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THE WASTE-TO-VALUE PARADIGM: ADVANCING CIRCULAR MATERIALS ENGINEERING THROUGH SYNTHESIS, CHARACTERIZATION AND APPLICATION

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ABSTRACT

The global depletion of coal and petroleum reserves and the amount of low-value industrial and agricultural waste necessitate an acceleration of the transition to renewable, circular, and energy dense resources. To face this difficulty, in this review paper, the results of seven separate experimental research projects, all under the "Waste-to-Value" paradigm, are collated. These studies encompass the synthesis of zeolite-X/NaP1 as an adsorbent from waste aluminum cans, achieving a Freundlich adsorption capacity for the synthetic dye Congo Red of 321.29 mg/g; TiO_2/zeolite nanocomposites using coal fly ash-derived zeolite, demonstrating 99.68% photodegradation efficiency of the synthetic dye Naphthol Blue Black under visible light; activated carbon derived from corn stalks and corn cobs for Fe(II) adsorption, achieving an adsorption efficiency of 67.65%; a continuous-flow hydroxyapatite column derived from eggshells, achieving 99.48% Cr(VI) removal; mesoporous TiO_2 nanoparticles synthesized via a green approach using Colocasia esculenta L. leaf extract at a calcination temperature of 500°C, achieving 98.64% methylene blue photodegradation; high-density biopellets produced from a mixture of walnut shells and palm kernels, with a calorific value of 4,684.4 cal/g, meeting the requirements of SNI 8951:2020; and the recovery of light and heavy fuel oils from maritime sludge through pyrolysis, producing fuels that comply with the specifications for low-sulfur marine fuels. To summarise, the studies discussed in this review establish a strong case for the conversion of industrial and agricultural wastes to high-performance materials for water remediation and renewable energy applications. The outcome is a validated risk-informed collaborative platform that is in line with the push toward the Sustainable Development Goals (SDGs), in particular SDG 6 (Clean Water and Sanitation), SDG 7 (Affordable and Clean Energy) and SDG 12 (Responsible Consumption and Production).

KEYWORD

Circular materials engineering, adsorbent, photocatalyst, biomass, renewable energy

INTRODUCTION

Global reserves of petroleum and coal are anticipated to last roughly 30 and 70 years, respectively. The industry is forced to switch to high-density renewable biomass immediately. At the same time, agricultural and industrial by-products such as walnut kernel shells, coal fly ash, corn stover and aluminum-can waste are massive, underutilised liabilities that must be transformed into valuable engineered assets. Walnut-shell production alone is reported to be approximately 248 Mt per year (Iryani et al., 2025). The problem is compounded by the complexity as well as the quantity of the contaminants. For example, synthetic textile dyes such as Congo Red and Naphthol Blue Black are resistant to natural decomposition, carcinogenic, and mutagenic (Iryani et al., 2024; Kusumawardani, Syahputri, et al., 2025). Hexavalent chromium in electroplating effluent is acutely hazardous even at low concentrations (Nur Ismawati et al., 2023).

In response to this global mandate, our research is structured around a circular engineering loop with four stages: (1) Source – Identification of high-potential precursors from negative-value materials including aluminium cans, coal fly ash, corn stovers, chicken eggshells, walnut shells and hazardous sludge oil; (2) Synthesise – Development of sol-gel, hydrothermal, alkali-fusion, precipitation, pyrolysis and green (phytochemical) synthesis routes to restructuring matter at a molecular scale; (3) Characterise – Rigorous verification via XRD, SEM, FTIR, DRS and BET for morphology, purity and performance; (4) Apply – Deployment of the resulting materials for wastewater remediation, heavy-metal capture and renewable energy generation (Figure 1).

