



Use of marble and iron waste additives for enhancing arsenic and *E. coli* contaminant removal capacity and strength of porous clay ceramic materials for point of use drinking water treatment

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ABSTRACT

This paper elaborates manufacture and performance analysis of new clay ceramic (CC) water filtration materials. The CC is manufactured from clay and sawdust mix. Waste marble powder and machined iron fines are used as additives to the mix for manufacturing the new modified materials. An equal volume of clay and sawdust were used to manufacture the control CC. Another ceramic, marble clay ceramic (MCC), was manufactured with distinct volume fractions of clay, sawdust, and marble (40:40:10). Third ceramic, ferrous clay ceramic (FCC), was manufactured from an equal volume of clay and sawdust and five percent by volume of iron fines. FCC showcased better arsenic (As (V)) contaminant removal from water at acidic pH while MCC showcased best As (V) removal at around pH of 8. Average flexural strength of MCC was comparatively better than FCC and CC. The modified materials showcased similar percolation rates at par with control CC. MCC showcased comparatively better *E. coli* removal capabilities than FCC and CC. Only limited volumetric addition of marble powder and iron fines were found to positively affect compressive strength. The results demonstrate new low-cost ways of modifying strength and specific water treatment characteristics of CC using waste materials from local marble-processing and iron-machining industries.

Keywords: Ceramic; Marble; Iron; *E. coli*; Arsenic; Strength; Waste

1. Introduction

Clean and safe water is a basic human need. However, lack of access to safe, reliable water resources remains a critical problem for millions of people worldwide, especially for the rural residents in developing nations [1]. Out of various heavy metals, arsenic contamination of drinking water sources is a serious issue of concern in India. Millions of people are exposed to unsafe levels of chemical contaminants

in their drinking water. The long-term exposure to chemical contaminants can have adverse health issues [2–4]. The map of India in Fig. 1 shows an approximate distribution of water contamination (element) [4]. The diarrhoeal disease caused by pathogenic bacterial infection is considered the other most predominant waterborne disease which spreads health risk associated with drinking water [5,6]. There are currently thousands of water filter setups in the market with the ability to purify water contaminated in many different ways. However, most of these filtering media are too expensive,

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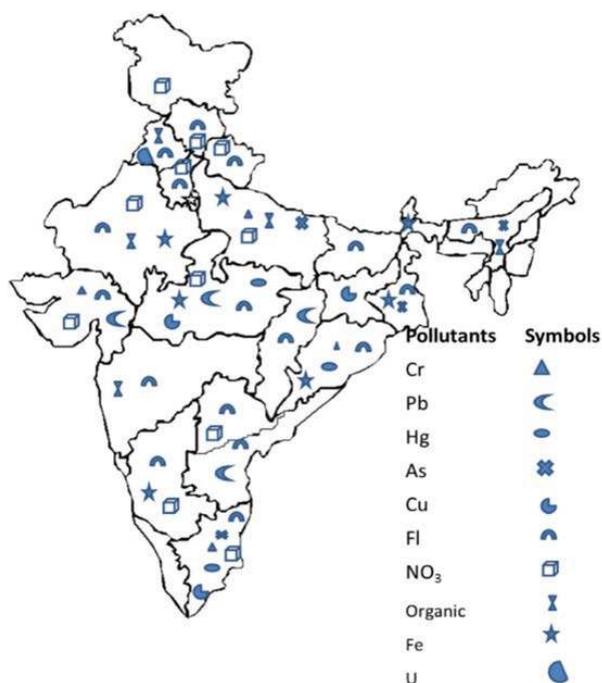


Fig. 1. Distribution of pollutants in water across different parts of India [4].

energy intensive, and do not meet the specific needs of many developing and underdeveloped nations [7].

Thus, there is a great need for sustainable solutions capable of providing clean drinking water in economically depressed regions.

Several low-cost materials have been employed by the researchers to remove arsenic and bacterial contamination from drinking water. Materials like clay, clay minerals, and sawdust have been used as adsorption surfaces to bind arsenic by chemical or physical forces [8–12]. The students of Massachusetts Institute of Technology prepared an experimental column setup for their Nepal project for arsenic treatment. The experiment involved treating raw water with Fe-oxide-coated sand. The principles of coagulation and filtration were used to effectively remove arsenic from water [13,14]. In 2001, sand and chemical powder (calcium hypochlorite) mixture were used in a filter column to treat arsenic-contaminated water of Bangladesh. During filtration, hypochlorite worked as an oxidant and sand behaved as an adsorbent to remove residual chlorine resulting from the use of hypochlorite and also aids to remove microbes in the water [15]. Fuhrman et al. established the Bauxsol technology which utilized calcium brine saturated red mud to adsorb arsenic [16]. Technical University of Berlin (Germany) tested adsorption of arsenic on granular ferric hydroxide (GFH) in fixed bed reactor. The GFH adsorbent was found to reduce levels of both As (III) and As (V) to less than 5 µg/L in drinking water [17].

Literature has suggested few remediation techniques like silver-impregnated ceramic filter, sediment filter with iron oxide (3-Kolshi filters and bio sand filter), and activated carbon filter which closely match the sustainable drinking water solutions [18]. Silver-impregnated ceramic filter is point of use filter which uses colloidal silver as a disinfectant to remove bacteria from the water. With the addition of

silver, ceramic filters are 99.99% effective at removing bacteria. However, the ingestion of too much silver can cause severe health risk through silver poisoning [6,19]. Sediment filters filter large amounts of water by taking advantage of multiple layers of sediment to naturally filter contaminants from water. The addition of nails or iron compounds or aluminum compounds to sediment filters makes them effective in removing hard metals like arsenic along with bacterial removal. The challenge with these filters is the formation of large pores which lower the degree of filtration [20,21]. The activated carbon filter eliminates chlorine and other organic compounds [18]. Plappally has formulated distinct clay-sawdust compositions to prepare simple and potable frustum-shaped ceramic water filters to optimize microbial removal efficiency [22]. In India, very recently, clay-ceramic (CC)-based G-filters were introduced for rural populace of Western Rajasthan, Bihar, and regions of southern India [23]. These clay-fired ceramic filters are investigated to treat heavy metals effectively. These filters are susceptible to breakage during operation and transportation [24]. Therefore, there is need to modify the ceramic filter composition that is capable of removing heavy metals from contaminated water at household levels and improving their mechanical strength.

In the earlier paragraphs, it can be observed that materials which have calcium and iron ingredients (chemical powders, processed granular iron) have been used either alone or in combination with clay/sand to treat arsenic- and bacteria-contaminated water. Thar Desert surroundings in Rajasthan are abundant in clay [25]. This Indian state generates tons of cellulosic, marble, and ferrous waste from wooden handicraft sector, mining, and metal industries [26]. Scientists have extensively studied use of marble waste powder in the context of removing various chemical contaminants (like fluoride, cobalt, etc.) and its application as a partial replacement for cement [27–29]. Similarly, ferrous-based materials have been utilized as supporting materials in removing many heavy metals and for improving the strength of construction materials [30–32]. The use of such industrial wastes as low-cost additive materials in the porous CC filters remains to be explored.

