Review article

Application of graphene for decontamination of water; Implications for sorptive removal

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A B S T R A C T

Graphene has shown to be a promising material for many different fields due to its unique structure with exceptional physicochemical characteristics and capacities. It has been widely used for water treatment as an adsorbent for heavy metal/loids and organic contaminants. Many studies have focused their attention on producing different graphene based nano materials such as graphene/metal nano particle composites, modified graphene, graphene-complex oxide composites and graphene/semiconductor hybrids along with improved performance and inherent qualities of graphene for contaminant removal in water. Its outstanding ability for excellent immobilization of various contaminants such as organic dyes; methylene blue (MB), methyl orange (MO), rhodamine, inorganic pollutants such as chromium, arsenic, uranium, mercury, fluoride, antimony, zinc, lead and copper are due to the high surface area and functional groups of graphene. Simultaneous removal of organic and inorganic contaminants with desorption studies have proved favorability of graphene as an adsorbent with reusability. In this review, an attempt has been made to assess recent developments of graphene's capacity to decontaminate wastewater and to explain the mechanisms of adsorption towards the future scope.

1. Introduction

Environmental remediation using nano materials has become a hot topic in the recent past as one of the prominent social aspects of nanotechnology where numerous applications of nanomaterials are considered in the decontamination issues of water, air, and soil. For instance, in early applications, such nano materials are being used in halocarbon decomposition of pesticide removal (Pradeep and Anshup, 2009). Different nano materials varieties such as carbon nano tubes (Long and Yang, 2001; Wang et al., 2008), graphene (Chandra and Kim, 2011; Chandra et al., 2010) and material composites based on carbon nano tubes (Yang et al., 2011b) have been extensively used as they have proven high sorption capacities in the organic and inorganic pollutant removal from aqueous solutions (Rao et al., 2007; Ren et al., 2011a; Seymour et al., 2012; Wang et al., 2012b).

Carbon has been considered as a versatile absorbent during the recent years in the pollutant removal from aqueous solutions (Ahmad et al., 2014; Suhas et al., 2016; Yin et al., 2007). Different forms of carbon and their particular composites were studied to improve

adsorption efficacy (Rajapaksha et al., 2016; Suhas et al., 2016; Yin et al., 2007). Since the characterization and isolation of graphene (Geim and Novoselov, 2007), which led the inventors sharing Nobel Prize in Physics for 2010, there has been a tremendous boom of interest in both academic and commercial arenas. It is the latest member of the family of carbon, believed to be one of the wonder materials of this century. In the past few years, graphene has become the centre of attention as another allotrope of carbon in many research fields. Graphene and its composites can be utilized in several applications thanks to its unique two dimensional nature and along with associated band structure (Rao et al., 2009). The structure of graphene is a carbon nanomaterial with a single layer of sp^2 hybridized carbon atoms which are arranged in six-membered rings with a theoretical specific surface area value of 2630 m^2/g (Zhu et al., 2010) apart from its in built strong electrical, thermal and mechanical properties (Liang et al., 2009; Su et al., 2009). It has a honeycomb lattice structure where the carbon-carbon bond length of 0.142 nm (Slonczewski and Weiss, 1958). GO is functionalized graphene which has varying oxygen containing groups on the surface (Wang et al., 2013). Graphene is obtained from graphite that exists in

GO and reduced graphene oxide (rGO).

As a consequence, graphene has been investigated for certain characteristics and properties suitable for the purpose of water purification (Aghigh et al., 2015) although most of the graphene composites were proposed for electronic and catalytic applications. Due to its unique structure with exceptional physicochemical characteristics, i.e. light weight of graphene allows it to withstand at high pressure, it has additionally gained a tremendous interest in environmental remediation by pollutant removal (Lü et al., 2012; Aghigh et al., 2015).

There have been several reviews on the application of Graphene Oxide (GO) and Graphene Nanosheets (GN) in many different fields (Huang et al., 2011; Lü et al., 2012; Machado and Serp, 2012; Yao et al., 2012b; Zhu et al., 2010) however, very few reviews are available on graphene as an adsorbent in water pollution remediation (Lü et al., 2012; Aghigh et al., 2015; Gandhi et al., 2016; Upadhyay et al., 2014). Reviews of sorptive use of graphene for water pollution remediation is limited (Jiang et al., 2016). Here we attempted to summarize recent research work for graphene based water purification intending benchmarking purpose to provide an insight into the focused knowledge in sorptive removal of pollutants. In this review, recent research advances are being discussed bearing this view point, and attention is directed towards the usage and applicability perspective of graphene in water remediation process for future.

2. Synthesis and characteristics

At present, several innovative techniques have been developed for the synthesis of graphene materials (via micro-mechanical exfoliation of highly ordered pyrolytic graphite, epitaxial growth, chemical vapor deposition etc), including GO and reduced graphene oxide (rGO) altering the previous methodologies such as hydrazine reduction, thermal annealing, $NaBH_4$ reduction (Singh et al., 2016). Mechanical exfoliation, epitaxial growth by chemical vapor deposition, chemical reduction of graphite oxide, liquid- phase exfoliation, arc discharge of graphite, in situ electron beam irradiation, epitaxial growth on SiC, thermal fusion, laser reduction of polymers sheets and unzipping of carbon nanotubes are among the most common synthesizing methods (Wang et al., 2013; Singh et al., 2016; Yu et al., 2016b; Chabot et al., 2014). It is well known that the GO is the most common precursor that is used currently for the synthesis of graphene materials. The four most common synthesis methods (Brodie, Staudenmaier, Hummers and Hofmann method), their modified and improved forms have been well reviewed in the literature (Singh et al., 2016; Khan et al., 2016; Yu et al., 2016b; Chabot et al., 2014). Afterwards, GO can be reduced to form graphene via various methods; chemical; thermal/solvothermal/hydrothermal; microwave; photo/photo catalyst reduction etc (Singh et al., 2016).

