Coal fly ash derived porous adsorbents as cost-effective and environmentally friendly sources of aluminosilicate for sequestration of aqueous and gaseous pollutants: A review

Seyed Mostafa Hosseini Asl[a], Maryam Khavarpour[a], Hamedreza Javadian[b,*]

[a]Department of Chemical Engineering, Ayatollah Amoli Branch, Islamic Azad University, Amol, Iran

[b]Universitat Politécnica de Catalunya, Department of Chemical Engineering, ETSEIB, Diagonal 647, 08028 Barcelona, Spain

[*]Corresponding author.

Email addresses: Hamedreza.javadian@yahoo.com; Hamedreza.javadian@upc.edu

Abstract

Coal fly ash (CFA) is an industrial by-product generated during coal combustion process in power plants. Due to its chemical and mineralogical composition, it is considered as an abundant and cheap source of aluminosilicate to produce advance materials (zeolites, catalysts, photocataltsts, geopolymeric) with high adsorption capacity towards aqueous pollutants such as petroleum compounds, dyes, heavy metals and radioactive contaminants. In this review, the utilization of CFA-based adsorbents as efficient and cost-effective alternatives to control the aqueous and gaseous pollutants was summarized. The ability of the prepared adsorbents to separate gaseous pollutants was discussed, and the recent investigations performed on applying various surface modification techniques to improve the adsorption capacity of CFA were explained.

Keywords: Environmentally-friendly adsorbents; coal fly ash; aqueous and gaseous pollutants.

1. Introduction

In the last few years, the great development of industrial activities have caused environment hazards (Wilson, 2013), and in many cases, irreparable damages to the ecosystems with consequences on human health (Gomes et al., 2017; Cong, 2018).

The effluents released by many industries such as textile, steel, metallurgy, electroplating, leather, paints and pigments, paper, plastics, tannery represent one of the main sources of wastewaters. Textile industry uses a large amount of chemicals during wet processing stages, and delivers considerable quantities of colored organic compounds (dyes) and other chemicals (Lin et al., 2016). Arora (2014) estimated that approximately 2% of the dyes produced are discharged directly in aqueous effluent, and 10% is subsequently lost during the coloration process. Moreover, the author considered that it is reasonable to assume that approximately 20% of the colorants are entered into the environment through effluents from the wastewater treatment plants. Many literature data have also reported the fixation percentage of dyes on different substrates and the relative loss percentage in textile effluents (Carmen et al., 2012). The presence of such toxic organic compounds in the industrial wastewaters creates many environmental problems due to negative effects on the biological life of pounds and rivers. Colored effluents cause high oxygen demand, fluctuating pH, resistance to biological oxidation and prevent sunlight penetration, decreasing photosynthetic activity in aqueous environment (Alcântara, et al., 2016).

Environmental problems are also arisen from the presence of heavy metals in the dyes' structures. Heavy metals have a catalytic role during dyes production and they form constituent structural elements (Visa et al., 2015). The presence of these toxic elements in wastewaters leads to serious risks for human and animal health by entering into the food chain (Mathur et al., 2007). Visa et al., (2012) underlined how some heavy metals such as copper, cobalt and, manganese in small concentration are accepted for living organisms, while their excessive levels can be detrimental to the organisms. Moreover, the authors, based on literature data (e.g Albanese et al., 2008), indicated that non-essential heavy metals (including cadmium, lead and copper) are dangerous for living organisms and create serious health effects since they tend to bio-accumulate.

Petroleum refinery wastewaters are the results of other industrial activities, resulting serious ecological impact on the environment (Al-Majed et al., 2012). The petroleum industry requires a

large quantity of water. Recently, de Abreu Domingos and da Fonseca (2018) explained that according to PETROBRAS (2015), a Brazilian petroleum refinery consumes an average of 0.9 m^3 water per m^3 of the processed oil. As a result, a large amount of wastewater is also generated (Coelho et al., 2006), which contains high levels of pollutants such as oil, ammonia, phenols, sulfides, heavy metals, and organic compounds (Nacheva, 2011) including the compounds that are refractory against biodegradation. For instance, the chemical oxygen demand (COD) in the raw wastewater of Abadan petrochemical complex (southwest of Iran) was reported to be as high as 20,000 mg O_2/L (Bahri et al., 2018; Shokrollahzadeh et al., 2008). The effluents of this complex and two other major petrochemical complexes together are discharged into Persian Gulf (Davodi et al., 2011), posing serious health hazards to all inhabitants and endangering the biological ecosystem of the region if they are not properly treated.

Furthermore, some industrial wastewaters contain radioactive pollutants (Lauer, 2018; Wen et al., 2016) that are even more hazardous than other contaminants for human and ecosystem. Radioactive wastewaters containing around 19% of total activity are perilous and considered as high-level wastes due to the high concentration of isotopes like ^{137}Cs, ^{134}Cs, and ^{90}Sr and the presence of long-lived actinides. The ecological risks of wastewaters containing medium-level radioactive wastes are precisely measured by the calculation of the main fraction of total activity in total volume of the polluted liquid (Lavrentyeva et al., 2014).

Besides the different kinds of wastewater pollution, air and gas pollutions seriously affect on human health and life. According to the data released by the World Health Organization in 2014, air pollution caused about 7 million deaths worldwide in 2012, indicating that air pollution is one of largest environmental health risks all around the world.

There are some conventional methods for the removal of pollutants, such as chemical coagulation, precipitation, oxidation, reverse osmosis, membrane, biological treatment, gamma radiations, adsorption that each of them has its own strengths and weaknesses (Golbad et al., 2017). Unlike most of these expensive treatment methods, adsorption techniques are much more favorable due to their cost-effectiveness, simplicity of operation and equipment, and significant removal efficiency. In addition, extended series of natural, synthetic and waste materials with high adsorption capacity can be applied as adsorbents (Javadian et al., 2018). Many literature data indicate that materials with mesoporous properties and regular pore structures are the best adsorbents used for air and water purification (Liu et al., 2013). However, beside various porous

materials, coal fly ash, a by-product of combustion process of coal in power plants, has attracted many researchers particularly in the field of wastewater treatment and separation of pollutants from gaseous flows, depending on its chemical, mineralogical and physical properties. Moreover, CFA is considered as an abundant and low-cost resource of aluminosilicate to produce efficient porous adsorbents like zeolite.

