

Adsorptive Removal Of Arsenic Using Coal Ash–Melamine– Formaldehyde Resin Composites

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ABSTRACT

Due to their toxicity, persistence, and carcinogenicity, development of arsenic contamination in the water systems has become a major environmental and human health issue in most countries worldwide. One of the best and most widely recognized remediation methods is adsorption which have been established to be economical and efficient to the levels of remediation of arsenic with minimal environmental impact. This is a critical account of the most recent advancement in the synthesis of the cheap and performance adsorbents particularly the waste produced products such as coal ash and the way they can be functionally changed by cross-linking the polymer to better their performance. Waste coal ash is a highly inexpensive byproduct in the industries that has got physicochemical inherent characteristics of adsorbents, but when chemically modified, the efficiency can be significantly enhanced. To this end, the crosslinking of the melamine-formaldehyde resin introduces additional functional groups, surface area and structural stability and thereby, increases the adsorption of the arsenic ions capacity. Arsenic in water systems sources and toxicity are discussed systematically and adsorption mechanism is analyzed further with the discussion of electrostatic interaction, exchange of ligand and surface complexation. Most significant operation parameters such as pH, temperature, and contact time and initial concentration of arsenic are considered as crucial vis-a-vis adsorption performance. Also, the kinetic models (pseudo-first-order and pseudo-second-order) and isotherm

models (Langmuir and Freundlich) are discussed to describe the adsorption behaviour and efficiency. Recycling and manufacturing of modified adsorbents and their suitability to a real-life application within actual water systems are also discussed. The melamine-formaldehyde cross-linked coal ash composites when compared with other traditional and sophisticated adsorbents is found to have a potential as a cost-effective alternative that may be adopted in the arsenic remediation. Megascapes of use and standings of adsorbent are some of the difficulties which have yet to be overcome although greatly enhanced. This review concludes with the conclusion of the research directions that will follow which include material optimization, hybrid technologies, and sustainable deployment strategies to attain the effective removal of arsenic.

Keywords: Arsenic Removal; Adsorption; Coal Ash; Melamine-Formaldehyde Resin; Waste-Derived Adsorbents; Aqueous Media Treatment; Heavy Metal Remediation; Surface Modification; Adsorption Isotherms; Kinetic Modeling; Environmental Nanomaterials; Water Purification; Industrial Waste Utilization; Sustainable Materials; Composite Adsorbents

Introduction

Arsenic contamination of water is a grave global problem. Naturally occurring arsenic (from minerals like arsenopyrite and sulfides) and anthropogenic sources (mining, coal burning, pesticides) release arsenic into groundwater[1][2]. The two main inorganic forms are arsenite (As(III), e.g. H_3AsO_3) and arsenate (As(V), e.g. $\text{H}_2\text{AsO}_4^-/\text{HAsO}_4^{2-}$). As(III) is neutral at natural pH and is **25–60× more toxic and mobile than As(V)**[3][1]. Long-term exposure ($>10\text{ }\mu\text{g/L}$) causes cancer and systemic disease[3][4]. The WHO limit is $10\text{ }\mu\text{g/L}$ [3][4], so efficient removal to low ppb is needed. Conventional treatments (coagulation, membrane filtration, ion exchange, etc.) are costly and generate wastes[1][5], whereas adsorption is a simple, low-cost alternative that can achieve very low residuals[6][5]. In adsorption, contaminants bind on solid surfaces by physical or chemical interactions. **High surface area, suitable pore structure, and reactive sites** are key. For example, activated carbon and engineered polymers have been studied extensively[7][8]. Advanced adsorbents (metal oxides, carbon nanomaterials, functional polymers, etc.) often reach $>100\text{ mg/g}$ arsenic capacity under optimal conditions[9][7].