Graphene possesses unique characteristics which creates it attractive material for contaminant removal in water although mostly studied are electrical and electronic features. Among many different properties, graphene and its derivatives offer the highest strength, highest specific surface area and low production cost (Perreault et al., 2015). At the same time, GO has abundant oxygen containing functional groups such as hydroxyl, carbonyl, carboxyl, epoxide etc. on the surface which plays a major role forming strong complexes with metal ions that allows GO to act as an adsorbent for heavy metal ion removal and preconcentration (Lü et al., 2012). There are differences between GO and rGO which can be observed in terms of functional groups on the surface, crystal structure, optical property and defective structure. Graphene Oxide shows strong acidity, with high adsorption for cations and basic compounds. On the other hand, graphene shows hydrophobic surface, and due to strong π-π interaction, it has high adsorption capability to chemicals. Hence, modification of graphene or GO can be associated with organics or metal oxides which can produce different kinds of nano composites while enhancing separation efficiency and adsorption capacity (Wang et al., 2013).

Both GO and rGO are now being focused as ideal materials for wastewater treatment from recent studies due to its high specific surface area, tunable morphological features, controllable porosity, inherent hydrophobicity and oleophilicity, chemical manipulability, tunable surface functionality, cost effective synthesis and good thermal/environmental/chemical stability (Liu et al., 2016; Chandra et al., 2010; Zhao et al., 2011b; Nadres et al., 2016). In fact, graphene's role in environmentally concerned applications like water purification has a limited scope due to the inability of large scale synthesis (Sreeprasad et al., 2011). Several studies even have used 3D rGO composite materials and have been proven to be effective removal of organic contaminants and dyes (Jayanthi et al., 2016; Maliyekkal et al., 2013; Ma et al., 2015). To avoid bare nanoparticle's aggregate formation, graphene has been used as a supporting material as it is highly stable with large surface area and in size (Liu et al., 2012b). Furthermore, chemical modifications have been used to enhance the properties of GO and rGO where several attempts have been reported in the production of GO and rGO composites (Liu et al., 2012d; Azizi et al., 2016; Liu et al., 2016). Magnetic graphene is one of the novel composite which has been widely tested for decontamination of water as a recyclable material (Vuong Hoan et al., 2016; Gandhi et al., 2016; Lu et al., 2014). It is being a reported fact that GO and rGO supported materials tend to have a high binding capacity comparatively to nano particles (Chandra et al., 2010).

3. Graphene as a material for water purification

Graphene seems to be the material which has been subjected to number of various functionalization and composite formation. Many different composites and functionalized graphite and its different forms have been tested for water purification ability. Although pure graphene is hydrophorbic which limits the use in water treatment due to lack in dispersion, GO and rGO are effective in water purification due to the presence of surface functional groups. Negative surface charge of the GO and rGO helps efficient removal of cationic contaminants such as heavy metals. It is well known that the electrostatic interaction is the mechanism behind the cationic heavy metal remediation. Since GO undergoes transformation with time, its application in water treatment has become limited. Over GO, rGO is becoming efficient material in water purification since it possesses large surface area. As rGO does not possess large negative charge, it shows effective removal of anionic impurities (anionic dyes, As(III), As(V), Se(V), and Cr(VI)) (Li et al., 2015; Ma et al., 2012). In order to improve the efficiency of graphene materials in environmental applications, functionalized graphene and graphene nanocomposites are prepared by anchoring them with specific functional groups and various nanomaterials, which have produced superior water purification applications compared with bare graphene, graphene oxide, and reduced graphene oxide (Kumarathilaka et al., 2016; Dinda et al., 2013; Jiang et al., 2015). Interestingly, like biochar, graphene has also become a material which can be used to remove many different contaminants from water including pesticides, pharmaceuticals, gasoline compounds etc. (Carmalin Sophia et al., 2016). The graphene-based composites showed great improvements in their performance toward adsorption capacity and catalytic activities (Hu et al., 2016; Sreeprasad et al., 2011).

4. Metal ion removal

As metal ions cannot be degraded by chemical reactions and bioprocess, adsorption is proven to be the most effective way in removal of heavy metal ions. GO has been used as a potential adsorbent due to strong functional groups on its surface (Wang et al., 2013). Effective removal of Cu(II) ions was suggested by interacting the positively charged Cu(II) ions and negatively charged oxygen groups on GO (Yang et al., 2010). Even, GO has been used in adsorption of Co(II), U(VI) and Pb(II) (Liu et al., 2014). Adsorption characterization of GO towards Cd,

Pb, Cu and Zn were investigated using powder X-ray diffraction (XRD), scanning electron microscopy (SEM), infrared spectroscopy (FT-IR) and X-ray photoelectron spectroscopy (XPS) (Sitko et al., 2013). Maximum adsorption capacities were found out to be at pH $=5$ were 530, 1119, 294, 345 mg/g for Cd(II), Pb(II), Cu(II) and Zn(II) respectively. Even maximum adsorption could be achieved for a wide range of pH values (4–8 for Cd(II), 3–7 for Pb(II), 3–7 for Cu(II) and 5–8 for Zn(II)) (Sitko et al., 2013). Competitive adsorption affinities were in the order of Pb (II) $>$ Cu(II) $\gg$ Cd(II) $>$ Zn(II). Data modeling suggested that metal ion sorption on GO is a monolayer coverage where the adsorption is being controlled by chemical adsorption by the involvement of surface complexation of metal ions with oxygen containing groups that are on the surface of GO. An adsorption capacity of 94 mg/g was achieved by GO/cellulose hydrogel (Chen et al., 2016).

Synthesis of layered GO nanosheets was carried out from graphite using modified Hummers method where it has been used to act as a sorbent for Cd(II) and Co(II) ions removal from aqueous solutions where a strong dependency on pH was evident by Cd(II) and Co(II) sorption on GO nanosheets while a weak dependency was on ionic strength (Zhao et al., 2011b). At pH 6.0 ± 0.1 and T $=$ 303 K, maximum sorption capacities of Cd(II) and Co(II) were about 106.3 and 68.2 mg/g, respectively with a highest reported value. For ex-situ environmental remediation by heavy metal ions, it has been suggested that GO nanosheets may be suitable although they are expensive compared to carbon materials (Zhao et al., 2011b).