This article provides an at-a-glance review of CFA-based adsorbents and a comparison of their removal performances. The aim is to summarize the latest researches conducted on using CFA as an efficient adsorbent to control the aqueous and gaseous pollutions.

2. Content analysis

The latest outlooks about the world production rate of CFA as one the most dangerous threats to the environment and the possibility of its conversion to the efficient adsorbents as the best opportunity to eliminate aqueous and gaseous pollutions were explained. The literatures on the fabrication of zeolites, geo-polymers, catalysts and photo-catalysts from CFA by chemical synthesis methods including hydrothermal treatment, fusion, microwave, ultrasonic were reported. In addition, the surface modification techniques used for CFA-based adsorbents reported in the literatures such as fictionalization, impregnation, coating and etc. were obviously discussed. The main aim of this review was the comparison of the adsorption capacity of CFA-based adsorbents towards various aqueous and gaseous pollutions.

3. Coal fly ash

Coal is the second major fossil fuel source of world energy production. It is largely distributed worldwide with approved reserves of approximately 1000 billion tones in total. Despite Paris climate agreement, coal plays a key role for power generation in the foresighted future considering the growing demands for energy particularly in developing countries. Globally, with around 28% of total generated energy reported in 2016, coal has the second rank among other energy sources (**Fig. 1a**) (BP Statistical review). The latest report published in 2017 by the U.S. Energy Information Administration (EIA) evaluates the worldwide coal consumption since 1990 to 2040 (International Energy outlook, 2017). Based on EIA outlook, the amount of coal consumption between 2015 and 2040 will be around 160 quadrillion Btu (a quadrillion Btu roughly equals to the amount of energy in 45 million tons of coal). The consumption rate in

China and US will decrease but it will be compensated by India. **Fig. 1b** shows that China will remain the largest consumer of coal with a partial reduction in 2040 (about 73 QBTU), while India's coal consumption will grow with an average rate of 2.6%/year from 2015 to 2040. It is predicted that before 2020, India will surpass the US and be known as the second largest coal consumer. This is the time when the consumption of coal in OECD (Organization for Economic Cooperation and Development) countries will decrease by an average of 0.6%/year during 2015-40 due to their growing competitions for natural gas and renewable sources. In other places such as Africa, the Middle East and non-OECD Asia, the rate of consumption will gradually enhance until 2040 (International Energy outlook, 2017).

In addition, the EIA predicts that the amount of coal utilization for generation of power remains stable and then drops slightly due to replacement of coal by the natural gas, renewable and nuclear power in OECD countries (**Fig. 2a**). EIA's projections show that the amount of electricity generation will grow up to 2040 (with a growth rate of 2% annually) that coal is a significant portion in that (**Fig. 2b**) (International Energy outlook, 2017).

The continuous demands for coal lead to more coal extraction and subsequently coal ash (CA) production (**Figs. 3a** and **3b**). In detail, during coal combustion process in power plants, 80% coal fly ash (CFA) and 20% bottom ash (CBA) are generated (**Figs. 3c** and **3d**) that their accumulation in landfills leads to destructive effects on living organisms and eco-system. Focusing on CFA, the world annual rate of its generation in 2012 was reported to be around 800 million tons, from which China, India, US & EU generated 500, 140 and 115 million tons, respectively (Belviso, 2018).

The major parameters affecting on CFA quality are chemical composition of coal burned and combustion conditions i.e. the rate of oxidation and pulverization. Besides the presence of oxides like SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, K_2O, Na_2O, TiO_2, coal fly ash composition also displays variable amount of trace elements (e.g. As, Se, Cd, Cr) (Querol et al., 1996; Jegadeesan et al., 2008) that make it potentially toxic. Therefore, the direct use of CFA could have hazardous effects on the ecosystem (Huggins et al., 2016). **Table 1** displays the different composition of CFA collected from diverse regions or sites.

On the other hand, chemical-mineralogical composition of coal fly ash allows its usage in many fields. The rates of CFA utilization in US, EU, India and China was estimated to be around 50%, 90%, 60%, 67%, respectively (Yao et al., 2015). Although, the global average of 25% is not

adequate. CFA as a pozzolanic material has been prevalently employed in the manufacture of cement and concrete as a supplement for saving cement consumption. However, due to its alkaline features with negatively charged surfaces, the literatures have also indicated the ability of coal fly ash for the adsorption of metallic ions from solutions. Moreover, in the last few years, CFA usage as a cheap source of aluminosilicate has attracted scientists who have shown the successfully transformation of this waste material into zeolites, depending on its mineralogical composition characterized by the presence of large amount of amorphous material together with crystalline phases such as quartz, hematite, magnetite, anhydrite, and lime. In our previous paper, the transformation of coal fly ash into zeolite was investigated (Belviso et. al., 2010, 2011, 2018) and the successfully application of the final synthetic products to remove toxic elements from both water and polluted soil was demonstrated (Belviso et al., 2010; 2011). The most prevalent applications and properties of CFA are reported in **Fig. 4**.

4. Reduction of aqueous and gaseous pollutants

Comprehensive investigations on CFA adsorbents demonstrate their considerable performance in degradation of different aqueous pollutants such as organic and petroleum compounds, dyes, heavy metals, radionuclides, chemical oxygen demand (COD) and suspended solid (SS) (Alinnor, 2007; Cho et al., 2005). Furthermore, CFA adsorbents were used to capture multifarious gaseous pollutions including CO_2, SO_2, H_2SO_4 with acceptable efficiency (Jha and Singh, 2016). All the mentioned contaminants were effectively removed by a series of CFA-based adsorbents such as raw types, modified types, zeolites, geo-polymers, catalysts and photo-catalyst (Belviso, 2018). Most adsorbents from CFA are synthesized with conventional methods like chemical modification through acidic and basic treatment and impregnation with metallic or non-metallic compounds. In addition, hydrothermal treatment, fusion method, microwave method, ultrasonic method and a mixture of them are used to produce various zeolites, geo-polymers, catalysts and photo-catalysts (Hosseini Asl, 2018). The results clearly illustrate that the removal efficiency of the synthesized adsorbents is remarkably higher than that of raw CFA (Zhuang et al., 2016). The following sections discuss the latest achievements regarding the environmental applications of various CFA-based adsorbents.