Therefore, the article investigated whether these low-cost industrial wastes (marble and ferrous scrap) could be used as an additive material in CC filter (G-filters) manufacturing. The effect of additive materials on the structural strength of modified ceramics was also examined.

First, the optimum quantity of industrial additives (marble and ferrous scrap) most suitable for sustaining almost same filtration rates was found by trial and error. Then construction of a simple square shaped ceramic briquettes by mixing exact proportions of clay and finely ground burnt out material (sawdust) and adding additives separately to the mixture was performed. The flow and strength characteristics were studied. Finally, contaminant removal characteristics from contaminated water using these novel ceramics were studied.

2. Materials and methods

2.1. Materials and chemical reagents

The raw materials used to prepare the ceramics were clay, carpentry sawdust, ferrous mill waste, and marble slurry. The clay obtained from Raital, Mokalsar, near Jodhpur, Rajasthan, India, was placed on the cemented floor

where it was crushed into very tiny particles using iron mallet; the clay was then sieved to a very tiny particle size less than 0.5 mm for investigation. The sieved clay was washed several times using distilled water and oven dried at 105°C for 12 h. Sawdust was used as organic pore former. It was freely available and collected from a local wooden handicraft shop (Timber mart Jalori Gate) in Jodhpur, Rajasthan, and sieved through a 1-mm sieve (ASTM 18 mesh size). The ferrous powder was obtained from Vishwakarma Fabricator, Basni Industrial area, Jodhpur. The ferrous mill powder was sieved through 0.074 mm sieve (ASTM 200 mesh size). Fine marble slurry as an additive was taken free of charge from marble-processing industry located near Bherju Ji Ka Than, Jodhpur, Rajasthan, India. The slurry was washed multiple times to remove unwanted dust or solid particles. The slurry was oven dried for 2 h at 125°C, then cooled at room temperature, packed in stopper bottle, and stored in desiccators for future use.

As (V) stock solution (100 mg/L) was prepared by dissolving the appropriate amount of $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$. NaOH (A6756, Merck, Darmstadt, Germany) and HCl were used for solution pH adjustment.

2.2. Manufacturing of clay-additive-based ceramic adsorbents

The ferrous (F) and marble (M) clay ceramic (FCC and MCC) adsorbents were separately manufactured. FCC was prepared by mixing clay soil (C), sawdust (C), and ferrous mill waste powder (F) in the ratio of 45C:45S:10F on a volume fraction basis. MCC was prepared from clay soil (C), sawdust (S), and marble slurry (M) in the ratio of 45C:45S:10M. The CC developed using an equal volumetric fraction of clay and sawdust (50C:50S) were used as control for comparison purpose.

The mixes were manually and independently blunged in deionized water to form a uniform wet paste. The wet paste was compacted into two different sizes: one in the form of small square pellets of $15 \text{ mm}^2 \times 15 \text{ mm}^2$ for the batch adsorption test. The second was $100 \text{ mm}^3 \times 100 \text{ mm}^3 \times 15 \text{ mm}^3$ dimensions for performing the mechanical strength test (as shown in Fig. 2). Oiling the mould is done before filling the wet clay-ferrous paste into the mould. The mixture is filled into the mould and pressed into a square form using a 50-t compaction machine (Model HBPO10, KEN-985-5000K, UK). The samples were kept for drying at ambient temperature conditions for a period of 1 week, and dried samples were then sintered at 850°C in an electric furnace (TEXCARE™ Muffle Furnace 220V, $250 \text{ mm}^3 \times 340 \text{ mm}^3 \times 180 \text{ mm}^3$ ($w \times d \times h$)) for 2 h to carbonize and produce pores. The square samples were cut using a ceramic cutter (Bosch GDC 120) to assess the compressive and flexural strength of samples.

2.3. Batch mode adsorption study

Batch mode experiments were carried out at room temperature ($25^\circ\text{C} \pm 1^\circ\text{C}$) in a 100-mL Teflon bottle. They were shaken at 80 rpm for 12 h. For contact time studies, 1 mg/L of arsenic (V) solution was kept in contact with 5 g of FCC and MCC adsorbents for the desired period of 5, 10, 15, 30, 45, 60, 75, and 90 min. The effect of pH from 3 to 11 was studied, and the desired pH was adjusted by adding

HCl or NaOH. The experiments were performed with an aqueous solution of the following As (V) concentrations: 50, 100, 500, and 2,000 $\mu\text{g/L}$. Freundlich and Langmuir's model isotherms were studied to evaluate the adsorption capacity and describe the arsenic adsorption onto solid ceramics. The filtrate concentration was analyzed within 24 h. All the experiments were carried out in triplicate to ensure reproducibility. The same parameters were applied for CC testings.

2.4. Microbial removal study

To check the microbial removal efficiency of ceramic wares, microbial removal test was carried out in controlled environments. Cultures of the nonpathogenic *Escherichia coli* DH10B™ strain were grown in Miller's Lysogeny broth at 37°C for 24 h with aeration by stirring at hot plate. Microbial removal tests were carried out until 500 mL of solution passed through the FCC, MCC, and CC filters prepared on a laboratory scale. The effluent was stored in 125 mL increments three times during each test. Two blanks were made for each set of controls. The *E. coli* in water was taken as per the procedure mentioned in Standard Methods for the Examination of Water and Wastewater [33].

2.5. Material characterization

X-ray fluorescence technique was used to decipher the chemical compounds present within the Fe-based stabilizer and FCC. The Fe waste, marble slurry, FCC and MCC were separately powdered using agate mortar and pestle. Powdered samples (2 mg) were analyzed in a Bruker S4 Pioneer facility (X-ray fluorescence instrument at JNU New Delhi) to determine their chemical compositions.

The mineralogical investigation of industrial wastes (ferrous mill powder and marble slurry) and the ceramic sample was carried out in a Bruker D8 ADVANCE diffractometer (at I.I.T. Jodhpur) using filtered copper $\text{K}\alpha_1$ radiations [$\text{Cu-K}\alpha$ radiation, $\lambda = 1.5418 \text{ nm}$] with a step size of 0.02 and scan speed of 0.5.

The microstructural evolution of FCC, MCC, and CC pellets before and after arsenic treatment was assessed using scanning electron microscopy (SEM) (FEI, Quanta 200, MNIT Jaipur, Jaipur, India). Gold-coated ceramics were placed in the SEM chamber for imaging analysis.

