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Reline deep eutectic solvent mediated synthesis of lanthanum titanate for heavy metal remediation and photocatalytic degradation

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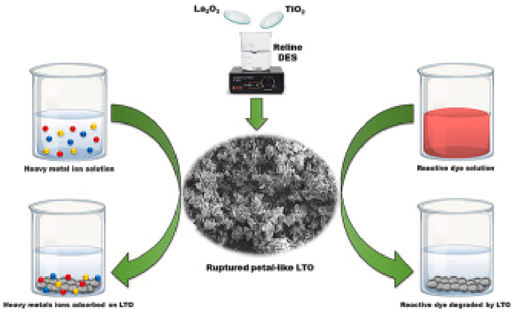
Abstract

Toxic heavy metal and dye contamination are potential threats that mutilate the essential triad of life; air, water and soil. Despite commercial applicability and importance, the over accumulation of these noxious toxicants has become a disturbing concern. As a result, their remediation has drawn greater fascination leading to the inexplicable quest for a material which can act as both an adsorbent and as a photocatalyst. The present work highlights a novel solid-state technique assisted with reline (Choline chloride: Urea) deep eutectic solvent for the synthesis of lanthanum titanate. The synthesized material was established with physical characterizations like PXRD, FT-IR, UV-DRS, BET, XPS, HR-SEM and TEM techniques. Further, the ruptured petal-like lanthanum titanate was integrated as an adsorbent for the removal of lead (Pb), arsenic (As) and chromium (Cr) heavy metals. The adsorbent presented increased adsorption efficiencies of 96, 74 and 71% towards Pb, As and Cr respectively. Dependence of the degradation efficiency over concentration, pH, contact time and competitive environments were analyzed and inferred. Furthermore, lanthanum titanate was used for the photocatalytic degradation of reactive black (RB5), red (RR198) and yellow (RY145) dyes. The degradation efficiencies were found to be 68.31, 85.2 and 96.8% for RB5, RR198 and RY145 dyes respectively. Variation in concentration and pH of the dye solutions were examined and reaction kinetics was also proposed. In conclusion, the as synthesized lanthanum titanate is assured to play dual roles as a versatile cost-effective adsorbent for the remediation of heavy metals and as a potential candidate for photocatalytic degradation.

Graphical abstract



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Introduction

The essential combination of air, water and soil is the triad that mirrors the well-being of the entire ecosystem. Various divergences that interrupt this triad simultaneously disrupt the nature's chain of evolution. Two such manifestations are the accumulation of toxic heavy metals (HMs) and dyes that are produced either by natural calamities or man-made actions. HMs are generally grouped as metals/metalloids whose atomic weights fall in the range 63.5–200.6 g/mol with atomic densities and specific gravities greater than 4000 kg/m^3 and 5 g/cm^3 respectively (Velusamy et al., 2021). They originate from a variety of sources including mining, petrochemical production, textile industry, fertilizers, pharmaceuticals, tanneries, electroplating etc (Zhou and Wang, 2019). Environmental conditions like flooding and soil erosion lead to the spread of these HMs to nearby water sources and eventually to human consumption. Adequate number of HMs such as lead, mercury, copper, nickel, cobalt, chromium, arsenic, cadmium, iron, selenium etc are present in the environment. Amongst these lead, arsenic and chromium are frequently found nonetheless hard to recover category of elements. Several environmental governing bodies have declared that the exposure to lead, arsenic and chromium HMs are lethal beyond 0.05 mg/L concentration (Malik et al., 2019). In terms of aquatic lives, filter-feeding creatures like crustaceans actively absorb HMs from seawater and influence human beings over future ingestion (Kumar et al., 2021).

Harmful effect of these HMs ranges from common ailments and extend to target-specific malignancy which ultimately leads to death. Lead (Pb) causes oxidative stress which results as a consequence of imbalance that takes place between the free-radical production and its counter antioxidants. Pb poisoning results in loss of appetite, fatigue, sleeplessness, renal dysfunction, allergies, paralysis, mental retardation, brain and kidney damage etc. Arsenic (As) on the other hand is a protoplasmic poison which can induce mitosis accompanied with breakdown of the cell enzymatic and respiratory processes. Some general effects of As toxicity include abnormal heart beat, pulmonary disease, nausea, skin lesions, hypertension, arsenicosis, keratosis, neurological and peripheral vascular disorder etc. Chromium (Cr) is a dynamic phytotoxic element capable of depressing the growth of roots and seed germination. Exposure to high levels of Cr leads to nasal septum ulcer, DNA damage, chromosomal aberration, and hinders the reduction of methemoglobin (Jaishankar et al., 2014; Rehman et al., 2021).