4.1. Dyes reduction

Industrial wastewaters with variety of dyes are one of the most important world concerns. Due to carcinogenic and mutagenic properties of dye contaminants, the release of colored wastewaters into the environment seriously damages the aquatic organism, ecosystem and particularly the human health. Literatures have indicated that materials such as CFA can be effectively used as the adsorbent for the degradation of dyes from industrial wastewaters, and also as raw material for the synthesis of micro and mesoporous adsorbent materials.

CFA was used as a support material to produce mesoporous Al-MCM-41 and applied for the reduction of Methylene Blue (MB) from aqueous solutions. The results showed that at the pH of 10, the maximum uptake rate of catalyst reaches 277.78 mg g^{-1} within 120 min. Since the experimental data were fitted well by pseudo-second-order kinetic model and the Langmuir isotherm model, the authors claimed that a monolayer chemical adsorption taken place. Also, the spontaneous and exothermic entropy-reduction properties of MB adsorption were confirmed by thermodynamic studies (Zhou et al., 2015). The photocatalytic degradation of MB was reported by a type of nano-zeolite photocatalyst (Fe^{2+}-Na-X)-based CFA. Fe_2O_3-Na-X photocatalyst exhibited 90.53% degradation efficiency of MB at the pH of 6.9 within 3 h (Sufha et al., 2015). Excellent photocatalytic degradation of MB (around 98%) was obtained using a stable Ag-doped ZnO/CFA nanocomposite (Kim et al., 2015). Jin et al. (2014) evaluated the depolarization of MB by means of CeO_2/FACs photocatalyst and showed that the degradation efficiency reaches 60% within 5 h. In another research, Fe_2O_3-TiO_2/CFA composite was used for the photocatalytic oxidation of MB. The maximum degradation rate was reported to be 86.81% within 1 h under visible light (Zhu et al., 2016). Saud et al. (2015) also performed numerous experiments aiming to decompose MB using CFA/TiO_2 nanofiber. It was observed that the photocatalyst completely removes MB from the aqueous solution within 120 min. Two kinds of CFA composites including zeolite/hydrous iron oxide and zeolite/hydrous zirconia were employed for MB adsorption. Both composites exhibited an excellent adsorption rate (~100%) at pH up to 7 (Lin et al., 2016). MB dye was effectively removed from aqueous solution with 90% adsorption rate by the hydrothermally modified CFA (Visa and Chelaru, 2014). Similar research was carried out using hydroxysodalite formed from CFA. The maximum adsorption capacity of 94% for MB was obtained at the pH of 5 (Fungaro et al., 2011). Bromo sodalite synthesized from CFA illustrated relatively good performance for the adsorption of MB with the removal efficiency around 90% (Borhade et al., 2017). The efficiency of geopolymer synthesized from CFA (FAG) for MB

removal from aqueous solutions was also investigated (Novais et al., 2018). Alouani et al. reported FAG as an efficient adsorbent with adsorption rate of 37.04 mg g^{-1} whose adsorption process was well described by Langmuir isotherm model (Alouani et al., 2018).

The adsorption rate and photodegradation of Rhodamine B (RhB) were studied using Fe–N-TiO_2/CFA photocatalyst, and the results indicated that the photocatalyst with Fe/Ti ratio of 0.71% exhibits the best performance for the photocatalystic degradation of RhB (89%) under irradiation of visible light within 4 h. The elimination rate of total organic carbon (TOC) was estimated to be 75.3% after 4 hours of irradiation (Song et al., 2016). The authors proposed the three-step mechanism of photocatalytic degradation process by Fe-N-TiO_2/FAC as shown in **Fig. 5**. Firstly, Fe and N contents of the photocatalyst generate photo-induced electron-hole pairs to be reacted with RhB. Secondly, the Fe–O–Ti and Ti–N are formed, and the band gap is narrowed. Under visible light, the photocatalyst is excited and generates the electron-hole pairs causing the water molecules to decompose to •OH and •H. H_2O_2 created from the recombination of •OH that is reacted (photonic reaction) with Fe^{3+} contents of the catalyst as well (Pnag and Abdullah, 2012). Finally, the FAC floats on the surface of water without any aggregation. Thus, the adsorption rate increases since more catalyst particles are exposed to visible light (Wang et al., 2011).

Recently, RhB dye was efficiently degraded under irradiation of visible light using TiO_2/$ZnFe_2O_4$/AFAC photocatalyst nanocomposite with a maximum removal efficiency of 97.1% (Fan et al., 2018). Lin et al. (2014) investigated the degradation of RhB by a novel CFA photocatalyst (PVP/BiOBr/FACs). Under visible light irradiation, the removal efficiency reached 87%. In another study, hetero-structured CFA-ZnO semiconductor photocatalyst was applied for the degradation of RhB, Reactive Orange 4 (RO), and Trypan Blue (TB) dyes from aqueous solutions. The results indicated that under natural UV light within 90 min, the degradation percentages of RhB, RO and TB reach 99%, 98% and 96%, respectively (Thirumalai et al., 2016). Tungsten oxide-CFA composite was used as an adsorbent for removal of Barmaid Blau (BB) and Bemacid Rot (BR) dyes from aqueous solutions. The removal percentages of BB and BR were up to 80% and nearly 80%, respectively (Visa et al., 2015). Wang et al. (2015) studied the catalytic performance of CuAg-FAC for the degradation of Orange IV (O_{IV}), Orange II (O_{II}) and Reactive Black 5 (RB_5). The study showed that the catalytic activity leads to sharp reduction

of dyes within 20 min with 98.4%, 97.5% and 98.8% removal efficiencies for O_{IV}, O_{II} and RB_5, respectively.