As (V) concentration prior and post adsorption was measured by atomic absorption spectroscopy (AA500, MRC, MNIT Jaipur, Jaipur, India). The pH studies were performed using a pocket-sized pH meter (Oakton pHTestr 30 Waterproof Pocket Tester).

Fourier transform infrared spectroscopy (FTIR) (Bruker™ Vertex, 70V, Germany) was used to analyze the chemical profile of ceramics before and after arsenic treatment.

2.6. Mechanical characterization

The flexural strength test was carried out on a laboratory-based universal testing machine (Model EZ-50, Lloyd Instruments, Germany). The ceramic specimens of $75 \text{ mm} \times 15 \text{ mm} \times 15 \text{ mm}$ size were processed with the three-point bend test setup at a loading rate of 0.1 N/s [22]. The results of flexural strengths were recorded using Nexygen

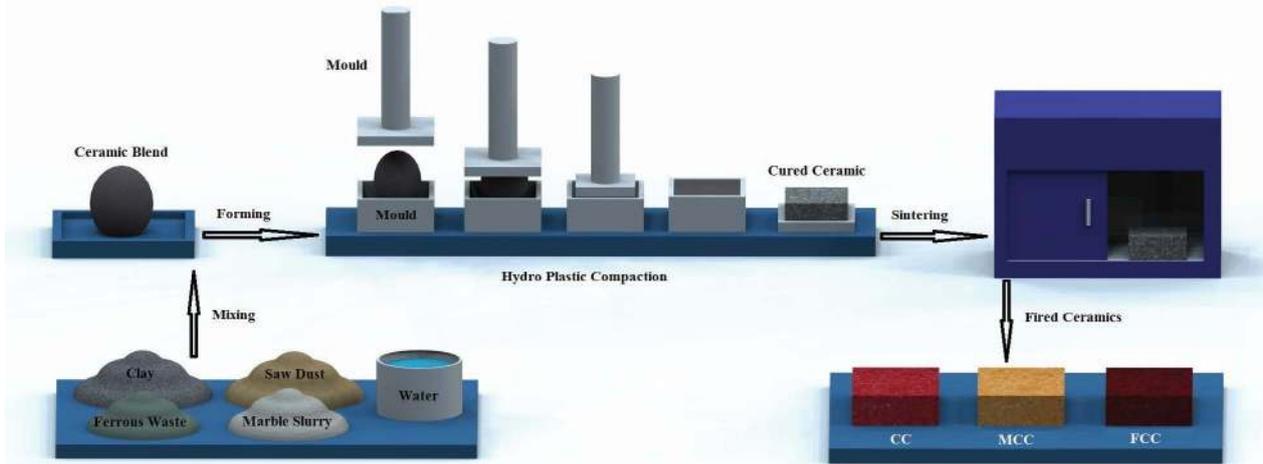


Fig. 2. Manufacturing of ceramics.

material testing software (Nexygen plus 01/3366, Lloyd Instruments, Germany).

For the compressive strength test, ceramic specimens of 35 mm × 15 mm × 15 mm cross-section were cut (Bosch GDC 120). A loading rate of 0.1 N/s was applied along the 35-mm axis until the sample was completely crushed [22]. Compressive strength data were recorded digitally using Nexygen plus material testing software.

3. Results and discussion

3.1. Properties of industrial waste and ceramic adsorbent

Table 1 shows the chemical composition of the ferrous waste and marble slurry. Table 1 depicts the high percentage of iron oxide and silica in case of ferrous waste, and the waste marble powder contains a major amount of calcium oxide followed by the considerable amount of silicon oxide.

FCC (Table 2) showcases the dominance of silica and oxides of iron (constituting approximately >80% weight percentage). The MCC contains a large fraction of silica as well as the oxides of aluminum, iron, calcium, magnesium, and potassium.

Table 2
Chemical composition (wt %) of porous ceramic adsorbent

Weight Percentage	FCC	MCC
Fe ₂ O ₃	34.642	5.134
SiO ₂	48.574	65.435
Al ₂ O ₃	9.959	16.682
CaO	2.386	7.32
MnO	0.895	–
MgO	1.681	2.339
Na ₂ O	–	0.964
K ₂ O	1.502	2.126
TiO ₂	0.361	–

Table 1
Chemical composition (wt %) of additives

Weight percentage	Ferrous waste	Marble slurry
Fe ₂ O ₃	55.545	0.844
SiO ₂	9.552	10.566
Al ₂ O ₃	1.874	1.574
CaO	0.482	37.12
MnO	0.444	–
MgO	–	18.9
Na ₂ O	–	0.192
K ₂ O	–	0.524
LOI	32.103	30.28

Fig. 3 shows the X-ray diffraction (XRD) pattern of the ferrous waste and porous ceramic adsorbent. The mill waste contained hematite as the dominant phase along with small amount of quartz. The sintered ceramic possessed main crystalline phases of quartz and hematite which were contributed by clay and iron oxide, respectively.

The XRD patterns illustrating mineral phase contents of the waste marble powder and sintered ceramics are shown in Fig. 4. The main phases in the waste marble powder are calcite (CaCO₃), quartz (SiO₂), and also minor dolomite (CaMg(CO₃)₂). The sintered ceramics include quartz, potassium feldspar, and calcite phases.

3.2. Effect of contact time

The effect of contact time of contaminated water with the FCC is premeditated during batch adsorption studies. The effect of residence time on As (V) removal was shown in Fig. 5. The curve showcases a high As removal gradient during the initial 20 min of contact and then peters slowly. An asymptote or saturation is reached with an hour. This behavior suggests the reduction in the amount of vacant surface sites with time. Based on these results, 90 min was taken

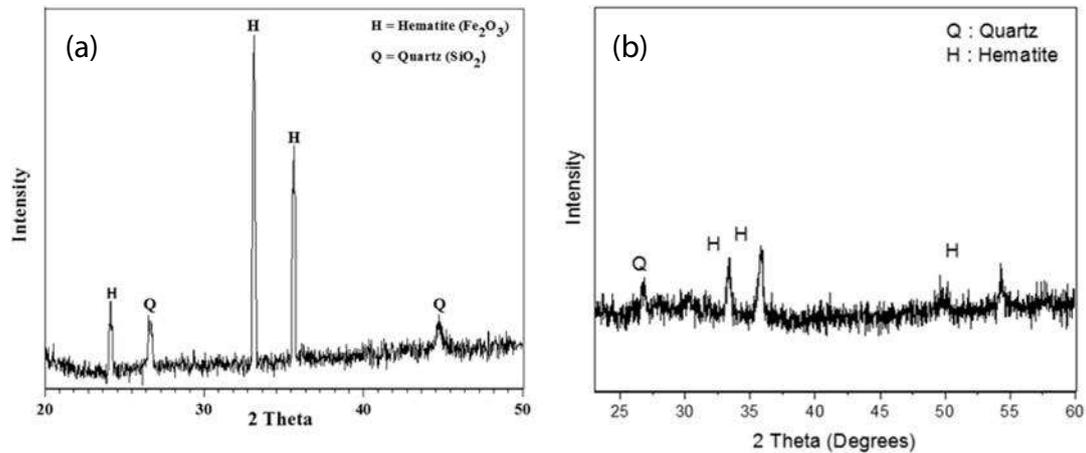


Fig. 3. XRD pattern of ferrous mill waste and ferrous-mixed porous ceramic adsorbent.