With metal oxides also GO and GNs can be incorporated and composites with GO and metal/oxide have been used as effective adsorbents in the past (Upadhyay et al., 2014; Hur et al., 2015b). A flower-like TiO$_2$ on GO hybrid (GO–TiO$_2$) was prepared and has been applied for the removal of Cd(II), Pb(II) and Zn (II) ions (Lee and Yang, 2012). The reported adsorption capacities of the GO–TiO$_2$ hybrid reached 72.8, 65.8 and 88.9 mg/g, respectively at pH 5.6 (Lee and Yang, 2012). The covalent anchoring of TiO$_2$ and rGO in the TiO$_2$-GO composite leads to a reduction in electron–hole recombination which in turn resulted high Cr(VI) reduction ability (Zhang et al., 2012b). Various magnetic graphene composites have been used by several research groups (Zhang et al., 2013b; Hur et al., 2015b; Ting et al., 2016; Yusuf et al., 2015). To overcome graphene's separation problems, the incorporation of magnetic particles with GO or GNs offers an effective approach. Hence by loading magnetite nanoparticles, it can avoid the possibility of agglomeration and restacking of graphene sheets, while consequently enhancing adsorption capacity and higher surface area (Sun et al., 2011).

Synthesis of rGO–metal/metal oxide composites were well received in water purification (Sreeprasad et al., 2011; Upadhyay et al., 2014). Inherent reduction ability of rGO was used and the data revealed that rGO composites are formed through a redox-like reaction between rGO and the metal precursor. It was reported that the model compounds 3-D rGO, Ag–rGO and MnO$_2$– rGO, were highly effective in the removal of Hg(II) with a maximum adsorption capacity of 185 mg/g for 3-D rGO (Sreeprasad et al., 2011; Wu et al., 2015). High distribution coefficient (K$_d$) greater than 10 l/g for Hg(II) uptake was given by rGO and the composites (Sreeprasad et al., 2011). By the NH$_4$OH reduction of FeSO$_4$ and GO, the metal ion induced self-assembly of α-FeOOH–rGO hydrogels which was effective in Cr(VI), Pb(II) and gasoline removal from polluted water (Cong et al., 2012). Magnetite Fe$_3$O$_4$/GO composite (M/GO) once has been used for the removal of different metal ions; Co(II), Pb(II), Ni(II) etc (Liu et al., 2011; Kireeti et al., 2016; Vuong Hoan et al., 2016). Comparatively to Fe$_3$O$_4$, M/GO was reported to have higher adsorption. Also, by magnetic separation, it can be separated and recovered. A SiO$_2$/graphene composite showed high efficiency towards Pb(II) ion with maximum adsorption capacity of 113.6 mg/g (Hao et al., 2012). The main interaction for the adsorption was believed to be the electrostatic interaction between metal cations and negative surface charge and/or electrons of the composite.

Microwave-assisted synthesizing method was used for Ni(II)

removal to a graphene-manganese dioxide (GNs/d-MnO$_2$) composite (Ren et al., 2011b). The adsorption capacity of Ni(II) onto the composite GNs/MnO$_2$ was 46.6 mg/g at room temperature and it was 1.5 and 15 times higher value than those of pure d-MnO$_2$ and GNs, respectively. Additionally, GNs/MnO$_2$ composite could be reused for 5 times with a recovery rate of 91%. Competitive adsorption isotherms of three divalent metals of Cu(II), Pb(II), and Cd(II) were examined on a magnetic GO, multiwalled carbon nanotubes (MWCNTs), and powered activated carbon (PAC) (Hur et al., 2015a). The adsorption could be described by the Langmuir isotherm model where the adsorption was highly dependent on pH. The maximum adsorption capacities of the adsorbents in the order of Pb(II) $>$ Cu(II) $>$ Cd(II) and were higher, while displaying the same as the degree of the electronegativity and the hydrated radius of the metals. Though antagonistic effects were affected by the type of metals and pH of the solution, they were higher in the order of the magnetic GO $>$ MWCNT $>$ PAC (Hur et al., 2015a).

Sulfonated magnetic graphene oxide composite (SMGO) was used to successfully remove Cu(II) from water (Hu et al., 2013) where the kinetic data fitted well with the pseudo second order while isothermal data also followed Langmuir equation. At 73.71 mg/L of Cu(II) concentration, 50 °C and 4.68 of pH, an optimum uptake of 62.73 mg/g was achieved. It was further found that the adsorption reaction was spontaneous and endothermic process from the thermodynamic parameters which were calculated from temperature dependent isotherms. Authors further advanced the synthesis and prepared magnetic graphene oxide–supported sulfanilic acid (MGO–SA) in aqueous solutions and investigated for Cd(II) with different background electrolyte concentrations (Hu et al., 2014). Results denoted that the Cd(II) adsorption was dependent on pH, background electrolytes as well as on ionic strength. For instance, at 0.01 mol l^{-1} Na$_3$PO$_4$ enhanced the Cd(II) removal when the pH > 5 while at pH < 5 it reduced the removal percentage. Not only the concentration but also the different electrolytes; NaNO$_3$, NaClO$_4$, NaCl and Na$_3$PO$_4$ influenced Cd(II) sorption. Freundlich model described the adsorption isotherm of Cd(II) onto MGO–SA in this case indicating a multilayer adsorption.

Magnetic Mn doped Fe(II) oxide nano particle implanted graphene (GMIO) has been used to investigate the sorption behaviors of Cu(II) and Cd(II) in aqueous solution by varying contact times, pH and concentrations (Nandi et al., 2013). With increasing temperature from 288 K to 333 K, monolayer sorption capacity values for the above cations of GMIO increased (130 mg/g to 144 mg/g for Cu and 88 mg/g to 127 mg/g for Cd). At optimized pH of 5.1 ± 0.1, kinetically the overall sorption process was governed by the pseudo-second order equation attributing a chemisorptions mechanism.

Polyethylenimine (PEI) modified magnetic mesoporous silica and graphene oxide (MMSP-GO) for the synergistic adsorption of heavy metal ions (Pb(II) and Cd (II)) and humic acid have also been studied (Wang et al., 2013). When magnetic mesoporous silica are being synthesized and functionalized with PEI molecules, many amine groups are available to conjugate with carboxyl groups on GO sheets which can enhance the affinity between the mesoporous silica and the pollutants. Promising data have been observed by MMSP-GO composites as adsorbents for efficient and simultaneous removal of Cd(II) and Pb(II) in wastewater treatment process where the maximum adsorption capacities of MMSP-GO shown for Cd(II) and Pb(II) were 167 and 333 mg/g, respectively.

Further for solidification and preconcentration of U(VI) ions from aqueous solutions, usage of magnetic graphene/iron oxides composite (Fe$_3$O$_4$/GO) have been studied (Zong et al., 2013). Ionic strength-independent and pH-dependent U(VI) sorption on Fe$_3$O$_4$/GO showed that at high pH values, U(VI) removal was achieved by inner-sphere surface complexation and simultaneous precipitation. At pH $= 5.5 \pm 0.1$ and T $= 293$ K the maximum sorption capacity of U(VI) on Fe$_3$O$_4$/GO was about 69.49 mg/g which was higher than the reported material and nanomaterials.