Two types of modified CFA zeolites were employed for the adsorption process of Crystal Violet (CV). The highest adsorption capacities of CV were calculated to be 36.7 mg g^{-1} and 21.1 mg g^{-1} for each type (Bertolini et al., 2013). Similar research carried out by Bertolini et al. (2013) on other two types of CFA-zeolites indicated that CV dye is efficiently removed by both adsorbents. Fungaro and Magdalena surveyed the adsorption process of Acid Orange 8 (AO8) by means of a modified nanozeolite synthesized from CFA. A good adsorption performance of the synthetic product was observed with the removal efficiency of 98% (Fungaro and Magdalena, 2014). Another type of nanozeolite fabricated from CFA was used as an adsorbent to remove four types of dyes including Solophenyl Navy (SN), Solophenyl Turquoise (ST), Solophenyl Navy Hydrolysed (SNH) and Solophenyl Turquoise Hydrolysed (STH) from simulated textile wastewater. The maximum removal percentages of 78%, 85%, 66% and 91% were attained for SN, ST, SNH and STH, respectively (Cunicol et al., 2016). Overall, all the aforementioned examples clearly indicate that CFA adsorbents are effective in adsorption and degradation of dangerous industrial dyes (**Table 2**).

4.2. Heavy metals and agricultural nutrients

The CFA modified by functionalized mesoporous silica was successfully used to adsorb Cu^{2+} from aqueous solutions. The large surface area of the functionalized adsorbent leads to high removal efficiency of Cu^{2+}~98%, and the experimental data were fitted well by Freundlich isotherm model (Pizzaro et al., 2015). Copper ions were also removed by means of a kind of porous CFA geopolymer modified by iron ore tailing. The results indicated the highest uptake capacity of 13.41 mg g^{-1} at 40 ºC and pH value of 6.0. The study reported that both Langmuir and Freundlich isotherm models can fit the adsorption data of the spontaneous and endothermic adsorption process (Duan et al., 2016). Al-Harahsheh et al. (2015) conducted a series of experiments on the adsorption of Cu(II) using CFA geopolymer. The authors reported that under experimental conditions of pH=6, T=45 ºC, contact time=120 min, and adsorbent dose=2 g L^{-1}, the copper ions are efficiently removed with 93.9% removal efficiency. Moreover, the kinetic and isotherm studies illustrated that the pseudo-second-order kinetic and Langmuir isotherm models fit the adsorption data. Another type of mesoporous aluminosilicates was synthesized from CFA by cetyltrimethylammonium bromide (C_{16}TMABr) under alkaline conditions and

applied in adsorption process of Cu(II) from aqueous solutions. The evaluations showed that the increase of Si/Al ratio causes a considerable enhancement of adsorption rate with the highest adsorption rate of Cu(II) around 221 mg g^{-1} (Wu et al., 2012). In our previous report (Javadian et al., 2015), the adsorption rate of Cd(II) from aqueous solutions was examined by a type of geopolymer synthesized from CFA. Under optimum conditions, the maximum removal efficiency of Cd(II) was 84%. The isotherm and kinetic studies indicated that the Langmuir and pseudo-second-order models were in good agreement with the adsorption data. We also converted CFA to zeolite by fusion method and used it as an adsorbent to remove Cr(VI) ions from aqueous solutions. The results of our investigations illustrated that more than 80% of Cr(VI) ions are removed and the equilibrium data are well described by both Freundlich and Langmuir isotherm models. The adsorption kinetic followed pseudo-second order and the thermodynamic studies confirmed the spontaneous and endothermic nature of the process (Hosseini Asl et al., 2013). Recently, we studied the adsorption capacity of hydrous iron oxide/aluminumhydroxide composite loaded on CFA to remove Cr(VI) from aqueous solutions. At optimum conditions, the highest adsorption rate reached 33.3 mg g^{-1}, and the adsorption data showed good conformity with pseudo-first-order kinetic and Freundlich isotherm models. In addition, the adsorption process was predicted well using a fuzzy logic modeling with the coefficient of determination (R^2) of 0.95 with low deviation from the experimental data (Hosseini Asl et al., 2017).

Zhou et al. (2016) worked on polypyrrole/CFA-iron composites. They used the final magnetic adsorbent to remove Cr(VI) ions from wastewater, and the results showed that Cr(VI) ions are effectively removed by the synthesized adsorbent, and the maximum adsorption capacity of the adsorbent was reported to be 119.33 mg g^{-1}. CFA was also converted to zeolite X and used to adsorb Cr(VI) ions from aqueous solutions. The maximum adsorption capacity was 1.549 mg g^{-1} at optimum conditions (C_0=40 ppm and contact time=2 h) (Wang et al., 2017). Wen et al. (2011) investigated the adsorption of Cr(VI) from aqueous solutions by means of a chitosan-CFA composite. The highest uptake rate of the bioadsorbent was 33.27 mg g^{-1} at the pH of 5 within 50 min. Newly, Zeolite LTA was synthesized from CFA and employed to remove mercury from industrial wastewater. The results indicated that Hg(II) are effectively removed with the maximum adsorption efficiency of 94%. The adsorption data showed the best affinity with Freundlich isothermal and pseudo-second-order kinetic models (Attari et al., 2017). In another

recent research, a synthetic zeolite from CFA was utilized to remove Hg(II) from wastewater, and the highest adsorption percentage of 91.27% was obtained at the pH of 2 (Tauanov et al., 2017). Liu et al. (2013) studied the adsorption process of mercury using a hybrid mesoporous aluminosilicate (HMAS) synthesized from CFA and impregnated with zeolite A. The study reported that the adsorption of Hg(II) follows the chemical ion-exchange mechanism, and the highest adsorption efficiency of 95% is reached at the pH of 6.