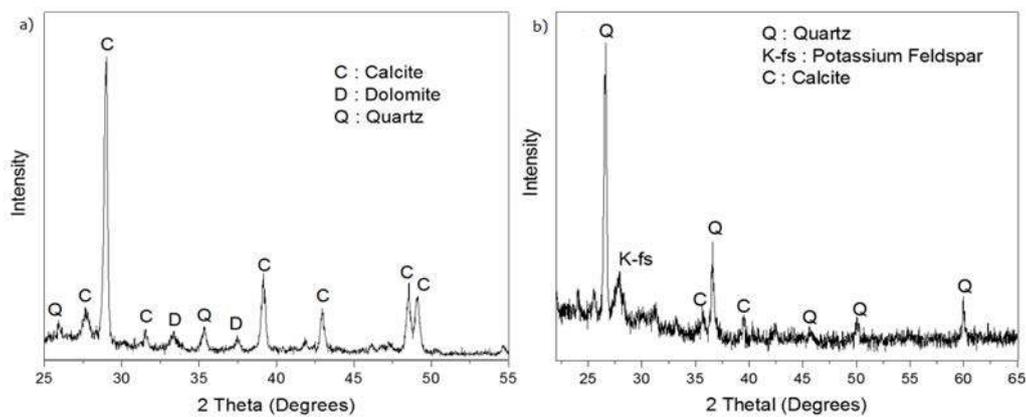


Fig. 4. XRD patterns of (a) waste marble powder and (b) sintered MCC.

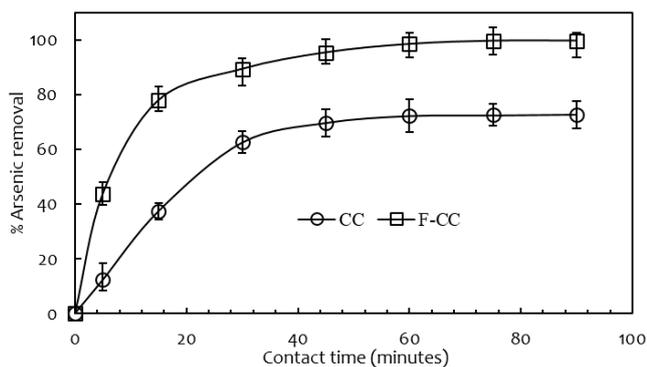


Fig. 5. Effect of contact time on the sorption capacity of As (V) ions onto FCC adsorbent (FCC pH = 6.5°C ± 1°C).

as the time for adsorption experiments to reach equilibrium. With the increase in contact time, the existence of repulsive forces between solute molecules in the aqueous solution and solid phase that causes difficulty in filling the vacant surface sites is characterized by layer formation which secludes other active surface sites as well [34,35]. This also means that FCCs can be used to achieve large removals within a short time interval. The addition of iron into the clay mixture has enhanced the arsenic removal efficiency.

The time required for saturation of MCC samples with arsenic is shown in Fig. 6. The arsenic-contaminated water of 1 mg/L is in contact with 5 gm MCC adsorbent for a distinct amount of time. Fig. 6 illustrates high As removal gradient in the first 30 min. Adsorption curve was observed to follow a birth process model with time [36]. Removal rates were the largest at 60 min beyond which MCC got saturated. The short saturation time implies faster removal efficiencies.

Initial concentration gradient on the MCC suggests rapid adsorption by MCC [37]. The rate of removal starts to ebb due to a consequent decrease in reaction sites of the MCC surface with time.

Sorption studies on clay minerals, soils, and other solid adsorbents used for arsenate removal have displayed similar faster kinetics in the initial stage [38,39]. The addition of marble slurry into the clay matrix has increased the calcite content, due to which Ca^{2+} cations are likely to enhance As adsorption by forming positively charged surface [40,41]. The formation of Ca-As precipitates has been reported which promote As immobilization in soils [42,43].

3.3. Effect on surface morphology

Fig. 7 shows the FCC surface characteristics prior to and after arsenic adsorption for a period of 90 min. Addition of

ferrous waste has resulted in dense morphology of the FCC. FCC in Fig. 6b had numerous pores prior to contact with arsenic-contaminated water. After As (V) adsorption, the surface in Fig. 6d showcases smoother surface with layers (similar to solidified lava flows) covering the pores.

The microscopic study of the MCC porous ceramic prior to and after As treatment is shown in Fig. 8. The micrographs revealed that MCC adsorbent had uneven surfaces with a large number of pore spaces. These surfaces provide sites for adsorption of arsenic either physically or chemically.

The MIP data displayed an average porosity of 42.24%, 41.19%, and 46.52% for FCC, MCC, and CC adsorbent, respectively. The mean pore diameter ranged between 1 and 5 μm .

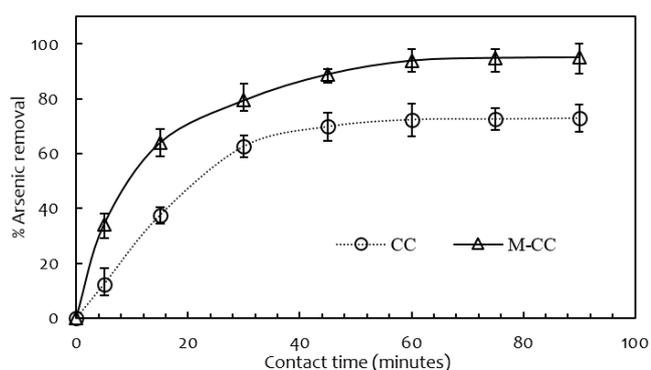


Fig. 6. Effect of contact time on the sorption capacity of As (V) ions onto MCC adsorbent (MCC pH = $8^{\circ}\text{C} \pm 1^{\circ}\text{C}$).

The ceramics modified using ferrous and marble industrial wastes produced similar flow rates (~ 960 mL/h) as obtained from CC. The details of the flow rate experimental procedure can be found from Gupta et al. 2016 [44].

3.4. Fourier transform infrared spectroscopy

Chemical profile of the modified ceramics (FCC and MCC) before and after arsenic treatment is shown in Fig. 9.