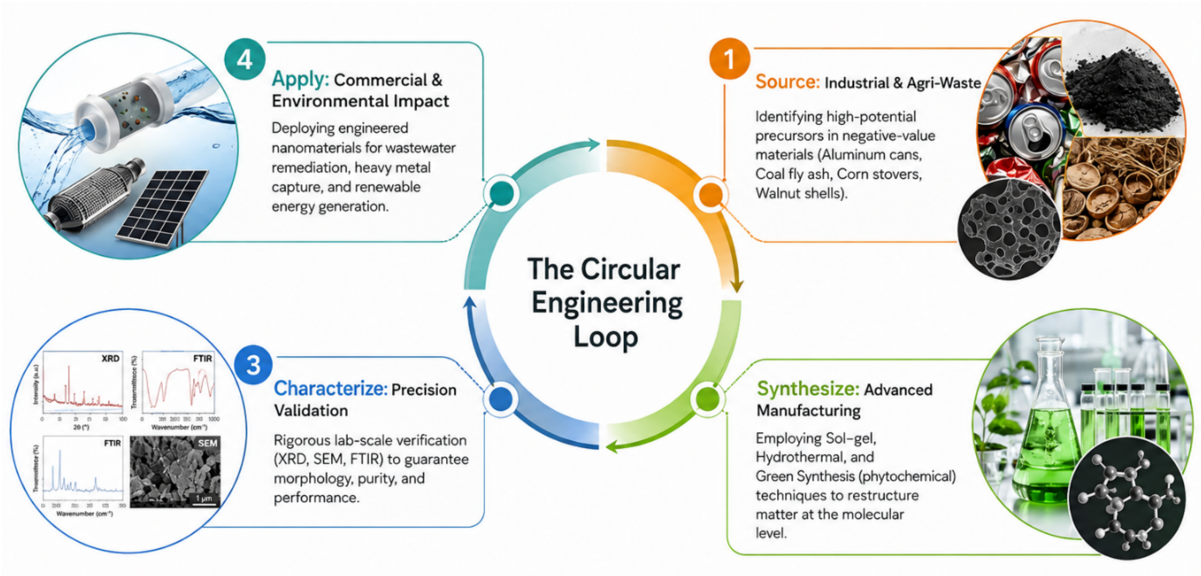

Figure 1: Our paradigm – the circular engineering loop (source, synthesize, characterize, apply).

The research architecture is based on 3 complementing pillars: Pillar 1: Advanced adsorbents and remediation including zeolites (Iryani et al., 2024), activated carbon (Iryani et al., 2026) and hydroxyapatite (Nur Ismawati et al., 2023) for the removal of synthetic dyes and heavy metals. Pillar 2, Renewable Energy and Resource Recovery: biomass bio-pellets (Iryani et al., 2025) and pyrolysis oil recovery from hazardous sludge (Kusumawardani, Sutanto, et al., 2025). Pillar 3, Green Nanotechnology and Composites, includes TiO_2/zeolite and plant-mediated TiO_2 photocatalysts. These three pillars combine to provide a complete and flexible toolkit to handle complicated industrial wastewater and energy problems with trash that would be otherwise dumped.

MATERIAL AND METHODOLOGY

The proposed approach is based on a "Waste-to-Value" cycle (Figure 2). Four major waste streams were utilized as feedstocks: household waste (eggshells and aluminum cans), agricultural waste (walnut shells, corn stalks, and corn husks), industrial waste (coal fly ash), and maritime waste (hazardous oily sludge). These feedstocks were converted through a combination of thermochemical and physicochemical processes into five main product classes: hydroxyapatite, zeolite, activated carbon, TiO_2-based nanocomposites, and recovered fuel oil/bio-pellets.