Hydroxysodalite zeolite prepared from CFA was used to remove lead ions from aqueous solutions. The maximum adsorption rate of 98.1% by the synthetic zeolite indicated that Pb(II) is efficiently removed from aqueous solutions (Golbad et al., 2017). Additionally, lead ions were removed from aqueous solutions by a type of CFA geopolymer. Under optimum conditions (pH= 5 and contact time=2 h), the maximum removal efficiency reached 90.66% (Al-Zboon et al., 2011). Yu et al. (2016) examined the adsorption capacity of a CFA zeolite to remove Ni(II) ions from aqueous solutions. The study compared the adsorption capacity of the synthetic zeolite and the commercial zeolite (4A), concluding that CFA zeolite has higher adsorption capacity (75.6 mg g^{-1}) than zeolite 4A (27.6 mg g^{-1}). A highly valuable research was conducted by Chen et al (2016) for providing four types of CFA-zeolite to adsorb Ni(II) from aqueous solutions, in which the final removal efficiencies were obtained in the range of 92.5%-96.2%. Shukla et al. (2013) utilized an artificial zeolite made from CFA to remove As(VI) from aqueous solutions. At a low initial concentration of As(VI) (C_0=60 µmol), the maximum uptake rate was reported to be 40.48 µmol g^{-1}.

The complete sequestration of Mn(II) from aqueous solutions (100%) was conducted by 10 g of synthetic CFA zeolite (Belviso et al., 2014). A type zeolite prepared from CFA was used for multi-cation wastewater treatment containing Pb(II), Zn(II) and Cd(II). The results showed that in comparison to Cd(II) with 60% adsorption efficiency, Pb(II) and Zn(II) are efficiently and selectively removed with 100% and 70% adsorption efficiency, respectively (Visa et al., 2012). Shyam et al. (2013) reduced the concentration of Pb(II), Ni(II) and Cr(VI) ions from aqueous solution by means of a CFA adsorbent modified with calcium carbonate and phosphoric acid. The study reported that under optimum conditions, 1:10 CFA remove up to 90% of Pb(II), 50% of Ni(II) and 30% of Cr(VI). In a similar research, the Brazilian CFA was used for the production of zeolite Na-P1 to cleanup contaminants from acid mine drainage. The authors announced that most of the contaminants like manganese (99.8%), iron (>98%), copper (>96%),

calcium (>98%), zinc (81%), etc. are effectively removed during the first 30 min (Cardoso et al., 2015).

The single and multiple systems of heavy metal ions were adsorbed separately by using a CFA-zeolite. The results illustrated that Langmuir isotherm model well describes the adsorption process and the highest adsorption rate of Pb(II), Cu(II), Cd(II), Ni(II) and Mn(II) were obtained to be 65.75, 56.06, 52.12, 34.40, and 30.89 mg g^{-1} for the single system and 45.28, 32.86, 26.93, 16.25, and 14.63 mg g^{-1} for the multiple system (He et al., 2016). The multi heavy metal ions available in wastewater were removed by another type of CFA zeolite (MCM-41). According to the reported results, approximately 90% of heavy metals were omitted from the wastewater (Hui et al., 2009). In the same way, the adsorption of multi heavy metal ions from aqueous solutions was investigated using CFA coated with chitosan. The authors claimed that the acceptable performance of adsorbent is observed within 3 h. The adsorption capacity of Cr(III, VI), Cu(II), Zn(II) and As(V) were reported to be 36.22, 28.65, 55.52 and 19.10 mg g^{-1}, respectively (Adamczuk, 2015).

CFA zeolite was applied to reduce the ammonium concentration in swine wastewater. The results demonstrated that ammonium concentration is declined from 1205 mg g^{-1} to half (η= 52%), and the maximum adsorption capacity of 31 mg g^{-1} is obtained (Cardoso et al., 2015). Jiang et al. (2016) utilized Na-A zeolite prepared from CFA to control ammonium contamination in aqueous solutions. At optimum conditions, the highest removal efficiency was 60% with the adsorption capacity of 60.6 mg g^{-1}. Furthermore, a type of CFA zeolite was chemically modified with hydrous lanthanum oxide and used for phosphate removal from lake water. Considering to the results, particularly the maximum adsorption capacity of 43.4 mg g^{-1} and the removal efficiency of 60%, the performance of zeolitic composite was relatively satisfactory (Wang et al., 2016). The sequestration of nutrients (NH_4^+ and PO_4^{3-}) from swine wastewater was conducted using a CFA nano-zeolite. Under optimum conditions (sorbent dosage=10 g L^{-1}), the maximum removal efficiencies of 51% and 91% were obtained for ammonium (N) and phosphate (P), respectively. It is worth noting that by increasing the dosage to 80 g L^{-1}, the efficiencies reach 95% and 99% for N and P, respectively (Chen et al., 2012). He et al. (2017) studied the sequestration of nutrients (N and P) from wastewater using Na-P1 zeolite synthesized from CFA. Magnesium as an ion exchange agent was added to the adsorption process through two methods: adding directly to the solution and adding to the zeolite. As a result, a rapid precipitation

occurred, and the amount of N and P reduced. The authors reported that under optimum conditions (Z-P1 dosage=20 g L^{-1}, mg^{2+}=4 mM and t=30 min), the adsorption rates for N and P reach 65.2% and 92.3%, respectively, and any increase in the amount of adsorbent dosage results in higher adsorption rates. In addition, the adsorption efficiency was higher using the direct adding method (He et al., 2017). Information regarding the adsorption of heavy metals and agricultural nutrients by CFA adsorbents is summarized in **Table 3**.