The FTIR spectra of the FCC and MCC adsorbents prior and post As (V) adsorption after a contact period of 90 min are shown in Figs. 9a and b, respectively. The bands at $1,030$ and 795.25 cm^{-1} in case of FCC and $1,052.8$ and 797.62 cm^{-1} in case of MCC indicated stretching vibration of the Si–O–Si group and Si–O deformation [45–47]. The peak observed at 676.69 cm^{-1} denoted the spectrum for iron oxide (Fe_2O_3) [48]. The peak bands at 540.17 and 448.33 cm^{-1} in case of FCC and 540.38 and 458 cm^{-1} in case of MCC resembled those of Si–O–Al and Fe–O groups, respectively [49,50]. After As (V) adsorption, the IR spectra displayed an increased percentage transmittance with reduction of stretching of peak bands of corresponding bands. Presence of new peak bands was not observed. This could be due to As (V) adsorption on the surface of the FCC and MCC adsorbents.

3.5. Effect of pH

From Fig. 10, it is observed that As (V) removal using FCC favored the acidic aqueous environments. FCC enumerated

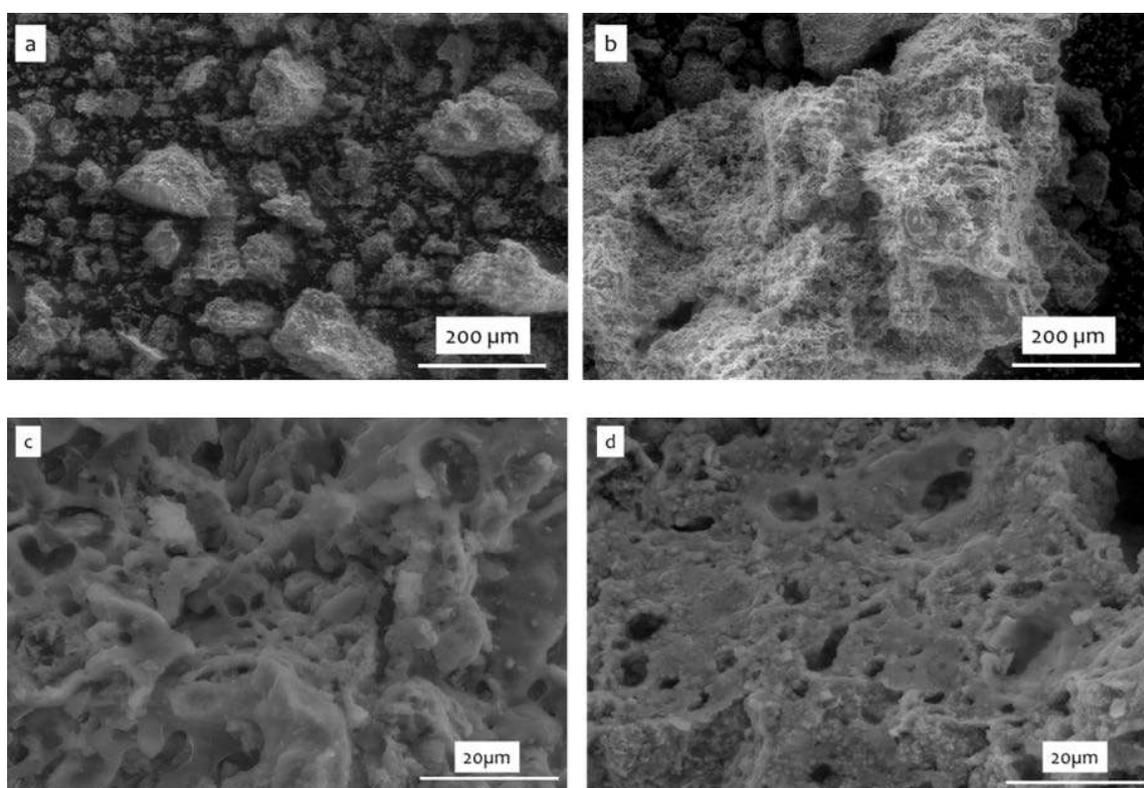


Fig. 7. Comparison between surface morphology of ceramics before arsenic treatment (a) CC and (b) FCC and after treatment (c) CC and (d) FCC at $500\times$.

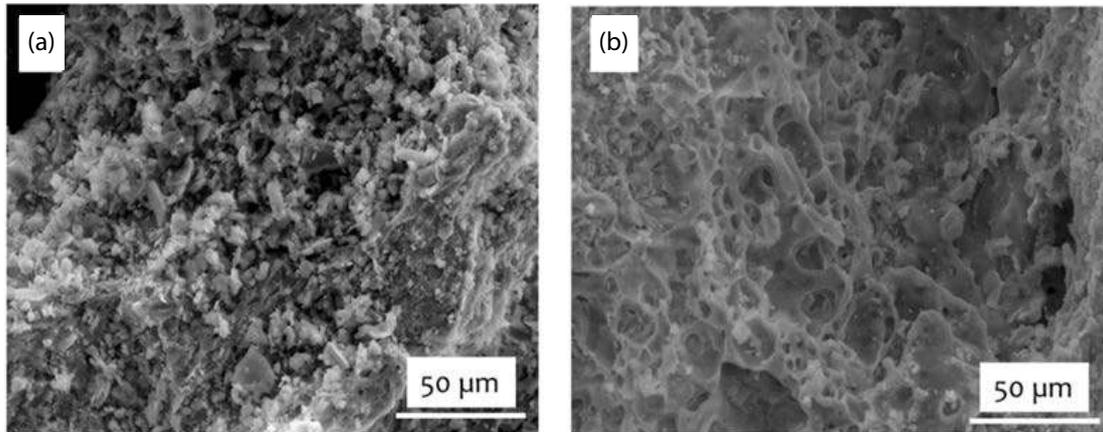


Fig. 8. Surface morphology of MCC (a) before and (b) after a contact time of 90 min (20,000×).

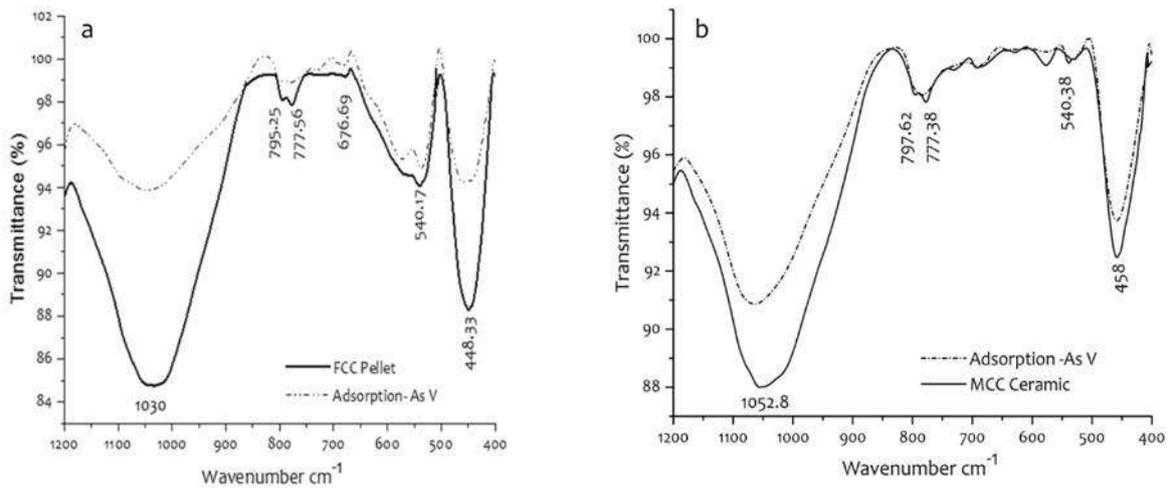


Fig. 9. FTIR spectrum of (a) ferrous ceramic before and after As (V) adsorption and (b) marble ceramic adsorbent.