For the removal of Pb(II) and Hg(II) from aqueous solution,

Table 1
Metal/metalloid adsorption capacities on graphene materials.

Metal	Adsorbent	Adsorption (mg/g)	Temperature (°C)	References
As(III)	GNs	138	RT	(Mishra and Ramaprabhu, 2011)
Cd(II)	GO	14.9	RT	(Lee and Yang, 2012)
		106.3	30	(Zhao et al., 2011b)
Co(II)	GO	68.2	30	(Zhao et al., 2011b)
	Fe_3O_4/GO	13.0–22.7	30–70	(Liu et al., 2011)
Cr(VI)	GNs	43	20	(Wooálee and Binákim, 2011)
	ED-RGO	5	33	(Luo et al., 2012)
	Fe^0-GNs	162.6	20	(Wooálee and Binákim, 2011)
Cu(II)	GO	46.6	RT	(Yang et al., 2010)
	GO-gelatin/chitosan	120.0	RT	(Zhang et al., 2011a)
Hg(II)	GO	35	RT	(Gao et al., 2011)
	GO-SH	190	RT	(Gao et al., 2011)
	GNs- SH	22	RT	(Gao et al. 2011)
	Ag- GNs	10.9	30	(Sreeprasad et al., 2011)
	MnO_2- GNs	10.8	30	(Sreeprasad et al., 2011)
	GNs- polypyrrole	980	20	(Chandra and Kim, 2011)
Pb(II)	GO	367	25	(Madadrang et al., 2012)
	GO-EDTA	479	25	(Madadrang et al., 2012)
	GO-Chitosan	99	RT	(Zhang et al., 2011b)
	GNs	22.4	30	(Huang et al., 2011)
	GNs-EDTA	228	25	(Madadrang et al., 2012)
Zn(II)	GO	30.1	RT	(Lee and Yang, 2012)

RT- room temperature.

magnetic cobalt ferrite ($CoFe_2O_4$)-reduced graphene oxide (rGO) nanocomposites ($CoFe_2O_4$-rGO) have been developed recently as an adsorbent in a simple, safe and cost effective method (Zhang et al., 2014). The highest adsorption for Hg(II) were 157.9 mg/g at pH of 4.6 and 25 °C and Pb(II) were 299.4 mg/g at pH of 5.3 and 25 °C. In addition, the data fitting indicated an involvement of chemisorption mechanism for metal removal. Adsoprtion capacities of some of the metal/metalloid ions on GO, GNs and their composites are being summarized below (Table 1).

5. Removal of arsenic

Arsenic exists as arsenate and arsenite in contaminated water and it is one of the most carcinogenic and toxic ion present specially in groundwater in many parts of the world. Water dispersible magnetite was synthesized by the simultaneous reduction of GO, $FeCl_2$ and $FeCl_3$, Fe_3O_4–rGO composite with super paramagnetic nature in room temperature efficiently bound both As(III) and As(V) (Chandra et al., 2010). Synthesis of cross-linked GO with ferric hydroxide (GO-Fe) exhibited high arsenic removal from drinking water (Zhang et al., 2010). Adsorption capacity value of 23.8 mg arsenate/g-composite was shown by the composite GO-Fe-5 while in a range of pH 4–9.

Even with an increment of the pH range, effective As(III) and As(V) adsorption was observed due to the introduction of MnO_2 nano particles into a Fe_3O_4–rGO material (Luo et al., 2012). It acted as a promising application since MnO_2 acts as an oxidant for As(III) without the need for adding other oxidants for effective adsorption of As(V). Recently, another research group (Zhu et al., 2012) has been able to develop graphene decorated with core–shell Fe–Fe_2O_3 nanoparticles which took the advantage of Fe core which makes the material magnetically separable whereas the total adsorption sites was increased by the use of Fe_2O_3. Graphene decorated with core–shell Fe–Fe_2O_3 nanoparticles showed an efficient capacity of As(III) removal in water.

One-pot microwave route has also been introduced for the production of 3D graphene-carbon nanotube-iron oxide nanostructures that contained an open pore network with high surface to volume ratio in arsenic removal (Vadahanambi et al., 2013). Carbon nanotubes are being standing vertically on graphene sheets whereas iron oxide nanoparticles are decorated on both carbon nanotubes and graphene in this nanostructure. Three-dimensional Fe_3O_4/graphene aerogels (GAs) and their application for the removal of arsenic (As) ions from water

also demonstrated effective removal of As(V) from water (Ye et al., 2015). Iron oxide nanoparticle decorated graphene owed a high adsorption capacity of 40.048 mg/g

There were reported attempts to synthesize graphene sheets by hydrogen induced exfoliation of graphitic oxide which was followed by functionalization (Mishra and Ramaprabhu, 2011). High concentrations of As(III) and As(V) removal by the usage of supercapacitor based water filter was a novel finding (Mishra and Ramaprabhu, 2011). Maximum adsorption capacities for As(V) and As(III) using Langmuir isotherms were found out to be 142 and 139 mg/g, respectively. Simultaneously, it was applied to desalination of sea water.

6. Removal of chromium

Stable, reusable, cheap and easy to prepare Ni nanoparticles-rGO composite material has been introduced to experience inherent properties of graphene as one of the recent approaches (Bhowmik et al., 2014). The synthesized material was found to be active in the formic acid induced reduction of aqueous Cr(VI) at the temperature of 25 °C. Though the rate of reaction is slow in room temperature, it could be enhanced by the introduction of an intermediate redox step with formic acid when Ni-rGO was present.

A highly reductive superparamagnetic graphene/Fe_3O_4 composite demonstrated efficient removal of Cr(VI) from waste water with a maximum adsorption capacity of 31.8 mg/g, which is greater than the graphene/γ-Fe_2O_3 composite (Ting et al., 2016). The possible mechanism for the Cr(VI) removal was observed by XPS and give as reduction of Cr(VI) to Cr(III) by ferrous hydroxide moieties in the graphene structure and resulted Cr(III) ions were easily captured by the negatively charged magnetic graphene surface. In addition, superparamagnetic graphene/Fe_3O_4 composite possessed a large surface area, negative charge and superior magnetism which could be applied to remove Pb(II), Cu(II), and Zn(II) with an efficiency of almost 100% (Ting et al., 2016).