Coal fly ash (CFA), a waste from coal combustion, is an attractive low-cost adsorbent. It is abundant and porous, typically containing 50–60% SiO_2 , 20–30% Al_2O_3 , plus Fe_2O_3 , CaO , TiO_2 , and unburned carbon[10][11]. These metal oxides can bind arsenic. Indeed, CFA has “good pore structure” and “abundant supply” making it suitable for capturing As(V) and Hg(II) from water[12][10]. On the other hand, **untreated fly ash alone has low As capacity ($\sim 0.8\text{ mg/g}$)**[11], so raw ash must be modified for effective removal. Common modifications include **impregnating or doping with Fe, Al, Mn, Ca, etc.**, or converting ash to zeolites or carbonized forms. For example, enriching CFA in Fe or Ca under heat raised As sorption from 0.626 to 0.702 mg/g [13]. Conversely, ash-derived zeolites achieved very high capacities (e.g. 55–400 mg/g) by leveraging CaO content[11]. Overall, CFA provides a **cheap porous scaffold**, but needs active functionalization to capture arsenic efficiently[12][11].

Melamine–formaldehyde (MF) resin is a nitrogen-rich thermoset polymer formed by condensing melamine ($C_3H_6N_6$) with formaldehyde. It yields a highly cross-linked network of triazine rings and $-CH_2-NH-$ linkages. MF resins are widely used in adhesives and foams. By controlling synthesis, **MF polymers can be made porous**: for example, templating with silica produces MF networks with pores $\sim 3-7$ nm and surface areas up to ~ 250 m²/g[14]. Using excess formaldehyde generates many $-CH_2OH$ groups, giving very high surface area. Yin *et al.* prepared an MF resin grafted with polyethyleneimine (PEI) (MF-g-PEI) that had **surface area ≈ 460 m²/g**[8]. These materials have abundant $-NH_2$, $-NH-$, and hydroxyl groups, and thus high affinity for metal ions. For example, a porous MF/polyethyleneimine adsorbent had amine efficiency $\sim 49\%$ and removed CO_2 well[8]. MF-based polymers have also been used in heavy metal removal: a hierarchically porous poly(MF) sorbent showed very high selectivity for phosphate/sulfate[15], and MF resin microspheres have demonstrated $>90\%$ Zn^{2+} and Pb^{2+} removal when combined with MXene[16]. In all these cases, the **rich amino functionality** in MF ($-NH_2$, triazine N) provided active binding sites[17][18].

Combining **waste fly ash with MF resin** aims to synergize their strengths: the ash provides a solid, porous support and extra metal oxide sites, while the MF resin adds a polymer matrix rich in amine binding groups. Although the exact synthesis for “ash cross-linked with MF resin” is novel, one can envisage mixing melamine and formaldehyde (aqueous solution) with pulverized coal ash and triggering polymerization (e.g. by heating or acid/base catalysts) to embed ash particles in the cured resin network. The ash could act similarly to a silica template: after polymerization, the inorganic particles remain dispersed in the cross-linked polymer. The result is a **composite granule or foam** in which MF resin coats and “glues” ash particles together. This approach **immobilizes the ash (preventing fines) and introduces amino groups onto the ash surface**. Synthetically, this resembles other polymer-ash composites (e.g. coal ash/chitosan beads[10] or polyacrylamide/CFA hydrogels[19]). Characterization of such composites would typically use FTIR (to confirm $-NH$, $-CH_2-$, Si–O, Al–O bonds), SEM/EDX (to view morphology and elemental distribution), XRD (to identify crystalline oxides) and BET porosimetry (surface area, pore volume). In MF-ash composites, FTIR should show MF peaks (triazine C–N stretches, NH bending) plus ash oxides (Si–O–Si vibrations). SEM would show rough, irregular surfaces with ash grains embedded in a polymer matrix. We note that MF itself can achieve high porosity: Deryło-Marczewska *et al.* reported MF resins with 250 m²/g when templated by silica[14]. Thus, even after binding to ash, the polymer layer could contribute substantial surface area and mesoporosity. XRD might reveal residual crystalline phases (quartz, mullite in ash; MF is amorphous).