4.3. Petroleum compounds

Studies show that CFA adsorbents can effectively remove petroleum compounds from wastewaters (Sasithorn et al., 2010). Asmaly et al. (2016) examined the adsorption capacity of aluminum oxide impregnated CFA to remove phenol from aqueous solutions. The investigation showed that phenol is completely removed with a maximum adsorption capacity of 2.105 mg g^{-1} at pH=7 within 120 min. The experimental data had good compatibility with Langmuir adsorption isotherm model. In another study, two CFA adsorbents were impregnated with aluminum (AFA) and iron (FFA) and then, applied for phenol removal from aqueous solutions (Chaudhary et al., 2015). The results illustrated that the modified adsorbents have higher adsorption capacities (AFA=14.07 mg g^{-1} and FFA=12.63 mg g^{-1}) compared to raw CFA (7.99 mg g^{-1}). The maximum removal efficiency of phenol was obtained to be 85.6%, 82.1% and 68% for AFA, FFA and raw CFA, respectively. Moreover, the authors reported that the Redlich-Peterson and Toth isotherm models adapt well with equilibrium data.

The degradation of O-methyl phenol (OMP) was conducted using a type of high aluminum CFA chemically modified by Fe(II)/Fe(III) (Meng et al. 2016). The fabricated catalyst (Fe-CFA) efficiently removed OMP from aqueous solution. High removal efficiency (100%) was achieved during catalytic ozonation of OMP at the pH of 11. The study claimed that the degradation process of OMP is described well using first order kinetic model. Ma et al. (2014) initially used CFA to adsorb Ni^{2+} ions and then, directly applied in zonation degradation of 2-chlorophenol. High adsorption rate (~100%) was observed by CFA catalyst, while the zonation oxidation process followed first order kinetic model. Similar research was conducted on oxidation degradation of phenol by CFA supported Co catalyst. The degradation rate of phenol was reported to be almost 100% at 90 min and 45 ºC. Also, the first order kinetic showed a good conformity with the experimental data, yielding the activation energy of 47 KJ mol^{-1} (Saputra et

al., 2012). CFA-cenospheres/Fe_3O_4 magnetic particles were used as a support to fabricate magnetic nanoparticles imprinted polymers (MNIPs) and magnetic molecularly imprinted polymers (MMIPs) adsorbents. Both adsorbents were surveyed for a competitive adsorption of nonylphenole (NP), 2-4 dichlorophenol (2-4 DCP), bisphenol A (BPA), 3-4 methylenedioxy phenol (3-4 MDP) and p-(tert-octyl) phenol (PTOP). The maximum NP, 2-4 DCD, BPA, 3-4 MDP and PTOP adsorption capacities were ~ 90, 85, 80, 90 and 90 mg g^{-1} for MMIPs and ~ 80, 80, 72, 73 and 75 mg g^{-1} for MNIPs. In addition, the authors asserted that the adsorption kinetic follows pseudo-second-order model, and the equilibrium data are fitted well with Langmuir isotherm model (Pan et al., 2013).

Furthermore, the modified CFA (impregnated with Al, Cd, Cu, Fe and Ni) was employed as adsorbent for organic acids. The removal efficiencies of β-napthaleneacetic acid, β-naphthoxyacetic acid, oxalic acid, trichloroacetic acid, cinnamic acid and indole-3-acetic acid were 100%, 95.25%, 85.71%, 78.26%, 75% and 63.63%, respectively (Li et al., 2012). In another research, CFA was modified through acidic and microwave treatment and used for reducing sulfonated humic acid (SHA) from aqueous media. The results indicated that only 3 g L^{-1} of CFA is needed for a complete removal of SHA at pH = 3 within 8 min (An et al., 2016). Evaluations illustrated that other organic compounds such as chrysoidine R (Matheswaran and Karunanithi, 2007), carbosyl group (Rao et al., 2006), and polychlorinated biphenyls (Nollet et al., 2003) can also be removed from the aqueous media using CFA-based adsorbents.

Recently, CFA-zeolite (Na-P1) was used to remove BTX from aqueous solutions. It was efficiently removed with the maximum uptake rates of 35%, 55%, 77% and 99% for benzene, toluene, o-xylene and p-xylene, respectively. The kinetic and isotherm studies demonstrated that the adsorption data are more adaptable with pseudo-second-order kinetic and Langmuir isotherm models (Bandura eta al., 2017). Another new achievement is related to the mixture of CFA and clay used in the purification of crude oil contaminations from aqueous media. The maximum turbidity and TDS removal efficiencies were reported to be 97.6% and 383.5 mg L^{-1}, respectively (t=20 min and pH=8.78) (Adams et al., 2017). Similar research was performed by Bandura et al. (2015) on the adsorption of oil products. Two CFA zeolites, Na-P1 and Na-X, were used to remove diesel fuels, yielding adsorption capacities of ~ 0.91 g g^{-1} and 0.79 g g $^{-1}$, respectively. The authors stated that CFA zeolites can be used as promising adsorbents for petroleum spills cleanup. The summary of some organic pollutants removed by the CFA-based adsorbents

together with the preparation method, adsorption conditions and the performance of each adsorbent is shown in **Table 4**.

4.4. Radioactive pollutants

CFA-based adsorbents have shown good adsorption performance towards radioactive pollutants available in wastewaters. Fungaro et al. (2012) studied the adsorption capacity of CFA zeolite-iron oxide magnetic nanocomposite to remove uranium from aqueous solution. The study reported that under optimum conditions (C_0=100 ppm, pH=3 and t=2.5 h), U(VI) is successfully removed (η=100%) from aqueous solution. The authors claimed that the monolayer adsorption with the highest adsorption capacity of 22.4 mg g^{-1} is obtained due to the good compatibility of adsorption data with Langmuir isotherm model. In another research, the mesoporous calcium-silicate material (MCSM) synthesized from CFA was used to remove a variety of radionuclides from wastewater. The results demonstrated that MCSM efficiently removes most of the radionuclides and hazardous metals up to 99.1 % of ^{58}Co, almost 100% of ^{60}Co and up to 98.7% of ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{95}Nb, ^{95}Zr, ^{99}Mo and ^{137}Cs. The authors reported that the adsorption rates of iodine (^{132}I and ^{133}I) and noble gases (^{133}Xe, ^{135}Xe, and ^{41}Ar) are not satisfactory due to the negatively charged iodine ions and low tendency of noble gases to enter in the reaction, consequently, they cannot participate well in the electrostatic interaction with the negatively charged surface of the adsorbent (Qi et al., 2015). In addition, the study proposed the adsorption mechanism of Co(II) with four consecutive steps including hydration, hydrolysis, surface complexation and ion exchange with Ca(II) as shown in **Fig. 7**. Firstly, the hydrated cobalt ions diffuse into the solution, reach the boundary layer and surround MCSM particles. Secondly, the hydrated cobalt ions diffuse into the boundary layer and the hydrated cobalt cations are released among water molecules. In the next step i.e. complexation, the surface interactions and ion exchange occur between Co(II) cations and SiOH or CaOH of the adsorbent and the calcium ions are adsorbed onto the surface and edge of the particles. Finally, in the hydration step, the diffusion of Co(II) cations to the interlayer of C-S-H(I) takes place and as a result, the cations are exchanged with calcium.