By refluxing GO with ethylenediamine (ED) using dimethyl formamide (DMF) as solvent has been used to form chemically reduced functionalized GO (Zhang et al., 2013a). Cr(VI) adsorption as well as the subsequent indirect reduction mechanism of adsorbed (VI) to Cr (III) contributed the Cr(VI) removal in this case. The total Cr removal was observed at the optimum pH value of 2 where the resulting adsorbent (ED-DMF-RGO) showed 92 mg/g of Cr(VI) removal capacity at
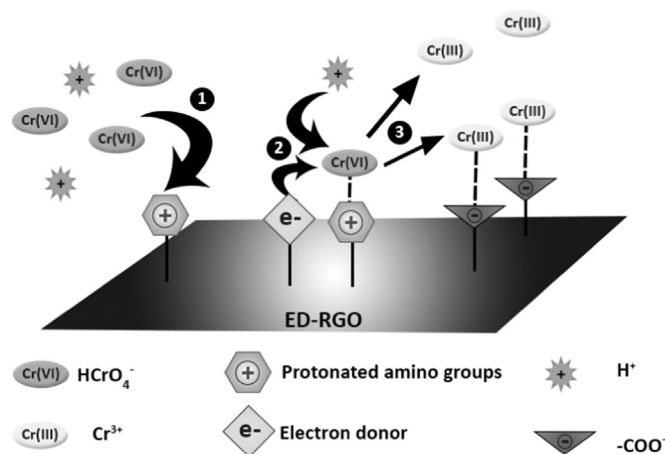
Fig. 1. Proposed mechanism of Cr (VI) removal by ED–RGO.

this particular pH. Ethylenediamine–reduced GO (ED–RGO) sheets have been also used for the effective removal of Cr(VI) (Luo et al., 2012). It was noted that Cr(VI) removal was through an indirect reduction mechanism with the help of p electrons on the carbocyclic six membered ring of ED–RGO. Mean while, part of Cr(III) species were bound to ED–RGO surface (Fig. 1).

Even via sodium borohydride reduction of GO, successful efforts were reported in synthesizing nanoscale iron decorated GNs (Wooálee and Binákim, 2011). Comparatively to bare iron nano particles, this material showed an enhanced adsorption capacity of Cr (VI) with improved surface area and magnetic property. Reported magnetic graphene nano composites (MGNCs) with a core@double-shell structure (Zhu et al., 2011) demonstrated fast removal of Cr(VI) along with a high removal efficiency. At conditions of [Cr(VI)] = 1000 g/L, [MGNCs] = 1 g/L and pH 7, complete Cr(VI) removal was occurred within 5 min.

A systematic investigation was carried out as a chemical bonding method to investigate the adsorption behavior of Cr(VI) on magnetic cyclodextrin-chitosan/graphene oxide (CCGO) which contained abundant amino and hydroxyl groups (Li et al., 2013). The adsorption equilibrium of CCGO followed Langmuir isotherm. A strong dependency of specific surface area and surface charge concentration were indicated on the adsorption performance of the novel adsorbent on Cr(VI) removal while exhibiting a better removal efficiency in solutions with low pH.

Apart from that, sacrificial template polymerization method has been utilized for enhanced Cr(VI) removal in aqueous solution by the synthesis of polypyrrole (PPy)/graphene oxide (GO) composite nanosheets where for the deposition of PPy, MnO_2 were chosen as a sacrificial template (Li et al., 2012d). It was found that the adsorption capacity of PPy/GO composite nanosheets were about two times as large when compared to conventional PPy nanoparticles. The method of fabrication was easy because of its independency in removing template than the conventional route.

7. Removal of other ions

There are different ions such as NO_3^-, SO_4^{2-}, ClO_4^-, PO_4^{3-} and F^- which are present in wastewater in excessive amounts. A 3D nanostructured graphene–polypyrrole (Ppy) composite was reported for ClO_4^- purification (Zhang et al., 2011b) where the particular nano composite exhibited an improved uptake capacity ClO_4^- comparatively to Ppy film individually. Reported maximum adsorption capacity was 89.4 mg/g and this adsorption was chemically irreversible and spontaneous. Efficient ClO_4^- removal (99%), 0.024 mg/g at initial perchlorate concentration of 2 mg/L and temperature of 298 K, was observed by using graphene indicating chemisorption mechanism

(Lakshmi and Vasudevan, 2013).

Usage of graphene prepared by liquid phase exfoliation of graphite exhibited an effective removal of phosphate ions due to the introduction of functional groups onto the surfaces of graphene enhanced phosphate adsorption (Vasudevan and Lakshmi, 2012). Green hydrothermal zirconium-loaded reduced graphene oxide (RGO-Zr) indicated high phosphate adsorption capacity of 27.71 mg/g at pH 5 and 298 K (Luo et al., 2016). The improved phosphate adsorption was due to the dispersion of ZrO_2 on the RGO surface. Interestingly, phosphate adsorption was found insensitive to pH increase but sensitive to the increase and vice versa for temperature. Phosphate sorption was not influenced by the presence of Cl^-, NO_3^- due to the weaker outer sphere complex formation by Cl^- and NO_3^-, however, SO_4^{2-}, CO_3^{2-} lowered the phosphate adsorption since SO_4^{2-}, CO_3^{2-}, HPO_4^{2-} form both outer and inner sphere complexes with the active surface sites. However, only high F^- could enhance the phosphate adsorption which could be caused due to the chemical reactions between ZrO_2 and F^- (Luo et al., 2016).

Graphene has been an excellent nitrate (NO_3^-) adsorbent with an adsorption capacity of up to 89.97 mg/g in chemisorptive nature at an initial NO_3^- concentration of 500 mg/L and temperature of 303 K (Ganesan et al., 2013). A drastic increase in the efficiency of nitrate removal from water is achieved over magnetically functionalized metal functionalized graphene; Fe@r-GO, Ni@r-GO, and Co@r-GO (Motamedi et al., 2014).

Similarly, high fluoride removal was observed from exfoliated graphene with monolayer adsorption capacity of 35.6 mg/g at 298 K and pH 7 (Li et al., 2011b). These studies started an effective adsorbent quality of graphene through an irreversible process. The porous and low-density 3-D rGO aerogel showed excellent removal capabilities for F^- where the adsorption data revealed the mechanism as chemisorptions with a maximum adsorption capability of 31.3 mg/g, indicating that the 3-D RGO aerogel is a suitable material for fluoride removal in water (Wu et al., 2015).