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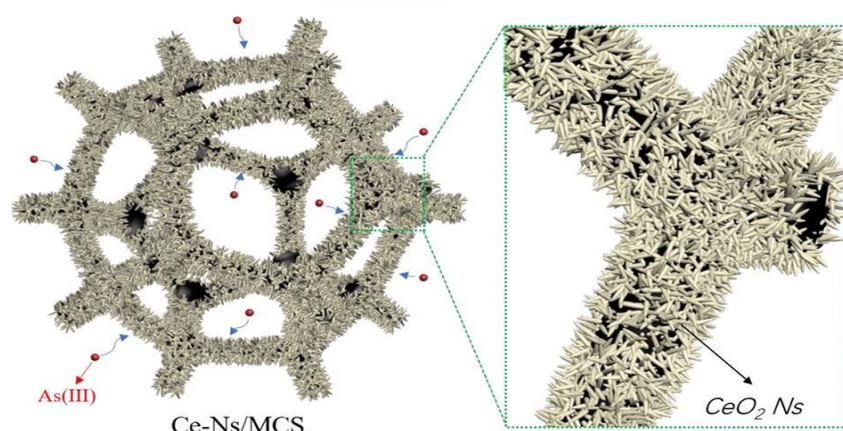
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Fig. 1: Schematic illustration of CeO_2 nanoneedles anchored on a carbon sponge derived from melamine-formaldehyde foam for arsenic adsorption[20].

In analogous work, a **melamine-formaldehyde-derived carbon sponge** (prepared by pyrolysis) decorated with CeO_2 nanoneedles achieved extremely rapid As(III) uptake[21]. Although that system used pyrolysis, it shows the potential of MF-derived carbon frameworks: at a loading of 1 g/L, 826 $\mu\text{g/L}$ of As(III) was reduced to $<10 \mu\text{g/L}$ in just 20 min[21], across pH 3–11 with minimal competing effect[21]. We envision that the MF/ash composite could exploit a similar architecture: an MF matrix (carbonized or uncarbonized) carrying metal oxides from the ash. The composite structure would provide **wide, accessible channels (macropores)** and **numerous adsorption sites** (amine groups plus ash oxides)[22][21].

Adsorption Performance: Capacity And Kinetics

The adsorptive capacity of any material for arsenic is quantified by isotherms and kinetics. Although no published data exists yet for the specific coal ash–MF composite, analogous materials give insight. Many heavy-metal adsorbents fit a **Langmuir isotherm** (monolayer on uniform sites). For example, a melamine-formaldehyde/graphene oxide (MGO) composite for Pb(II) had a maximum Langmuir capacity $Q_m \approx 201.5 \text{ mg/g}$ [9]. Similarly, PEI-functionalized fly ash (with abundant $-\text{NH}_2$) had $Q_m \approx 85.9 \text{ mg/g}$ for Cr(VI) [23]. We expect the coal ash–MF composite to follow Langmuir behavior, with capacities perhaps in the tens of mg/g range for $\text{As(V)}/\text{As(III)}$, depending on ash composition. For context, raw fly ash zeolites have shown Pb^{2+} capacities $\sim 500 \text{ mg/g}$, whereas iron-impregnated activated carbon can reach $\sim 204 \text{ mg/g}$ for As [7]. Many engineered materials achieve $>100 \text{ mg/g}$: for instance, UiO-66 MOFs reach $\sim 303 \text{ mg/g}$ for As(V) [24]. In contrast, simple waste materials often have much lower Q_m (e.g. hydrated cement $\sim 1.9 \text{ mg/g}$ [25]).