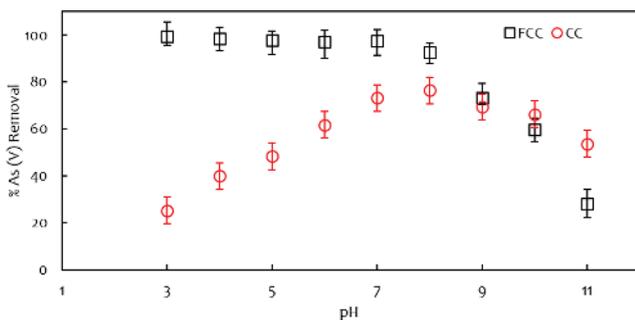


Fig. 10. Effect of pH on arsenic removal efficiency of FCC and CC.

no significant change in adsorption over the pH range from 3 to 7. The removal attenuated above pH of 9. According to Chen et al. 2010, As in aqueous media exists in distinct states [51]. These states are discussed in Table 3.

Chemical transformation of As in water with varying pH [51] is displayed below.

Following the theory provided in Table 3, the plot in Fig. 9 illustrates that FCC was more positively charged

Table 3
Transformation of As (V) in water

pH of aqueous solution	Form of As (V)
<2.2	H ₃ AsO ₄
2.2–6.98	H ₂ AsO ₄ ⁻
6.89–11.5	HAsO ₄ ²⁻
>11.5	AsO ₄ ³⁻

during a pH range of 3–7 which confirms electrostatic attraction between As and FCC surface [51]. The abrupt decrement in the removal efficiency beyond pH of 9 symbolizes electrostatic repulsion between the As (V) and FCC. The change in pH influences the FCC surface charge which affects surface-based adsorption kinetics [51].

The effect of pH on the removal of arsenic using an adsorbent mass of 5 gm for optimum 60 min contact time was shown in Fig. 10.

It was found that As adsorption followed the parabolic profile. The arsenic removal using MCC material was low

in acidic conditions. The efficacy of separation is found to increase with an increase in pH from 3 to 9. Fig. 11 illustrates the peak of arsenic contaminant removal in an aqueous solution at a basic pH of 9. The increase in the basic nature of the water decelerates As removal.

The improvement in adsorption of As (V) between acidic and neutral pH values may be due to the formation of As–Al and As–Fe complexes at the surface of MCC [52–54]. The decrease in the adsorption behavior at extreme acidic and alkaline pH values was likely due to the hydrolysis of clay, which resulted in the dissolution of clay minerals (Goldberg, 2002). Though the adsorption curve of the MCC and CC was found to follow the same pattern, the As (V) ions showed higher arsenic (V) removal capacity between 3 and 9 pH values. For MCC, the adsorption capacity slightly increased beyond pH 10. This may be attributed to coprecipitation of As (V) due to dissolved calcium (due to marble slurry addition) as reported by other researchers [55–57].

The findings indicate that variation in pH influences the surface charge of the MCC, which positively influences the sorption kinetics and sorption capacity of the metal ions on the MCC surfaces [58].

3.6. Effect of initial concentration

High arsenic removal is showcased at low concentration of arsenic in aqueous media from Fig. 12 [59]. FCC displayed higher arsenic removal in comparison with MCC and CC

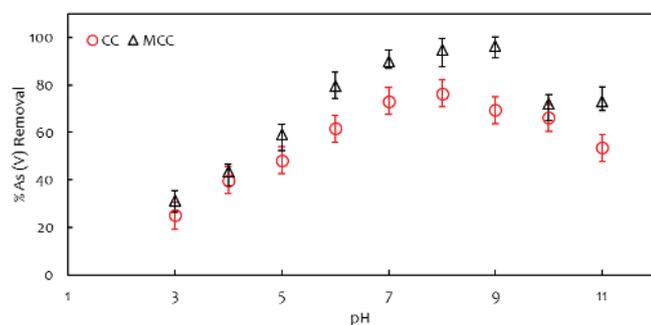


Fig. 11. Effect of pH on arsenic removal efficiency using MCC.

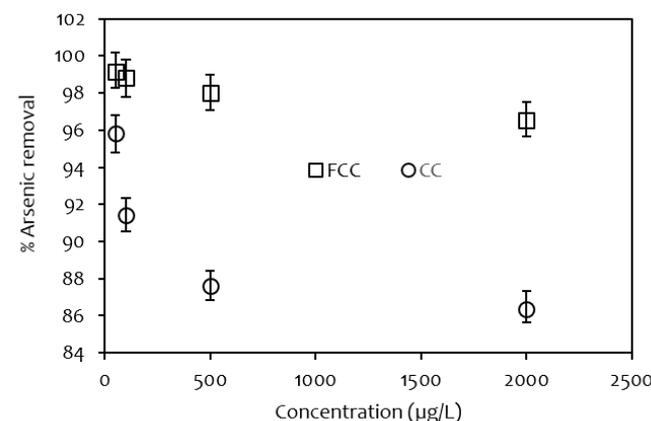


Fig. 12. Effect of adsorbate concentration on arsenic adsorption of FCC and CC.

at higher arsenic concentrations. The percentage of arsenic rejection decreased with increasing initial concentration. The result attests that large surface area to concentration ratio value showcases high As removal [54]. The CC and industrial additive in FCC and MCC jointly provided the more active surface of contact for greater adsorption of metal ions at lower concentrations.

The results (Fig. 13) indicated that arsenic removal efficiency decreases with the increase in concentration.

The transport mechanism of As to the MCC surface includes diffusion through the surface fluid film and diffusion through the pores in the media [60]. In the initial phase of adsorption, the As concentration gradient between the surface film and the pores was large. Therefore, the rate of adsorption accentuated. The rate of adsorption petered in the later stages due to the plummeting As diffusion into the MCC. At low concentration, the ratio of MCC area to the As concentration was large. Therefore, As separation from aqueous volume was high [60,61].