Thus the HM contamination owed to the non-degradable, bioaccumulation and toxic nature becomes a ubiquitous threat to nature (Zhao et al., 2021) and it is of primary concern to remediate these HMs from the environment rather than only detection. Conventional methods used for the removal of HMs are membrane separation, chemical precipitation, reverse osmosis, ion exchange, electrochemical treatment, coagulation-flocculation and adsorption (Chakraborty et al., 2020). In comparison, adsorption serves as an exclusive technique in terms of its lower cost, eco-friendliness, tunability and superior efficiency in HM remediation. Over the years, researchers are enhancing the efficiency of traditional adsorbents at the same time innovative adsorbents are also being discovered. Semiconductors, polymers, ceramics, bio-waste, metal-organic frameworks and carbon based materials are gaining tremendous attraction towards adsorption.

Alike HM accumulation, water resources face yet another severe contamination due to textile dyes. According to a study, extreme amounts (280,000 tons) of textile dyes are discharged into water streams every year (Solís et al., 2012). It is noteworthy to mention that beyond various safety measures, almost fifteen percent textile dyes escape from textile processes and reach the common water system reluctantly (Saggioro et al., 2011). Synthetic dyes can be normally considered as cationic, anionic and non-ionic categories. Among the anionic classification reactive dyes play a vital role in terms of their augmented use in commercial dyeing. Reactive dyes are known for their versatile chemical structures that contain various functional groups which impart specified properties like extended shelf-life, wear and tear resistance etc. More specifically, industrial dyes such as reactive black (RB5), reactive red (RR198) and reactive yellow (RY145) contain azo linkages (-N═N) and sulphonic linkages (-SO^{3-}) which enhance their physical and chemical properties (Shindhal et al., 2021; Sintakindi and Ankamwar, 2021).

Water contaminated with these dye moieties tend to turn mutagenic and carcinogenic to living beings. Some acute conditions like heart disease, kidney failure, splenic carcinoma, dermatitis, metabolic stress and reproductive dysfunction can be triggered over ingestion of such contaminated water. Water bodies experience an increase in turbidity which in turn blocks sunlight from reaching the water-based organisms. This leads to slow decomposition of micro-organisms and produces foul odor in the neighborhood (Yeow et al., 2021; Nithya et al., 2021). Various physical, chemical and biological methods have been developed for removing such toxic dyes from industrial effluents and water reservoirs (Can et al., 2006; Sathishkumar et al., 2019). Owed to scalability, cost effectiveness, eco-friendly nature and ease of process, photocatalysis has gained over the traditional methods for degradation of textile dyes. Photocatalysis takes place when photons trigger the production of highly active reactive oxygen species such as hydroxyl radicals (Li et al., 2021).

Conclusively, this investigation will pioneer a novel reline (choline chloride and urea in 1:2 ratio) deep eutectic solvent assisted solid-state strategy for synthesizing lanthanum titanate (LTO). Further, the synthesized lanthanum titanate will be incorporated as an adsorbent for aiding the adsorption-based remediation of heavy metals such as lead (Pb), arsenic (As) and chromium (Cr). Besides, lanthanum titanate will also be used as a photocatalyst for degrading reactive dyes such as reactive black 5 (RB5), reactive red 198 (RR198) and reactive yellow 145 (RY145) under several concentrations and pH.

Section snippets

Chemicals and instruments

Analytical grade lanthanum oxide, titanium dioxide, choline chloride, urea, lead acetate, sodium arsenite, potassium dichromate and reactive dyes RB5, RR198 and RY145 were bought from Sigma. Ultrapure distilled water was used during the course of the analysis. PXRD data was obtained at 10-80° 2θ range with CuKα radiation in a Bruker D8 advanced powder X-ray diffractometer. FT-IR spectrum in a 400-4000 cm^{-1} range was studied with a Spectrum two instrument. UV-DRS spectrum in 200–800 nm…

PXRD

The distinctive diffraction pattern displayed in Fig. 1 evidenced the crystallinity and phase-pure nature of LTO. Owing to the formation of LTO, significant peaks at 2θ 21.2°, 23.1°, 27.8°, 29.8°, 32.2°, 33°, 40°, 43.1°, 43.8° and 48.1° pertaining to crystal planes (210), (002), (400), (−212), (020), (212), (022), (412), (420) and (−104) were observed. These results presented the presence of monoclinic phased $La_2Ti_2O_7$ with $P2_1/m$ space group and was also affirmed with the standard JCPDS 27–1182 (…

Conclusion

In summary, a novel reline deep eutectic solvent system was used to synthesize lanthanum titanate via solid-state technique. The synthesized material was incorporated as an adsorbent material towards potentially toxic heavy metals such as lead, arsenic and chromium. The adsorption efficiency of LTO was evaluated at different conditions including pH, contact time, concentration, multiple ion environment etc. The kinetics and isotherm of adsorption followed by LTO was also verified. LTO showed…

Credit author statement

Roselin Ranjitha Mathiarasu: Investigation, Methodology and Writing - Original Draft **Kurinjinathan Panneerselvam, P. Senthil Kumar, Raghu Subashchandrabose:** Formal analysis, Data curation, Validation. **Gayathri Rangasamy, Mary George:** Conceptualization, Validation and Supervision.…

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.…

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