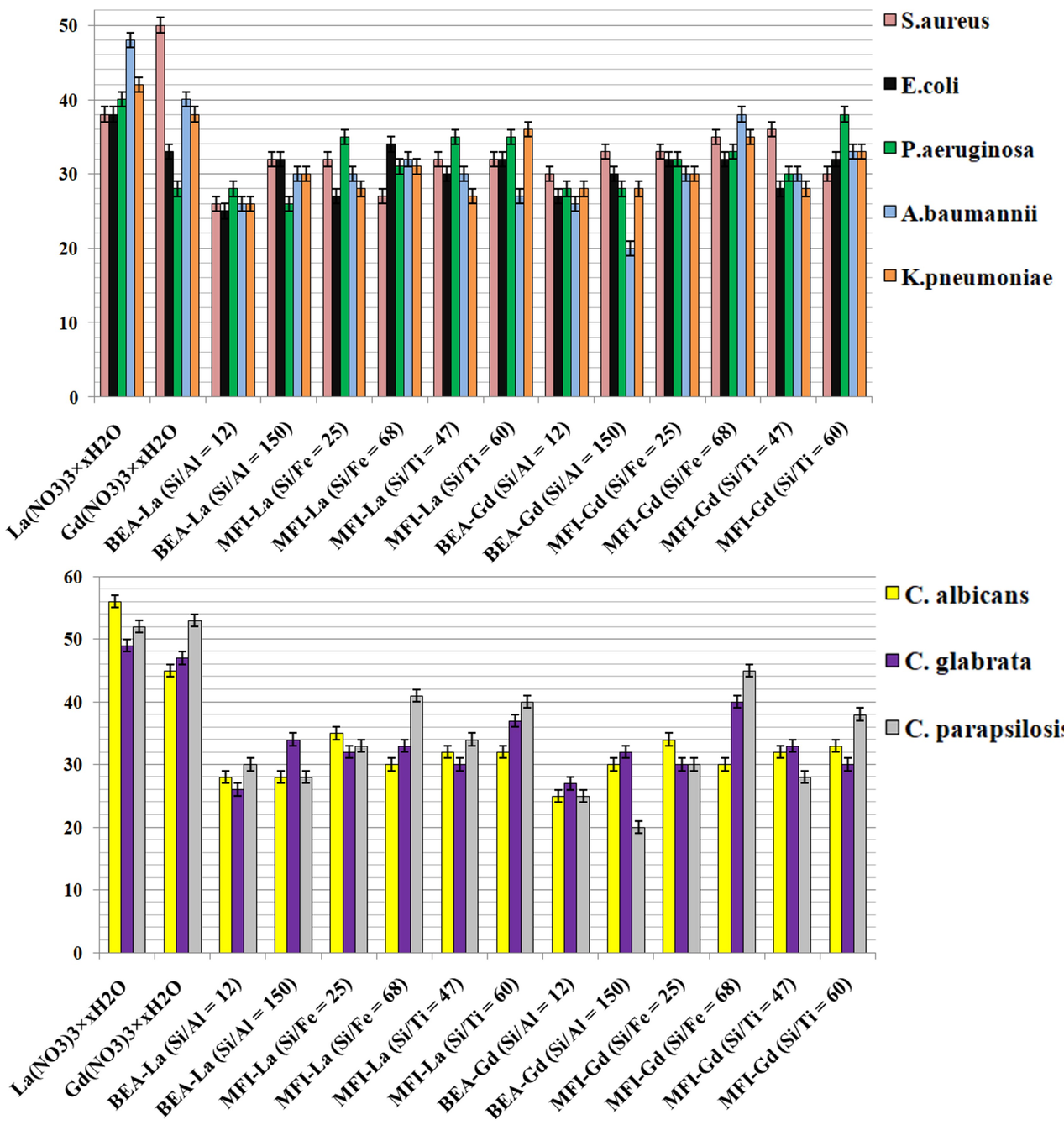
Characteristic SEM images:

BEA-La (Si/Al = 150) **MFI-La (Si/Fe = 68)**



The composites in the system **"MFI zeolites–*RE*$(NO_3)_3\times6H_2O$ (RE = La, Gd) salts"** have larger particle sizes (*N*=12.5-35 μm) than composites **"BEA zeolites–RE$(NO_3)_3\times xH_2O$ (RE = La, Gd) salts"** (*N*=7.5-8 μm) except for samples with MFI (Si/Ti=60) the particle sizes of which are comparable with BEA-based ones.

ANTIMICROBIAL PROPERTIES



Growth inhibition zone of bacteria and fungi on salts change in the range of 28-50 mm with D_{max} for *S.aureus* on *Gd*$(NO_3)_3\times6H_2O$, and in the range of 45-56 mm with D_{max} for *C.albicans* on *La*$(NO_3)_3\times6H_2O$, which is a record for these microorganisms; D=0 mm for zeolites. Microorganisms showed high sensitivity to composites with **D_{max}=45 mm** for *P.aeruginosa* on composite **MFI zeolite–*Gd*$(NO_3)_3\times6H_2O$ (Si/Fe=68)**, but less compared to salts, while maintaining their excellent biocidal properties.

CONCLUSION

Introduction of ***RE* = La, Gd** ions in the form of ***RE*$(NO_3)_3\times6H_2O$** salts into biocompatible zeolites (**MFI and BEA types**) makes it possible to implement antimicrobial properties that are almost not inferior in D values to the salts, and to ***significantly reduce the cost of finished products***. The antimicrobial activity of the obtained composites is higher than that of the broad-spectrum antibiotic penicillin, which makes such systems ***promising for biomedical purpose***.

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