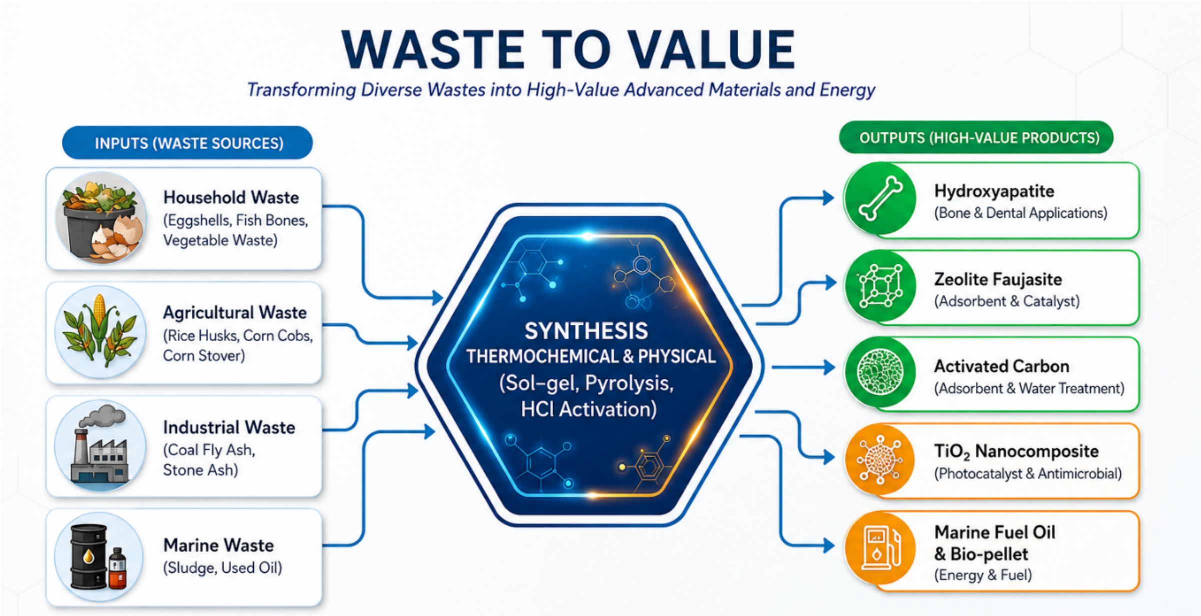

Figure 2: The Waste-to-Value synthesis flow, from waste inputs through thermochemical/physical synthesis to engineered outputs.

The synthesis of zeolite from aluminum-can waste was carried out according to the method of Iryani et al. (2024). The aluminium was obtained from clean cans through digestion with nitric acid, HCl, and H_2O_2 and then converted into sodium aluminate with 1 M NaOH, dried at 100 °C, and calcined at 650 °C for 3 hours. The sodium aluminate was then stirred with 58% waterglass and NaOH for 1 h, hydrothermally treated at 100 °C for 24 h, washed to pH 8 and dried at 100 °C to prepare the zeolite.

The preparation of TiO_2/zeolite coal fly ash nanocomposite was carried out according to the method of Kusumawardani et al. (2025). In the first step, the zeolite component was obtained by calcination of coal fly ash at 800 °C, washing with 10% HCl, fusion with NaOH (1:1.6 w/w) at 550 °C and hydrothermal treatment at 110 °C for 8 hours. Separately, TiO_2 was prepared by sol-gel technique using titanium tetra-isopropoxide. This was mixed with the synthesised zeolite (2:1 w/w) in water and aged for 14 h and then calcined at 500 °C for 2 h to make the nanocomposite. The prepared material was characterised by FTIR, DRS, XRD and SEM. The photocatalytic activity of the prepared material was checked for the degradation of Naphthol Blue Black (NBB) under 1000 W halogen lamp. Effect of H_2O_2 dose, pH, catalyst dosage, initial dye concentration and irradiation time were optimised systematically. Reaction kinetics was studied by Langmuir–Hinshelwood model.

Activated carbon from maize stalk and husk (Iryani et al., 2026). Corn stalks and husks were combined, carbonised at 400 °C for 3 hours, sieved through a 100 mesh screen, activated with 3 M HCl (1:10 ratio, 7 hours), washed to neutrality, and dried at 110 °C. Moisture content (105 °C/3 hours), ash content (550 °C/3 hours), iodine adsorption (titration), and methylene blue (UV-Vis 665 nm) are all used in quality testing, while FTIR and SEM-EDS are used for characterisation. The optimisation of Fe(II) adsorption uses 1.25 g of carbon in 100 mL of solution at varied pH (3-6), initial concentration (5-12 mg/L), and contact period (30-75 minutes), with the final analysis utilising AAS. The equilibrium data are then fitted to the Langmuir and Freundlich isotherm models to obtain the optimal adsorption constant and model.

Hydroxyapatite from chicken eggshells. CaO was prepared by washing, drying, and calcining chicken eggshells at 1000 °C for 5 h. The resulting CaO was then dissolved in HNO_3 and mixed with $(NH_4)_2HPO_4$ at a Ca:P molar ratio of 1.67 using the precipitation method (Ismawati, Iryani, & Kusumawardani, 2023). The resulting hydroxyapatite was characterized by XRD, FTIR, and SEM, and subsequently packed into a continuous-flow adsorption column (½″ diameter, 30 cm height) for the removal of Cr(VI) from simulated electroplating wastewater as per figure 3. The adsorption experiments were conducted at a fixed flow rate of 25 mL/min, with pH (3–7), contact time (30–90 min), adsorbent mass (2.0–4.0 g), and initial Cr(VI) concentration (13.86–57.64 ppm) investigated as operating variables.