8. Removal of organic pollutants

Successful applications of graphene based materials in organic pollutants adsorption have been applied to different forms of polycyclic aromatic hydrocarbons, gasoline and dyes.

9. Dye removal from graphene based materials

The ability to test GO for removal of various dyes were reviewed extensively focusing the catalytic and degradation activities (Upadhyay et al., 2014). Among many different graphene composites, TiO_2-graphene composite have been widely used for photodegradation of dyes. At the same time, many metals and metal oxides composites were tested for their abilities in photocatalytic degradation of dyes. Among them, methylene blue was the most common dye tested for its degradation followed by rhodamine B (Upadhyay et al., 2014). Not only photocatalytic degradation but also dye adsorption has been investigated by various graphene materials (Sun et al., 2014; Konicki et al., 2017; Jayanthi et al., 2016). Highly porous, ultralight graphene oxide foams were applied for carcinogeneic dyes, rhodamine B (RB), malachite green (MG) and acriflavine (AF) where high removal capacities were observed; 446, 321 and 228 mg/g, respectively (Jayanthi et al., 2016). The particular 3-D GO was synthesized by a facile and cost effective lyophilization technique and the direct use of the foam without any pretreatment such as ultrasonication was possible due the 3D architecture which is one advantage in terms of practical application. Simultaneously, these foams were also exhibited high antibacterial activities against E. coli bacteria in aqueous and nutrient growth media which again indicates its potential to be used in water treatment (Jayanthi et al., 2016). Methylene blue (MB) and malachite green (MG), in aqueous solution has been reported via electrostatic attraction and

the reported adsorption capacities of MB and MG were 351 and 248 mg/g, respectively (Bradder et al., 2010). These values were seemed to be much higher than those of activated carbon and graphite. Even other research groups also discovered the capacity of GO as an adsorbent for MB with adsorption capacity of 714 mg/g (Yang et al., 2011a) and 1939 mg/g (Zhang et al., 2011).

A maximum adsorption capacity of 153.8 mg/g has been observed by GN for adsorption of MB at 20 °C (Liu et al., 2012c). In this particular study, the effects of temperature, contact time, pH and dosage on adsorption properties of MB were also investigated. Kinetic study showed that it was fitting to pseudo second order model. Better than Freundlich model, the isotherm of experimental data followed Langmuir model while the thermodynamic parameters indicated that MB adsorption was spontaneous and an endothermic process. In situ reduction of GO with sodium hydrosulfite to graphene was able to enhance adsorption of acridine orange (AO) (Sun et al., 2012) with a maximum adsorption capacity of 1.4 and 3.3 g/g for GO and rGO respectively, under identical conditions.

Triphenylmethane (TPM) dye is considered as one of the most prevalent and recalcitrant water contaminants in the wastewater. Magnetic Fe3O4@rGO nano-composite showed quick and high adsorption capacity towards different TPM dyes with 64.93 mg/g adsorption for crystal violet (Sun et al., 2014). Successful attempts were made to produce a $Mg(OH)_2$–rGO by using NaOH in the simultaneous reduction of $MgCl_2$ and GO (Li et al., 2011a). Adsorption capacity of Methyl Orange (MO) increased 12 times with the inclusion of rGO into CdS nanoparticles (Pan and Liu, 2012). It was revealed that the 3D materials displayed controllable desorption of pollutants and they are more efficient pollutant absorbers than 2D graphene materials (Tiwari et al., 2013).

Many composites of GO and GNs were found to be effective for dye removal (Gong et al., 2017; Liu et al., 2012a; Kharismadewi et al., 2016). Methyl orange removal by GO nanosheets was 138.69 mg/g at pH 2.0, which is considered to be larger than that of other carbonaceous adsorbents (Gong et al., 2017) due to the presence of hydrogen bonding and π–π interaction between methyl orange and graphene oxide nanosheets. Batch experiment data modeling indicated the presence of both chemisorption and physisorption for the removal of MO (Gong et al., 2017). The GO/Fe3O4 nanohybrids demonstrated a 100% removal for methylene blue while a remarkably high removal for Rhodamine B (Jiao et al., 2015). A biocompatible and thermoresponsive polymer was prepared by grafting poly(2-hydroxyethyl methacrylate) (PHEMA) to GO surface and examined for the removal of MB (Kharismadewi et al., 2016). Adsorption observed to be an interaction of chemisorptions and physisorption with a capacity of 39.41 mg/g at 25 °C and could remove 99.8% of the dye in 45 min (Kharismadewi et al., 2016). Fabrication of carbon nanotube (MWCNT)–graphene hybrid aerogel was reported to have adsorption capacities of 191.0, 180.8, 150.2 and 35.8 mg/g for MB, Fuchsine, Rhodamine B (RB) and acid fuchsine respectively (Sui et al., 2012). A biocompatible and hydrophilic 3D chitosan–graphene mesostructures were used for removal of reactive black 5 (RB5) from aqueous solution with 97.5% of removal efficiency at initial RB5 1.0 mg/mL concentration (Cheng et al., 2012).

Reduced graphene oxide-titanate (rGO- Ti) hybrids with higher pore volumes and large surface area were reported to have been fabricated where the MB uptake reached 83.3 mg/g over the hybrid in comparison with tubular titanates and pure GNs (values of 36.5 and 48.7 mg/g respectively) (Nguyen-Phan et al., 2012). A magnetic chitosan–rGO composite material has improved MB sorption by the electrostatic mechanism between MB and the charged amino acid groups (Fan et al., 2012).