Noli et al. (2016) investigated the adsorption of barium (Ba) and europium (Eu) radionuclides from aqueous solutions using a type of CFA zeolite containing Na-Y and SOD. They reported that CFA zeolite shows excellent performance in the adsorption process of both radionuclides

(η=100%) particularly at optimum conditions (T=308 K, pH=4 and t=2 h). The adsorption properties of two kinds of modified CFA zeolites (A and B) to remove uranium from aqueous solutions were also examined. The results indicated that U(VI) is effectively removed within 1 h with maximum removal efficiencies of 90% and 99.5% for A and B, respectively (Xie and Zhou, 2017). Lieberman et al. (2015) conducted a series of experiments for the stabilization of four radionuclides on the surfaces of two types of CFA. The study showed that Ce(III) and Ce(IV) ions are completely removed from aqueous solutions, while the adsorption rate of Cs(I) is not satisfactory (30%) and fixation of Sr(II) is not observed. Strontium ions are converted to $SrCO_3$ (a carbonate salt) and successfully trapped on the surfaces or in the pores of the adsorbents. Furthermore, the authors proposed the fixation mechanism of the radionuclides by CFA adsorbents. The study remarked that Cs^+ interacts with CFA surface through ion exchange mechanism (**Fig. 7a**), while in the case of $Ce^{3+/4+}$, the complexes of cesium and FA are formed due to the rapid equilibrium and precipitation. It was claimed that the main reason for this phenomenon can be the interaction between cesium radionuclides and the functional groups available on the surface of the adsorbent (**Fig. 7b**). The fixation mechanism of strontium ions on the surface of CFA is shown in **Fig. 7c**.

4.5. COD and SS reduction

The chemical oxygen demand (COD) and suspended solid (SS) are considered as important factors in the quality evaluation of water and wastewater. In fact, COD and SS are the indexes for the identification of the amount of organic compounds and the presence of solids in water and wastewaters. COD rate indicates the mass of oxygen required for oxidation of organic compounds in water or wastewater. The high concentration of both COD and SS in water and wastewater are detrimental for human health and particularly aquatic organisms. One of the most effective adsorbents employed to remove COD and SS is CFA. Hu et al. (2017) surveyed the adsorption process of COD and SS from soybean and dairy wastewaters by means of the aluminum and iron-based coagulants prepared from CFA. It was observed that the maximum removal efficiencies of COD and SS achieved at the pH of 8 are around 20% and 80% for soybean wastewater, and around 65% and 92% for dairy wastewater. A series of experiments were conducted to remove COD from aqueous solutions containing o-methyl phenol (OMP) by using a CFA catalyst modified by Fe(II)/Fe(III) oxides. The results demonstrated that at pH=11,

the highest removal efficiency of COD is 44.5 % (Meng et al., 2016). Devi and Dahiya (2006) utilized CFA to eliminate COD from domestic wastewater. They announced that the maximum removal percentages of COD by CFA, brick kiln and commercial activated carbon are 87.84%, 83.22%, and 99.35%, respectively. The operational conditions to reach the reported results are the adsorbent dosage of 180 g L^{-1} within 3 h, 60 g L^{-1} within 4 h, and 45 g L^{-1} within 5 h for active carbon, CFA and brick kiln, respectively.

4.6. Capturing gaseous pollutants

The utilization of solid waste residual generated by industries as an adsorbent to capture and store (CS) various gaseous pollutants has attracted the attentions of many researchers. CFA as an abundant waste is considered as a cheap source of aluminosilicate that is able to capture gaseous contaminants (Wee, 2013). In one study, the removal of CO_2 was carried out using raw CFA through surface adsorption and carbonation process. Carbon dioxide capturing was performed in a tubular reactor with the possibility of controlling the gas flow temperature and pressure using a PID controller (**Fig. 8**). As can be seen in **Fig. 8**, initially, nitrogen gas is injected to carry all gas samples from the reactor to Chromatography (GC). Then, CO_2 is released to be captured by solid CFA and the amount of CO_2 in the outlet is measured by GC equipped with a thermal conductivity detector (TCD). The highest rate of CO_2 capturing was obtained up to 304.7 µmol g^{-1} CFA consisting of 2.9 and 301.8 µmol g^{-1} of CFA for adsorption and carbonation, respectively (Siriruang et al., 2016). Carbon dioxide capturing was also conducted by two types of CFA adsorbents modified with NaOH and CaO (WNCF and WNC). Both adsorbents showed excellent performance with high CO_2 adsorption efficiencies of 94.7% and 105.1% for WNCF and WNC, respectively (Lee et al., 2014). Dananjayan et al. (2016) investigated the capturing and storage capacity of CFA for sequestration of CO_2. The authors mentioned that the highest capturing and wet carbonation capacities of CFA are 26.3 g kg^{-1} of CFA at 10 bar within 1h and 50.3 g kg^{-1} of CFA at 4 bar within 2 h. The uptake capacity of CFA for capturing CO_2 was surveyed by Mazzella et al. (2016) at different pressures and temperatures. The results indicated that CO_2 is completely captured by CFA under specific operational conditions (P=15 bar, T=45 ºC and t=134.8 min). CO_2 sequestration was conducted at high temperature using a CFA modified by potassium (K-FA). The outcomes of experiments illustrated that at 700 °C, CO_2 uptake rate reaches 1.45 mmol g^{-1}. In addition, the author reported that in the presence of