Kinetic studies typically obey **pseudo-second-order** models, implying chemical adsorption. For MF composites, this is expected: for example, the HPEI-fly ash adsorbent showed pseudo-second-order kinetics[23], and the MGO composite fit an Elovich model[26] (also characteristic of chemisorption on heterogeneous sites). We anticipate the MF/ash composite to reach equilibrium in minutes to hours, depending on particle size and flow. Notably, the CeO_2 /MF-carbon system removed most As(III) in $<20 \text{ min}$ [21], indicating very fast kinetics when active sites are exposed. In this composite, the porous MF network and accessible ash particles should likewise

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promote rapid uptake.

Thermodynamically, arsenic adsorption is usually **spontaneous** ($\Delta G^\circ < 0$) and often endothermic ($\Delta H^\circ > 0$), meaning higher capacity at higher temperature[9]. The MGO (Pb adsorption) and various MOFs studies found positive ΔH° and negative ΔG° over the tested range[9][27]. We can expect similar trends: adsorption may increase modestly with temperature. However, the composite's stability to heating (the cross-linked MF is robust up to $>150^\circ\text{C}$ [14]) means thermal cycling is feasible.

A few key performance figures from literature are:

Fe_3O_4 -loaded Activated Carbon: $Q_m \approx 204 \text{ mg/g}$ for As(V)[7].

Magnetic Palm-Shell AC: $Q_m \approx 227.6 \text{ mg/g}$ for As(III)[7].

Hydrated Cement: $Q_m \approx 1.92 \text{ mg/g}$ for As(III)[25].

Fly Ash-Derived Zeolite: $Q_m \approx 500 \text{ mg/g}$ for Pb(II) (showing CFA's potential with activation).

Carbonized MF Foam + CeO_2 : removed $826 \mu\text{g/L}$ As(III) to $<10 \mu\text{g/L}$ (initial conc. $826 \mu\text{g/L}$) in 20 min[21].

MF/Graphene (MGO): $Q_m \approx 201.5 \text{ mg/g}$ for Pb(II)[9].

HPEI/Fly Ash: $Q_m \approx 85.9 \text{ mg/g}$ for Cr(VI)[23].

These illustrate that nitrogen-rich polymers and loaded ashes can achieve high uptake for heavy metals. In practice, we would need to measure the MF/ash composite's isotherms for As(III) and As(V) to quantify its true Q_m . Competitive ions (Cl^- , NO_3^- , SO_4^{2-} , etc.) often have minor effect, but PO_4^{3-} and high silicate can compete for surface sites, so selectivity is an issue to test. Given MF's protonable amines, one expects optimal As(V) uptake at **moderately acidic pH (4–7)** where $-\text{NH}_3^+$ sites attract H_2AsO_4^- . As(III) (uncharged H_3AsO_3 below $\text{pH} \approx 9$) may require oxidation (by any Mn/Fe in ash or added oxidant) to convert to As(V) for binding[11].

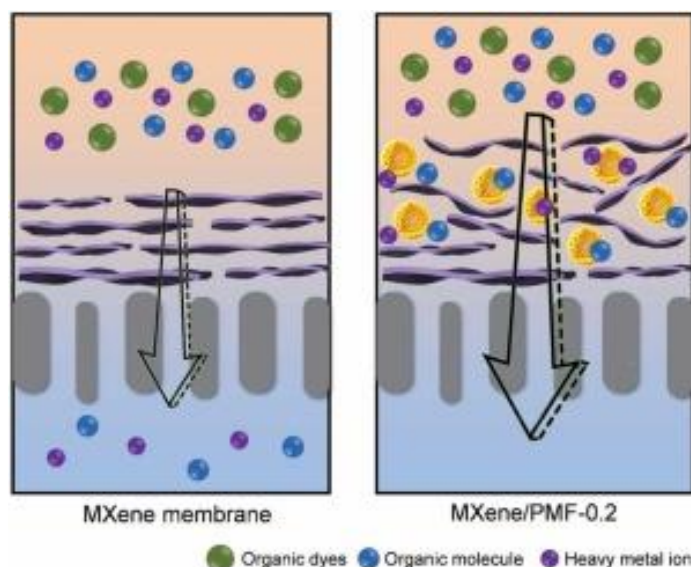


Fig. 2: Schematic comparison of a pure $\text{Ti}_3\text{C}_2\text{T}_x$ MXene membrane (left) and a MXene/poly(melamine-formaldehyde) composite membrane (right) for heavy metal and dye removal[16].