In a similar manner, various groups have reported about adsorption capacities of iron containing and magnetic graphene composite materials (Sun et al., 2011; Ai et al., 2011; Yao et al., 2012a; Guo et al., 2012; Yang et al., 2012). A synthesis of graphene nanosheet (GNS)/ Fe3O4 composite by a one step solvothermal method have been

demonstrated (Ai et al., 2011). It was achieved by in situ conversion of FeCl3 to Fe3O4 and simultaneous reduction of GO into graphene in ethylene glycol solution. The resulting composite has shown fast adsorption rates for MB removal. Meanwhile the equilibrium adsorptions and kinetics could be described by Langmuir model and pseudo second order kinetics respectively. Also, magnetic Fe3O4/graphene composite (FGC) have been reported (Yao et al., 2012a) for MB and Congo red (CR). The maximum adsorption capacities obtained were 45.3 and 33.7 mg/g, for MB and CR respectively. GO as a supporting matrix has been also been reported to synthesize Fe nanoparticles@graphene composites (FGC) (Guo et al., 2012) First, GO was treated with Fe^{3+} to get Fe^{3+}@GO complexes. GO and Fe^{3+} are reduced in situ to graphene and Fe respectively by adding NaBH(4) solution while forming FGC hybrid composites. They have high MB removal capacities due to the increased adsorption sites and it was achieved by the reduction of the size of Fe nanoparticles and inhibition of particle aggregation.

Quick and high removal efficiency of fuchsine dye was observed from a magnetic graphene nano composite (Wang et al., 2011). Even several researches have been conducted in the removal of dyes using 3D graphene-based materials too as they can be easily separated from the solution (Liu et al., 2012a; Niu et al., 2012). A superparamagnetic GO–Fe3O4 composite (GO–Fe3O4) was tested for MB and neutral red (NR) which showed fast removal of only 30 min and 90 min for MB and NR respectively and the reported adsorption capacities for MB and NR were 167.2 and 171.3 mg/g, respectively (Xie et al., 2012). The magnetic Ni–rGO nano composite prepared using the simultaneous hydrazine reduction of $NiCl_2$ and GO showed a completed adsorption of organic dyes RB and MB from water within 4 h (Li et al., 2012a). Adsoprtion capacities of some of the dyes on GO, GNs and their composites are being summarized below (Table 1) (Table 2).

Again, GO was examined for the adsorption of anionic azo-dyes such as Acid Orange 8 (AO8) and Direct Red 23 (DR23) indicated chemisorption and diffusion as mechanisms for the removal (Konicki et al., 2017). The hydrophobic nature hinders the possibility of submerge the graphene sponge into water though it has attracted a strong interest. To overcome the particular issue, graphene–melamine-sponge (GMS) was invented with a superhydrophilic melamine skeleton and investigated its adsorption behaviors for methylene blue (MB) and orange G (OG) (Du et al., 2016). The GMS efficiently adsorbed MB and OG with competitive capacities of 286.5 and 80.51 mg/g, respectively indicating monolayer chemisorptions (Du et al., 2016).

10. Aromatic compounds and gasoline

Extended studies using GO and GNs for adsorption of phenolic compounds, drugs, pesticides and humic acids have shown promising results (Wang et al., 2013). Generally, the interactions with hydrogen bonds, hydrophobic effect, π-π bonds, electrostatic and covalent bonds have been identified in carbon materials. These are believed to be playing a major role in the adsorption of organic chemicals of nano sized carbon particles (Wang et al., 2012a). It was found that the GO exhibited higher adsorption capacity compared to graphite for humic acid (HA) (Hartono et al., 2009). From the Langmuir isotherm, maximum adsorption capacity of the GO was 190 mg/g, which was even higher than reported different values of activated carbon.

Ni–rGO nano composite material was used to remove organic pollutants such as toluene, benzene and dimethylbenzene from a solution (Li et al., 2012c). It can be separated magnetically from water and easily recycled is an added advantage to be used in water treatment. The use of 3D graphene-based materials as adsorbents where the material formation was done by a GO self-assembly process have been successfully applied in gasoline removal (Cong et al., 2012). Synthesis of superhydrophobic rGO-coated porous scaffolds by a melamine sponge which was dipped into graphene dispersed ethanol exhibited high removal of organic solvents and oils (Nguyen et al., 2012). Monoliths of graphene and CNTs in 3D foams which can effectively

Table 2
Dye adsorption capacities on some graphene materials.

Dye	Adsorbent	Adsorption (mg/g)	Temperature (°C)	References
MB	Fe_3O_4-GO	167.2	25	(Xie et al., 2012)
	GNs	154–204	20–60	(Liu et al., 2012c)
	G-aerogel	152.8	RT	(Sui et al., 2012)
	Fe_3O_4-GNs	43.8	25	(Ai et al., 2011)
	$MnFe_2O_4$- GNs	34.7	25	(Bai et al., 2012)
MG	Fe_3O_4-GNs	22	RT	(Sun et al., 2011)
MO	$CoFe_2O_4$- GNs	71.5	–	(Li et al., 2011c)
Methyl Blue	GO	43.5	30	(Fan et al., 2012)
	Fe-GNs	75–375	RT	(Guo et al., 2012)
AO	GO	1428	–	(Sun et al., 2012)
	GNs	3333	–	(Sun et al., 2012)
RB	G-aerogel	133.6	RT	(Sui et al., 2012)
	Fe_3O_4-GNs	13.2	RT	(Sun et al., 2011)
Fuchsine	G-aerogel	142.7	RT	(Sui et al., 2012)
NR	Fe_3O_4-GO	171.3	25	(Xie et al., 2012)
CR	Fe_3O_4-GNs	33.7	–	(Yao et al., 2012a)

*RT- room temperature.

retain adsorbents have a large internal area and showed a functional similarity to rGO-coated melamine sponges (Dong et al., 2012).

Pharmaceutical traces have been classified as emerging pollutants in the aquatic ecosystem due to their persistence, therefore, its removal is essential and graphene plays a major role in sorptive removal of pharmaceuticals in water (Carmalin Sophia et al., 2016). The adsorption of some specific antibiotics on GO was reported high maximum adsorption capacities of 212, 313 and 398 mg/g for oxytetracycline, tetracycline and doxycycline respectively attributed to cation-π bonding and π-π interaction (Gao et al., 2012). Sulfapyridine and sulfathiazole removal by two reduced graphene oxides were examined at the influence on pH and Dissolved Organic Carbon (DOM). Adsorption of sufonamides (SAs) by rGOs was highly pH and speciation dependent and the responsible mechanisms for SAs removal were suggested as hydrophobic interaction, π–π EDA interaction and electrostatic interaction (Liu et al., 2016). Interestingly, the inhibition impact of DOM on was low for rGOs than carbon nano tubes and graphite, which might be due to higher oxygen contents in rGOs (Liu et al., 2016).