Li_2CO_3, this rate increases to 2.38 mmol g^{-1}, and CO_2 uptake of 90% is reached within 5 min (Sanna and Maroto-Valer, 2016a). Similar research was carried out using CFA adsorbent modified by Na_2CO_3 (Na-FA). The maximum CO_2 uptake rate of 9.24 wt% was achieved at 700 °C in the presence of 12.5% CO_2 and 12% H_2O. This rate increased to 11.2 wt% by the addition of 20% Li_2CO_3 (Sanna and Maroto-Valer, 2016b). Yan et al. (2016) studied the adsorption process of CO_2 at different operational conditions using high-alumina CFA. The study showed that at the temperature of 323.15 K and the flow rate of 95 mL min^{-1}, the highest CO_2 uptake rate reaches 8.67 mg g^{-1}.

Zeolite A and 13X synthesized from CFA were employed in the CO_2 capturing process. Based on the reported outcomes, both zeolites showed good performance for the capture of CO_2 (Zeolite A=145 mg g^{-1} and 13 X=220 mg g^{-1}) (Soe et al., 2016). Kalvachev et al. (2016) examined the adsorption of CO_2 using Zeolite X synthesized from CFA. It was observed that the uptake rate increases to 60 mg g^{-1} at 22 °C within 1 h. Recently, MCM-41 fabricated from CFA was used as a cheap silica support for impregnating polyethyleneimine (PEI). Thereafter, this novel adsorbent (MCM-41-PFA) was applied to capture CO_2. The results exhibited that CO_2 equilibrium uptake efficiency of 90% is obtained at 75 °C within 1 h, and the uptake rate sharply increases to more than 11 wt% in the first 5 min (Panek et al., 2017). Another recent report is related to the CO_2 capturing using CFA-based zeolite X modified with PEI by wet impregnation technique. According to the findings, the adsorbent (Na-X-PEI-600) efficiently captures CO_2 with the maximum adsorption capacity of 26 mg g^{-1} (Dindi et al., 2017). The capturing capacity of CO_2 using zeolite 13 X prepared from CFA was studied by Zhang et al. (2017). The results indicated that CO_2 is effectively adsorbed with the highest adsorption capacity of 225 mg g^{-1}.

Newly, in an interesting research, the adsorption and carbonation capacity of CO_2 were examined using CFA to produce carbonated CFA for application in cement manufacturing. The highest carbonation efficiency and CO_2 uptake rate were reported to be 83.5% and 32 g kg^{-1} of CFA, respectively (Ebrahimi et al., 2017). Shu et al. (2015) investigated the adsorption capacity of CFA adsorbents to remove sulfuric acid mist from lead-acid battery plants. The adsorption experiments were performed using a fixed bed apparatus with the possibility of placing 300 g of the adsorbent in a vertical organic glass reactor equipped with an inlet for nitrogen as a sweeping gas (**Fig. 9**). As can be seen in **Fig. 9**, NaOH bath is placed after the reactor to ensure a complete removal of H_2SO_4 mist. The results demonstrated that H_2SO_4 mist is completely removed

(η=100%) by NaOH/CFA and CaO/CFA, while the maximum removal efficiency of H_2O/CFA is only 60%. It is asserted that the adsorption mechanism is the combination of electrostatic interaction and chemical precipitation at the interface of the adsorbent and H_2SO_4 mist. In a similar study, the oil fly ash (OFA) was activated by adsorption of CO_2 and chemicals to remove H_2S from a gas stream. It is reported that during the first 15 min, H_2S is effectively removed (η=100%) using AC-NH_4OH with the highest uptake rate of 0.3001 mg g^{-1}(Aslam et al., 2015). Magnetic catalyst ($CuCl_2$-MF) synthesized from CFA was employed to eliminate mercury from the flue gases. At optimum conditions, the highest uptake percentage of Hg^0 was reported to be 90.6% (Yang et al., 2016). In a creative research, modified CFA was used to control the secondary release of mercury from the flue gases of a coal-fired power plant. The authors reported that by adding $CaBr_2$ to CFA before coal combustion process, the removal efficiency of Hg reaches 94% (He et al., 2016). Furthermore, the catalytic reduction of NO was conducted by two types of zeolite Y (LY-Cu and LY-Fe) synthesized from CFA. The denitration mechanism of CFA catalysts modified with various gases (O_2, N_2 and Ar) was recently studied using low-temperature plasma technology. It was reported that at optimum adsorption conditions, NO uptake efficiency reaches 80% ((Nie et al., 2017). The performance of various CFA adsorbents in the adsorption process of gaseous pollutants is summarized in **Table 5**.

9. Conclusions

The opportunities and challenges of CFA as a by-product of coal factories and coal-fired power plant were investigated. Based on the published outlooks, the production rate of CFA will increase until 2040 due to the global growth in demand for energy. The irregular disposal of CFA in landfills leads to irreparable damages to the environment. The remarkable amount of aluminosilicate available in CFA can be applied to fabricate valuable advance materials such as zeolites, catalysts, photocatalysts, geopolymers, etc. As a result, the conversion of CFA to valuable products seems to be necessary. The adsorption capacity of CFA-based adsorbents produced by various conversion techniques consisting of both chemical syntheses and modification methods were analyzed. In comparison to commercial adsorbents, the CFA adsorbents as an efficient and environmentally-friendly product can be used to clean up the real wastewaters containing dyes, petroleum compounds, heavy metals, agricultural nutrients, radionuclides, COD, SS. Moreover, the literatures confirmed that CFA-based adsorbents have

good performance in the reduction of different gaseous pollutants such as CO_2, SO_2, H_2SO_4, H_2S, Hg, NO, BTX. Based on our findings during several years of research, this profitable by-product can be utilized to provide both a healthy environment and a high rate of income for producers.

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