The clay and marble additive in MCC jointly provided the more active surface of contact which promoted greater adsorption of metal ions at lower concentrations. The results indicated that adsorption efficiency of MCC was high at lower concentrations in comparison with CC. The behavior pattern is consistent with the works done by other researchers [62–65].

3.7. Adsorption isotherms

Langmuir and Freundlich’s isotherms are used to model the distribution of arsenic adsorption with varying initial concentrations. As (V) adsorption isotherms are obtained from FCC and CC treatments and are shown in Figs. 14 and 16. These isotherms represent the adsorption behavior of As (V) on different adsorbents as a function of increasing aqueous As (V) concentration for a contact time of 60 min.

The Freundlich isotherm model was also used to analyze the result of As (V) adsorption on different adsorbents (Fig. 14). The Freundlich model can be expressed by Eq. (1).

$$\ln q_e = \ln k_f + \left(\frac{1}{n}\right) \ln C_e \quad (1)$$

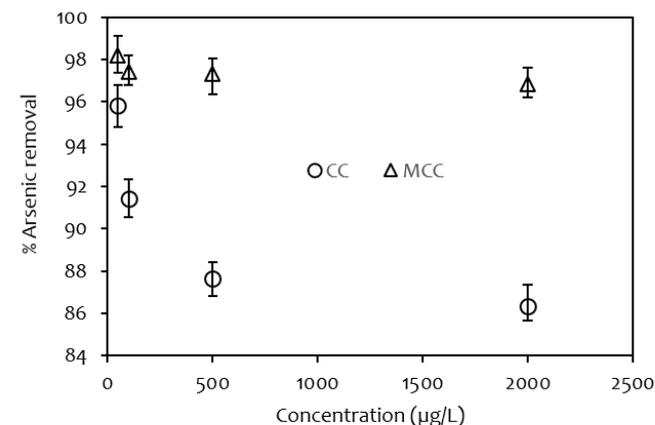


Fig. 13. The influence of initial adsorbate concentration of As (V) removal using MCC.

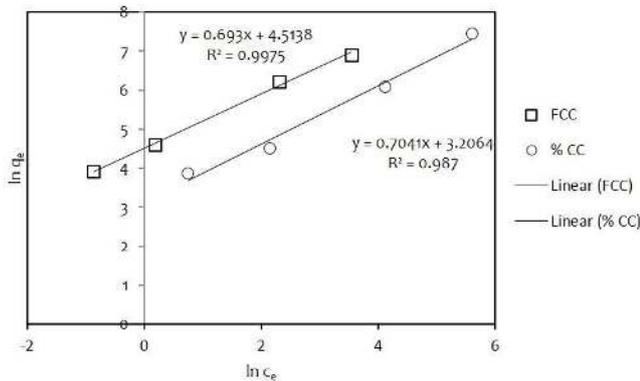


Fig. 14. Freundlich isotherm plot of FCC and CC in contact with arsenic-contaminated water.

where k_f and n are constants related to the adsorption capacity and affinity, respectively. The constants can be determined as the intercept and slope of a plot of $\ln q_e$ versus $\ln C_e$. The values of k_f and $1/n$ for the ceramics are tabulated in Table 4. The characteristic of the adsorption can be investigated using the value of $1/n$. The adsorption is said to be favorable when $1/n$ has a value between zero and unity, irreversible when $1/n$ is unity, and unfavorable when $1/n$ greater than 1 [66]. In this present case of FCC, value for $1/n$ for As (V) was 0.693, which indicates favorable adsorption.

The MCC displayed good fit to Freundlich isotherm model with the coefficient of determination of 0.995. The Freundlich parameters, k_f and n , were enumerated by plotting $\ln q_e$ vs. $\ln C_e$ (Table 5 and Fig. 15).

The Freundlich Eq. (1) deals with physicochemical adsorption on heterogeneous surfaces [67].

Table 4

The parameters obtained from fitting the sorption data of As (V) onto various porous ceramics using Freundlich Eq. (1) and R is the regression coefficient for the linear plot

Parameters	FCC	CC
$1/n$	0.6925	0.7041
k_f	91.16763913	24.68504
R^2	0.9975	0.987

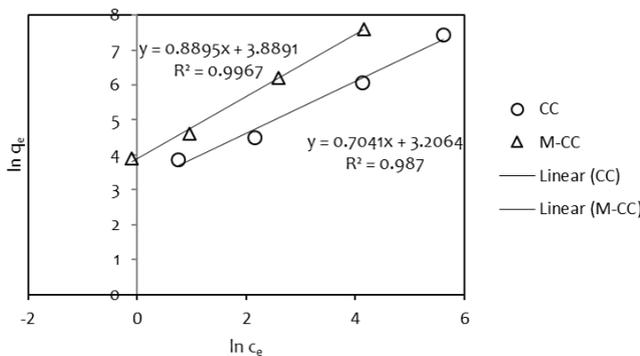


Fig. 15. Freundlich isotherm curve for MCC in contact with arsenic-contaminated water.

The value of n indicated the nature of adsorption. The value of $1/n < 1$ (as shown in Table 5) suggested that the adsorption process was heterogeneous [67,68].

The results of As (V) adsorption (Fig. 16) on FCC were analyzed by using the Langmuir model to evaluate parameters associated with the adsorption behavior. The linear form of the Langmuir equation at a given temperature is represented by Liu et al.

$$\frac{C_e}{q_e} = \frac{1}{bq_{max}} + \left(\frac{1}{q_{max}}\right)C_e \quad (2)$$

where C_e is the ionic concentration of arsenic ($\mu\text{g/L}$), q_{max} is the maximum amount of As (V) adsorbed onto 1 gm of the adsorbent ($\mu\text{g/g}$), b is the adsorption constant (L/mg) related to the energy of adsorption.

Experimental isotherm data acquired were correlated with the linear form of Langmuir model. The isotherm parameters related to the model are listed in Table 6. It could be seen that both q_{max} and b remain higher for As (V) adsorption onto ferrous-waste-mixed sintered CC compared with CC without ferrous waste.

Isotherm data fitted well to the Freundlich isotherm, suggesting that As (V) adsorption occurred in multilayer on the heterogeneous surface of the adsorbent [69].