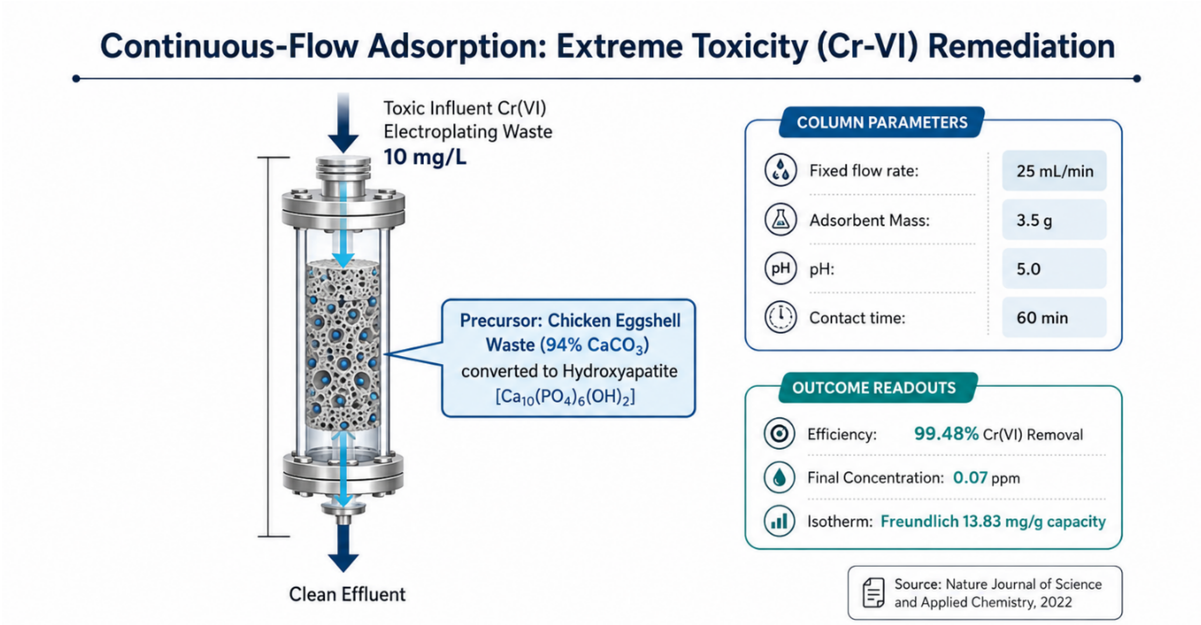

Figure 3: Continuous-flow adsorption column for extreme-toxicity Cr(VI) remediation using eggshell-derived hydroxyapatite.

Green synthesis of TiO_2NPs. The leaves of *Colocasia esculenta* L. (taro) were macerated in 96% ethanol to achieve a phenolic-rich extract (CELEs; total phenolic content 33.20 mg GAE/g), which was used as a bioreductor and capping agent for titanium tetra-isopropoxide at a TTIP:CELEs ratio of 1:3 (v/v) (Kusumawardani et al., 2026). The resultant TiO_2–CELEs nanoparticles were calcined at 300, 400, 500, and 600°C for 3 hours each, characterised by DRS, XRD, BET, SEM-EDS, and PSA, and tested for methylene blue photodegradation under light and dark conditions.

Production of bio-pellets from walnut and palm kernel shells (*Canarium indicum* L.) from Bogor: walnut kernel shells were sun-dried, crushed to 60 mesh, and mixed in the ratio of 30:70, 50:50, 70:30, and 100:0 (walnut:palm) with 20% starch binder, and pressed at 200-230°C and 100 kg/cm² into bio-pellets (Iryani, Kusumawardani & Juniansyah, 2025). The calorific value, moisture, ash, volatile matter, fixed carbon, sulphur content, and bulk density of bio-pellets were tested according to Indonesian National Standard SNI 8951:2020 for power-plant biomass fuel.

Pyrolysis of toxic sludge oil. (B3) Hazardous and toxic maritime sludge oil, such as used hydraulic lubricating oil, refinery sludge and tank-bottom residues, was pyrolysed in a 1000°C reactor at temperatures of 250, 300 and 350°C with reaction times of 5, 6, 7 and 8 hours, condensing vapour at 215, 270 and 310°C, respectively (Kusumawardani, Sutanto, & Krisanthy, 2025). Pyrolysis oils were analysed for density, kinematic viscosity, calorific value and flash point and then classified as Light Fuel Oil (LFO) or Heavy Fuel Oil (HFO) according the Indonesian Marine Fuel Oil (MFO 180/380) standard (Decree of the Director General of Oil and Gas No. 0179.K/10/DJM.S/2019).