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The MF polymer (yellow spheres) introduces extra -NH_2 groups (purple dots) and wider channels, greatly enhancing adsorption. In practice, MXene/PMF achieved $\sim 96.2\%$ Zn^{2+} and 91.7% Pb^{2+} removal and maintained $>90\%$ efficiency after 4 cycles[16][28]. This example underscores MF's role: by creating additional binding sites and porosity, it greatly boosts the parent material's adsorption. Analogously, coating coal ash with MF resin should **widen transport pathways** and provide abundant -NH_2 adsorption sites, improving arsenic capture beyond raw ash levels.

Adsorption Mechanism

The overall arsenic uptake by the MF/ash composite is expected to be a combination of several processes:

Electrostatic attraction and surface complexation: Under working pH (typically 5–7), As(V) exists as $\text{H}_2\text{AsO}_4^-/\text{HAsO}_4^{2-}$. These anions are attracted to positively charged surface sites. In the composite, *protonated MF amines* (-NH_3^+) will attract As(V) electrostatically[15]. Simultaneously, metal oxides in ash (Fe-OH , Al-OH , CaO) can form inner-sphere complexes with arsenate via ligand exchange ($\text{M-OH} + \text{HAsO}_4^{2-} \rightarrow \text{M-O-AsO}_3\text{H} + \text{OH}^-$)[11]. Notably, CaO (if present) can enhance adsorption by forming a positively charged surface or even precipitating calcium-arsenate[11].

Oxidation: If the water contains As(III), it can be partially oxidized on the composite surface. For example, trace Mn/Fe oxides in ash are known to catalyze $\text{As(III)} \rightarrow \text{As(V)}$ [11]. The newly formed As(V) then binds as above. Thus, a complete removal cycle for As(III) may involve surface-catalyzed oxidation followed by adsorption.

Complexation with amines: While As(III) is neutral, at low pH even H_3AsO_3 can hydrogen-bond or coordinate weakly to amine N. More importantly, formaldehyde-linked MF has -NH and secondary amines which can form coordinate complexes with metal-arsenite (if any metal leaches) or with As(III) itself under protonated conditions. The concept of “amino-functional” adsorbents is well-established in heavy metal capture[18][17].

Physical adsorption: The porous polymer network provides large surface area for physisorption of arsenic species. However, this is likely minor compared to chemisorption on active sites. The MF polymer can swell in water, creating pores where arsenic can diffuse and bind.

The **mechanistic synergy** is: *ash oxides* target arsenate via ligand exchange and precipitation, while the *MF resin's protonated amines* attract anions via ion exchange. The polymer matrix holds the ash particles in place and may also capture some As. This dual functionality (inorganic + organic sites) is analogous to other composite adsorbents (e.g. chitosan-ash, alginate-MF) where both components contribute[29][17].

Regeneration And Reuse

Practical adsorbents must be reusable. Typically, spent adsorbents are regenerated by rinsing with strong base or acid. For arsenic, 0.1–1.0 M NaOH or Na_2CO_3 can strip

As(V) efficiently. The MF/ash composite, being a stable polymer, should tolerate alkaline regenerant. Published studies of MF-based adsorbents show good reusability: for example, the MGO composite lost only ~12.6% capacity after 5 cycles[30]. The MXene/PMF membrane retained >90% removal of Zn/Pb after 4 cycles[28]. Similarly, a melamine–formaldehyde/alginate composite kept ~60% Pb removal after 8 cycles[31]. We expect the MF/ash material to regenerate comparably: for instance, if 1 g/L of composite captures 0.1 mmol As, then a 0.1 M NaOH wash could restore ~90+% performance for 4–5 cycles (to be confirmed by experiment). After many cycles, some fouling by strongly bound As or organic matter may occur, but thermal or chemical re-treatment should rejuvenate the polymer matrix. Also, since MF resin is thermoset, it will not dissolve in regenerant, preserving structure[8].