3.8. Microbial removal

The percentage of *E. coli* removed by ceramics with and without the addition of ferrous waste and marble slurry is shown in Table 7. There was a slight uplift in the log removal rate due to the addition of ferrous material. Likewise, the use

Table 5

The parameters obtained from fitting the sorption data of As (V) onto various porous ceramics using Freundlich Eq. (1) and R is the regression coefficient for the linear plot

Parameters	MCC	CC
$1/n$	0.8895	0.7041
k_f	48.86689	24.68504
R^2	0.9967	0.987

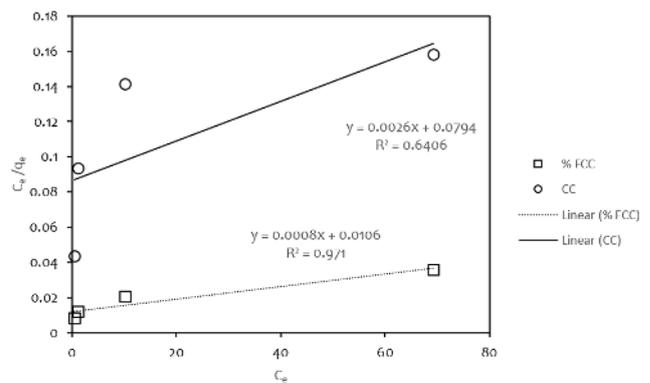


Fig. 16. Langmuir isotherm for FCC and CC in contact with arsenic-contaminated water.

Table 6

The parameters obtained from fitting the sorption data of As (V) onto various porous ceramics using Langmuir Eq. (2) and R is the regression coefficient for the linear plot

Parameters	FCC	CC
q_{\max}	1,250	384.6153846
b	0.075471698	0.032745592
R^2	0.971	0.6406

of iron compounds to enhance virus removal in a clay mixture has been tested by Brown and Sobsey in 2009 [70]. High *E. coli* removal efficiency was observed in MCC. The addition of marble slurry into the CC leads to no trace of *E. coli* from the effluent. Marble-based products are supposed to showcase antibacterial and antimicrobial properties [71]. Addition of marble can be used for remineralization of reverse osmosis or desalinated water as illustrated in the document of Omyaqua [72].

3.9. Mechanical characterization

The compressive and flexural strength of the CC experienced an increment with the addition of ferrous

powdered waste and marble slurry (Fig. 17). Addition of industrial waste lowered the porosity and surface roughness of the samples, thereby making the structure denser (Figs. 17a and b). The presence of a ferrous element (in FCC) increased average flexural strength ~2 times, and compressive strength increases ~1.33 times the strength of CC.

MCC displayed the highest strength characteristics. This may be due to the joint contribution of quartz and calcium carbonate in the marble-added ceramics. The presence of calcium carbonate (from marble slurry) created an extra surface area for the nucleation and growth of hydration products which might have developed additional strength in MCC [73,74]. The results agreed with the studies which involved incorporation of marble reject into ceramics for the construction and building material application for better structural properties [75–78].

4. Conclusion

- The investigation highlights the sustainable utilization of industrial wastes as additives in improving the arsenic removal efficiency and mechanical properties of porous CC.
- The fluorescence and mineralogy study of FCC and MCC displayed the evolution of hematite and calcite peaks, respectively, along with quartz.

Table 7

E. coli removal data from MCC, FCC, and CC

Ceramics	Initial concentration (I)	Final concentration (F)	I/F	Log reduction value (LRV) = $\log_{10} I/F$	Final/initial	The efficiency of <i>E. coli</i> filtration ($1 - F/I$)
MCC	4×10^8	No viable <i>E. coli</i> trace found	–	–	0	1
FCC	4×10^8	14×10^6	28.57143	1.455932	0.035	0.965
CC	2.4×10^6	4.5×10^3	533.3333	2.726999	0.001875	0.998125

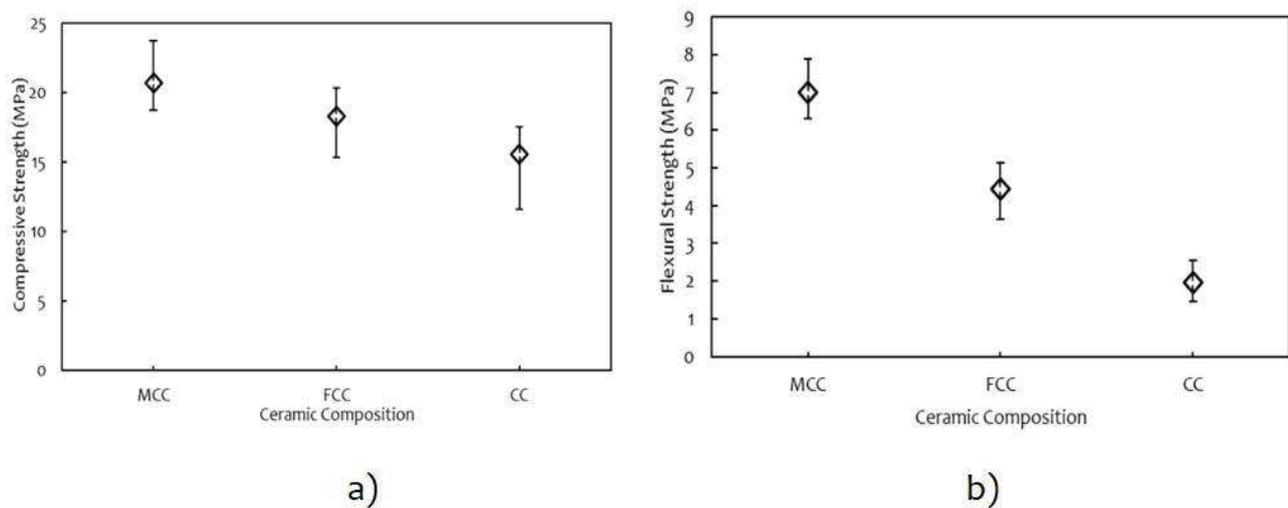


Fig. 17. Comparison of (a) compressive and (b) flexural strength behavior of ceramics with and without the addition of ferrous and marble slurry additives.

- The incorporation of marble slurry and ferrous waste served as a pore blocker to make the ceramic matrix structure dense.
- FCC gave arsenic removal of ~99% and MCC showed ~95% arsenic removal in a contact period of 90 min from an aqueous solution of 1 mg/L of As. In the batch adsorption study, the addition of marble slurry and ferrous waste to the CC, therefore, enhanced the arsenic removal efficiency.
- The effect of adsorption on the morphology of the ceramics was clearly distinguishable from SEM and FTIR curves. The surface of the ceramics became smooth and contained small pores.
- FCC showed a slight variation in arsenic removal percentage up to pH 7, and an abrupt fall in arsenic removal efficiency was observed after pH 9 due to changes in the surface charges. MCC illustrated a parabolic variation of arsenic removal with varying pH.
- Out of the Freundlich and Langmuir isotherm studied, the Freundlich isotherm provided the best quality fit for the experimental data for As (V).
- The compressive and flexural strength of ceramics increased after the addition of ferrous waste and marble slurry, which means improvement in the mechanical sustainability of the ceramics.
- The high *E. coli* removal from the additive-based ceramics was observed. The addition of marble into the filter composition has enhanced the antimicrobial properties of the filters. Incorporation of marble encourages the use of ceramic filter units without silver coating. This would reduce the cost of the filter unit.

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