Ciprofloxacin was chosen to compare the removal capacities of GO, commercial graphene and TiO_2 reduced graphene and the maximum adsorption capacities were seemed better in graphene and TiO_2 reduced graphene than the commercial graphene, 178.6, 181.8, and 108.7 mg/g respectively (Zhuang et al., 2015). Tetracycline strongly loads on the GO surface via π-π interaction and cation–π bonding in chemisorptions bonding with a maximum adsorption of 323 mg/g (298 K) (Ghadim et al., 2013). A complex strontium titanium trioxide doped graphene oxide based magnetic nanocomposite showed high adsorption capacities of 65.78 and 18.21 mg/g toward tetracycline and cefotaxime, respectively (Rashidi Nodeh and Sereshti, 2016). The π-π interactions between aromatic rings on pharmaceuticals and graphene materials have been proved as the main adsorption mechanism (Peng et al., 2016).

A graphene-coated biochar material was utilized as an adsorbent for polyaromatic hydrocarbons where the adsorption capacity of biochar could be increased by 20 times with a coating of graphene (Zhang et al., 2012a). Sulfonated graphene (thickness around 3 nm) sheets have been tested for its capability to adsorb naphthalene (~2.3–2.4 mmol/g) with strong π-π interaction between the aromatic molecules and graphene sheets indicating the utilization as an effective adsorbent in pollution management of the near future (Zhao et al., 2011a). The effects of contact time, pH, temperature and dosage on phenol adsorption using GNs showed a maximum adsorption capacity of 28.26 mg/g (Li et al., 2012b). Further, the adsorption of bisphenol A (BPA) on GNs reported a maximum adsorption of 182 mg/g via hydrogen bonds, single atom layer which was sp^2 hybridized and π-π interaction (Xu et al., 2012).

Adsorption of pesticides such as malathion (ML), endosulfan (ES), and chlorpyrifos (CP) on to GO and GN reported a high removal capacities for GN with 800, 1100, and 1200 mg/g were found for ML, ES and CP respectively (Maliyekkal et al., 2013). The adsorption of phenanthrene, onto the loaded graphene nanosheets exhibited an increase up to 100 fold compared with pristine graphene at the same level (Yang et al., 2015). More than 70% adsorption capacity was reported for 6 different Triazine pesticides with cellulose-graphene composite and it was the best among five other sorbents (graphite carbons, primary secondary amine (PSA), graphite carbon black (GCB), cellulose, and graphene) (Zhang et al., 2015). Again, Fe_3O_4/reduced graphene oxide (rGO) nanocomposite efficiently employed to remove five harmful pesticides namely ametryn, prometryn, simazine, simeton and atrazine where the removal mechanism was identified as electrostatic interaction between the pesticides and $Fe3O4$/rGO nanocomposite through chemisprotion (Boruah et al., 2017) with a maximum adsorption capacity of 54.8 mg/g at pH 5. The presence of Mg^{2+}, Ca^{2+}, Na^+ and SO_4^{2-} enhanced sorption and maximum (63.7 mg/g) for ametryn adsorption was found in seawater medium (Boruah et al., 2017).

Dibenzothiophene (DBT) in fuel resulted environmental and industrial problems due to the presence of sulfur compounds (Nazal et al., 2015). The potential of synthesized GO, commercial coconut activated carbon (AC) and MWCNT loaded with 5% and 10.9% aluminum (Al) in the form of aluminum oxide (Al_2O_3) particles were tested to improve the chemical properties of their surface demonstrated a highest adsorption capacity for DBT (29 mg/g) with 5% Al (Nazal et al., 2015). Benzene, aniline and naphthylamine adsorption on rGOs were investigated by batch experiments and theoretical density functional theory (DFT) indicated that the sequence of maximum adsorption capacity is naphthylamine > aniline > benzene on rGOs (Yu et al., 2016a).

11. Recent advancements and future outlook

The GO and GNs are the new carbonaceous materials that have been used as adsorbents to remove various pollutants in water. Due to loss of oxygen species, the π-π interaction between the adsorbate and GN plays a vital role for adsorption on many different organic contaminants including pharmaceuticals. On the contrary, GO depicts strong acidity and negative charges where it is able to exhibit high adsorption for inorganic ions together with heavy metals. Because of strong interplanar interaction, there is a tendency to aggregate in a layer by layer mode in GO and GNs. As a result, GO and GNs can be functionalized to improve the adsorption capacity by forming composites. Particularly, magnetic GO/GNs nano composites promote convenient magnetic

separation along with the reusability of the adsorbents. Yet, the large scale production of graphene with a low cost without using toxic chemicals is being viewed as a daunting challenge to the scientists. There were suggested green methods to synthesize graphene material from cane sugar (Gupta et al., 2012; Parvathi et al., 2015). With proper engineering, such green approaches which are quick and cost effective must be adhered into water filtration.

Environmental friendly nature of graphene with large surface area and high stability has paved the way even to environmental applications especially water decontamination according to this review article. As a result, graphene and its composites have been applied by various research groups in this perspective. Usage of porous GO foams to design unique 'tea bags' by University of Aveiro, Portugal as a purification method for the removal of mercury and usage of graphene which is an allotrope of graphene for desalination applications can be viewed as few of novel research attempts which had opened new horizons in the magnitude of graphene's application very recently. Graphene's hydrophobic nature is one of the remarkable traits that can be useful in water remediation. When narrow pores are made in graphene, rapid water permeation can be seen although graphene naturally repels water. Once the production of micro pores by research attempts are fruitful, there will be an increased usage of graphene in water filtration and desalination areas as well.

The experiments to date demonstrate that the water remediation aspect of graphene is based on the environmental conditions hence, future research is essential to make graphene into a good universal material for water treatment. At the same time, different test methods, synthesis techniques and functionalization have produced hundreds of graphene materials, and tested for many different contaminants in different ranges in various environmental conditions, which does not allow finding the best material. Hence, it is important to standardize the laboratory techniques in order to compare the results and investigate the best material among many.

Most research carried to examine the capacity of graphene and related materials for water treatment in literature is limited to batch adsorption studies and hence research on column and prototypes are recommended. The evaluation and analysis of graphene materials life cycle, impacts, toxicity and distribution in the different ecosystem should also be understood well since it will be finally released into the environment. Finally, recent optimistic approaches should be motivated with proper regulations of environmental policies in sustainable wastewater management systems in which graphene would be able to manufacture in an industrial scale to address certain environmental issues. Hence, the scope of graphene application in water decontamination seems to be still evolving into different calibers of its capacity with recent developments.

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