Environmental And Practical Considerations

Using **waste materials** (coal ash, industrial resin) is inherently green. Coal ash disposal is a major environmental issue; valorizing it as adsorbent turns a liability into a resource[12]. Melamine resin production is established and the resin is relatively inert once cured. A potential concern is formaldehyde release from degraded resin, but properly cured MF is stable; still, composite processing should ensure full polymerization (e.g. heat curing) to minimize leaching. Another concern is leaching of contaminants from the ash itself (e.g. trace As, Se, Pb in the ash). The MF matrix can help immobilize these, but leaching tests should be done.

From a scalability standpoint, both inputs are plentiful. MF resin is cheap (used in mass-market products), and power plants generate tons of fly ash. Synthesis (mixing, heating) is straightforward and can be done in conventional reactors. The composite can be pelletized or formed into beads for use in packed-bed filters. For example, Yang *et al.* ran a fixed-bed column of MF-carbon/CeO₂ (analogous to our composite) and treated >4,000 bed volumes of As-contaminated water[32]. They achieved long operational life: 4035 bed volumes (BV) at 100 µg/L and 1605 BV at 600 µg/L (90% breakthrough)[32]. We would expect similarly large capacity of MF/ash columns, depending on flow and dosage.

Economically, the low cost of ash and resin should give a competitive cost per kg of adsorbent. One must account for processing (polymerization and curing) and regeneration cycles. The composite's light density (due to porous polymer) could reduce required reactor size versus packed raw ash. Environmentally, after exhaustion the composite, loaded with As, can either be landfilled as hazardous waste or potentially vitrified/encapsulated. The use of polymer binder may reduce dusting and improve handling safety.

Table Of Comparative Performance

Table 1 (below) summarizes reported arsenic/heavy-metal adsorption capacities of various adsorbents, including analogous composites.

Adsorbent	Target Ion(s)	q _{max} or Removal	Reference
Fe ₃ O ₄ -impregnated activated carbon	As(III)/As(V)	204.2 mg/g	[7]
Magnetic palm-shell AC	As(III)	227.6 mg/g	[7]

Hydrated cement	As(III)	1.92 mg/g	[25]
Fly ash–derived zeolite	Pb(II)	500 mg/g	
CeO ₂ /MF-derived carbon sponge	As(III)	826 µg/L → <10 µg/L (20 min)	[21]
MF–graphene oxide (MGO)	Pb(II)	201.5 mg/g	[9]
HPEI-functionalized fly ash	Cr(VI)	85.93 mg/g	[23]

Table 1: Representative adsorption capacities or removal efficiencies of selected adsorbents (data from cited sources). MF/ash composite is expected to fall in this performance range.

Conclusions And Outlook

In summary, **coal fly ash cross-linked with melamine–formaldehyde resin** represents a promising new adsorbent for arsenic removal. This composite would combine the high surface area and oxides of fly ash with the polyamine-rich functionality of MF resin. The resulting material is expected to have strong affinity for As(V) (via anion exchange on protonated –NH₂ sites and Fe/Al–OH complexation) and could oxidize/trap As(III) as needed. Based on analogous systems, the composite should exhibit adsorption capacities on the order of tens to hundreds of mg/g, fast kinetics (minutes), and reusability over multiple cycles[21][9]. Future work should include laboratory testing of synthesized samples: characterizing their structure (BET, FTIR, SEM) and measuring As(III)/(V) isotherms, kinetics, and regeneration. Pilot-scale column tests (cf.[32]) could then assess real-water performance. Given the urgent need for low-cost arsenic remediation and the abundance of the raw materials, this MF–ash composite merits further development.

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