RESULT AND DISCUSSION

Zeolite synthesised from aluminum-can waste (96.5% pure Al) achieved a synthesis yield of 92.67%, and XRD confirmed a mixed Zeolite-X/NaP1 phase (average crystal size 24.08 nm; purity 55.44% and 44.53%, respectively) and SEM showed a faujasite-type morphology (Iryani, Kusumawardani, & Destiani, 2024). The zeolite exhibited max. Freundlich adsorption capacities of 321.29 mg/g and 77.54 mg/g for Congo Red and Naphthol Blue Black, respectively. The Freundlich isotherm was better fitted than the Langmuir isotherm (R^2 = 0.9929 and 0.9636) for both dyes indicating heterogeneous, multilayer adsorption dominated by weak Van der Waals interactions.

The TiO_2/zeolite coal-fly-ash nanocomposite contained Ti–O–Si and Ti–O–Al functional groups, a band gap narrowed to 2.85 eV (3.22 eV for pure TiO_2), and a crystallite size reduced from 62–75 nm (zeolite alone) to 10.3 nm with 98.2% crystallinity after TiO2 introduction (Kusumawardani, Syahputri, Apriansyah, & Fathurrahman, 2025). The nanocomposite exhibited NBB degradation efficiency of 99.68% at optimised circumstances (1.2% v/v H_2O_2, pH 2, 0.1% w/v catalyst, 12 mg/L NBB, 75 min visible-light irradiation). The degradation process obeyed pseudo-first-order Langmuir–Hinshelwood kinetics (k = 0.0312 min^{-1}, R^2 = 0.9044; Qmax = 11.82 mg/g). The LC-MS/MS revealed the mineralisation of benzoic acid, maleic acid, and phenol as degradation intermediates to CO_2 and water.

Among the tested combinations, the activated carbon prepared from corn stalk and husk with a ratio of 30:70 (BJ30KJ70) showed the highest surface area (870 m^2/g) and decreased Fe(II) concentration by 67.65% (from 10 mg/L to 3.235 mg/L) at pH 3, initial concentration of 10 mg/L and 30 min contact time according to the Langmuir monolayer model (qm = 0.5996 mg/g) (Iryani et al., 2026). However, the single-precursor carbons BJ100 and KJ100 conformed to the Freundlich multilayer model (qm = 1.4766 and 0.1684 mg/g, respectively). This demonstrates that the combination of maize husk and stalk waste generates a more uniform and greater capacity adsorbent surface for iron removal.

The continuous flow Cr(VI) remediation trial showed that the eggshell-derived hydroxyapatite column could remove 99.48% of Cr(VI). The optimum condition was at pH 5, contact time 60 min, adsorbent mass 3.5 g, and initial concentration of 10 mg/L, and the final concentration was 0.07 mg/L (Ismawati, Iryani, & Kusumawardani, 2023). The adsorption fitted the Freundlich isotherm (R^2 = 0.9960) with a maximal capacity of 3.93 mg/g and the kinetics fitted a second order model (R^2 = 0.8163) consistent with the exchange of Ca(II) sites in hydroxyapatite for Cr(VI) through the phosphate group. Table 1 summarises the resulting advanced remediation capacity matrix, with each precursor waste linked to its designed material, target pollutant, and validated removal performance.

Table 1: The advanced remediation capability matrix – a versatile toolkit for complex industrial wastewater challenges.

Precursor Waste	Engineered Material	Target Pollutant	Active Mechanism	Reference
Aluminum cans	Zeolite-X / NaP1	Soluble & insoluble textile dyes	High-capacity adsorption	Iryani et al., 2024
Coal fly ash	TiO_2/Zeolite nanocomposite	Complex organic compounds	Visible-light photodegradation	Kusumawardani, Syahputri, et al., 2025
Corn stalks & husks	Highly porous activated carbon	Heavy metals (Fe)	Micro-pore trapping	Iryani et al., 2026
Chicken eggshell waste	Hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$)	Cr(VI) electroplating waste	Continuous-flow column adsorption	Nur Ismawati et al., 2023

In the domain of green nanotechnology, a different trade-off between surface area and crystallinity at calcination temperatures was seen for TiO_2 nanoparticles synthesised using Colocasia esculenta leaf extract (TiO_2–CELEs) (Kusumawardani, Winanti, Iryani, Nurlela, Mahendra, & Haryani, 2026). The sample calcined at 400 °C showed the highest BET surface area (106.31 m^2/g) and the smallest crystallite size (6.33 nm) and achieved 96.64% methylene blue photodegradation, whereas the sample calcined at 500 °C, though with a lower surface area (60.13

m^2/g), showed the highest degradation efficiency (98.64%) due to better crystallinity (82.8%) and more effective charge separation. Both samples showed lower band gaps than pure anatase TiO_2 (3.29 and 3.24 eV vs 3.22 eV) establishing CELEs as a viable eco-friendly biotemplate.

The 30:70 walnut to palm shell bio-pellet mixture in the renewable-energy pathway was optimised to provide the best overall balance of thermal and mechanical properties with calorific value of 4,684.4 cal/g, moisture content of 8.83%, ash content of 1.85%, volatile matter of 69.16%, fixed carbon of 19.30%, sulphur content of 0.02%, and bulk density of 1.18 g/cm3, completely fulfilling the SNI 8951:2020 standard for utility-quality power plant fuel (Iryani, Kusumawardani, & Juniansyah, 2025). Pure walnut-shell pellets (100% W) achieved a calorific value of 4,593 cal/g, while the highest overall single-formulation calorific value was 4,683 cal/g at 70% palm / 30% walnut, suggesting that palm kernel shells are a high-purity, low-ash complement to locally available walnut trash.

Pyrolysis of hazardous maritime sludge oil indicated that oil yield and categorisation were controlled by pyrolysis and condensation temperature (Kusumawardani, Sutanto, & Krisanthy, 2025). Total oil yield rose with temperature and reaction time, reaching 60.86, 65.75 and 70.69% at 250, 300 and 350 °C respectively (8 h reaction time). The oil condensed at 215 °C and 270 °C passed the specification of Light Fuel Oil (MFO 180: density 938 kg/m3, viscosity 50.51 cSt) and the oil condensed at 310 °C was similar to Heavy Fuel Oil (MFO 380: density 980 kg/m^3, viscosity 187.35 cSt). Both fuel classes met the Indonesian low-sulfur Marine Fuel Oil standard, indicating that hazardous B3 waste can be converted into marketable marine fuel.

Taking these lab-scale achievements into the commercial arena requires navigating what is commonly called the "valley of death" in clean-technology advancement. In this model of collaboration, the university would contribute basic characterisation, proof of concept, material-synthesis optimisation, and rigorous lab-scale validation in all seven studies described above, while industry partners would contribute pilot-scale testing, manufacturing integration, supply-chain distribution, and commercial deployment. This engineered alchemy portfolio directly aligns to major international grant priorities – SDG 6 (Clean Water, via zeolite, activated-carbon and hydroxyapatite adsorbents), SDG 7 (Affordable and Clean Energy, via bio-pellets and pyrolysis fuel oil) and SDG 12 (Responsible Consumption, via the upcycling of hazardous, agricultural and food waste) – collectively offering a de-risked foundation for joint funding applications.

CONCLUSION

To summarise, the Waste-to-Value paradigm demonstrates that low-value industrial and agricultural residues – aluminium cans, coal fly ash, corn stovers, chicken eggshells, walnut shells, taro leaves, and hazardous sludge oil – can be systematically re-engineered via sol-gel, hydrothermal, alkali-fusion, precipitation, pyrolysis, and green synthesis routes into high-performance zeolite adsorbents, visible-light photocatalysts, activated carbon and hydroxyapatite adsorbents, and renewable bio-pellets and recovered fuel oil. The resulting materials present strong and quantifiable performance, including dye-adsorption capacity of up to 321.29 mg/g, dye photodegradation of 99.68%, Cr(VI) removal of 99.48%, and fuel products that meet SNI 8951:2020 and national Marine Fuel Oil standards, while directly contributing to SDG 6, SDG 7, and SDG 12. This portfolio is a comprehensive and collaborative platform for joint grant applications, co-development and technology licensing between academic researchers and industry partners to produce sustainable, circular solutions to wastewater and energy concerns.

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