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Recent advances in the treatment of contaminated soils by ball milling technology: Classification, mechanisms, and applications

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ABSTRACT

Soil pollution has become an important environmental issue worldwide. Various pollutants in soils may harm human health through the food chain. At present, the commonly used remediated methods mainly include thermal treatment, electrokinetic remediation, stabilization/solidification, soil washing, and bioremediation. Among them, ball milling is a non-combustion technology with simple operation, green, and mild reaction conditions as well as wide applicability, and it is promising in the decontamination of soil pollutants. Ball milling techniques have been gradually used for the detoxification of persistent organic pollutants and heavy metals in contaminated soils, while related reviews focusing on ball milling for soil remediation are limited. It is necessary to fully understand the potential and applied value of ball milling for remediation of contaminated soils. Ball milling has different effects and mechanisms for different pollutants and soil types. Therefore, there is a need to systematically review and digest the relevant knowledge on ball milling remediation of contaminated soils. In this review, the main types of ball milling and influencing factors are summarized, and the mechanisms of ball milling for soil remediation are analyzed. Finally, the problems which need to be solved are briefly described and the future research direction and trend are pointed out. This review is expected to promote the application and its in-depth research of ball milling technology on soil remediation.

1. Introduction

With the increasing urbanization, a large amount of soil and water environments have been seriously contaminated (Chen et al., 2020; Feng et al., 2022; Lian et al., 2019). According to the Food and Agriculture Organization of the United Nations, only 12.6% of the world's land is available for agricultural activities (FAO and ITPS, 2015). While agricultural production and food safety are seriously threatened after various pollutants (e.g., heavy metals, polycyclic aromatic hydrocarbons, and pesticides) enter the soil (Pepper, 2013). The above processes may affect human health, leading to various diseases and immunocompromise. With the increasing negative impacts of soil pollution on human health, identifying and refining soil remediation technologies becomes even more critical.

To solve the severe soil pollution problem, there is a great need to develop effective soil remediation technologies, so that contaminants can be effectively decontaminated. In recent years, several treatment techniques have been used to remediate contaminated soils, including thermal treatment (Vidonish et al., 2016), electrokinetic remediation (Rosestolato et al., 2015), stabilization/solidification (Bian et al., 2013, 2014; Sun et al., 2021), soil washing (Andreozzi et al., 2020; Ferraro et al., 2016a; Yun et al., 2015), and bioremediation (Wang et al., 2018). Among them, the high energy consumption of thermal treatment causes relatively high costs, and high temperatures may also lead to changes in

soil function and even deteriorate soil physicochemical properties (Gong et al., 2018). Electrokinetic remediation is generally applied to low permeable porous matrices, leading to the removal of contaminants through mechanisms such as electromigration, electroosmosis, and electrophoresis, but its side effects result in high energy consumption and low mass transfer rates (Wen et al., 2021). Although stabilization/solidification and soil washing are high efficiencies, low-risk, and possible for the recovery of metals, the longevity of the stabilization/solidification materials and the safe disposal of the washing solution may need to be pondered (Bian et al., 2016; Ferraro et al., 2015, 2016b; Race et al., 2016). Bioremediation is often rather slow and greatly influenced by environmental variables (Gong et al., 2018). Since the above-mentioned methods have a series of disadvantages and cannot completely solve the problem of soil pollution, an efficient and cost-effective remediation technology is highly needed to develop.

Mechanochemical treatment is characterized by easy operation, high efficiency of treatment, and mild reaction conditions, which is considered as a green soil remediation method (Montinaro et al., 2007), especially it can remediate heavy metals and organic contaminated soils simultaneously (Aresta et al., 2003; Concas et al., 2007). Under certain working conditions, high-energy grinding can limit the toxicity of contaminated substances or reduce their solubility to achieve the remediation of soils (Concas et al., 2020b). Mechanochemical treatment refers to the materials in the milling jar under the constant collision of the grinding body (generally a ball), grinding, impact or shearing, and other effects occur to obtain a large amount of mechanical energy, which initiates the solid-state reaction (Cagnetta et al., 2018; Gilman, 1996). Mechanochemical activation enables inorganic or organic pollutants to interact with soil components, accompanied by crystal structure destruction, frictional wear, and fracture of soil particles (Friščić, 2018; Plescia et al., 2003). The above processes may lead to piezoelectric effects, surface plasma formation, high-energy emission of electrons, neutral particles or photons, and functional groups generation (Friščić, 2018; Wang et al., 2018a, 2018b). Generally, inorganic contaminants are irreversibly adsorbed and entrapped into soil particles (Concas et al., 2007). Organic pollutants are mineralized to amorphous carbon, graphite, and a mixture of light hydrocarbons, while halogen elements are formed inorganic halides (Cagnetta et al., 2016).

Ball milling, as a typical mechanochemical method, has been initially used for the effective removal of persistent organic pollutants (POPs) (Rowlands et al., 1994; Zhang et al., 2001). A high density of energy is supplied to the contaminated soil by countless collisions or shear to satisfy the mechanochemical reactions. The energy supplied to the particular soil/reactant/pollutant system can promote different physic-chemical transformations (Cagnetta et al., 2016; Concas et al., 2020b; Montinaro et al., 2007, 2009), which may lead to the effective immobilization or degradation of various pollutants. Compared with other methods, ball milling has been successful in treating many types of contaminated soils, such as polychlorinated biphenyls (PCBs), organic bromide, per- and polyfluoroalkyl substances, and heavy metals (e.g., Pb, Zn, and Cr) (Montinaro et al., 2008; Turner et al., 2021; Wang et al., 2017; Yi et al., 2021; Yuan et al., 2019a; Zhang et al., 2014, 2020). Compared with traditional techniques, ball milling is a non-combustion treatment with few technological processes and no solvent, gas, and temperature requirements (Qin et al., 2020; Wang et al., 2019; Zhao et al., 2022). Searching through the Web of Science with the keywords of "ball mill" and "soil", it was noted that the remediation of soil using ball milling has been gaining increasing attention (Fig. 1). The functions of ball milling are diverse, such as the ability to treat various classes of pure waste chemicals or solid waste, and the remediation of contaminated soil (Turner et al., 2021). Moreover, compared with conventional technologies for soil remediation, the ball milling method is relatively energy-saving. The ball milling method mainly relies on ball mill jar rotation to produce a centripetal force to drive the grinding ball movement (Gao et al., 2021; Jiang et al., 2009), and realize the remediation by milling the soil through countless hits. In brief, ball milling can be rapidly and simply applied, enabling economical, efficient, and green soil remediation effects.

There have been some studies on the application of mechanochemical methods (Cagnetta et al., 2016, 2018; Nasser and Mingelgrin, 2012), but the conclusion is relatively scattered. The ball milling method for soil remediation is also rarely reported. As far as we know, the mechanism of ball-milled is still unclear, thus a comprehensive comparison of the applicability and effectiveness of ball milling is highly necessary. Therefore, a systematic review of ball milling for soil remediation can provide the latest knowledge in this field. Based on the characteristics of ball milling, the important types of ball milling and the influencing factors of soil remediation are reviewed. Finally, the remediation
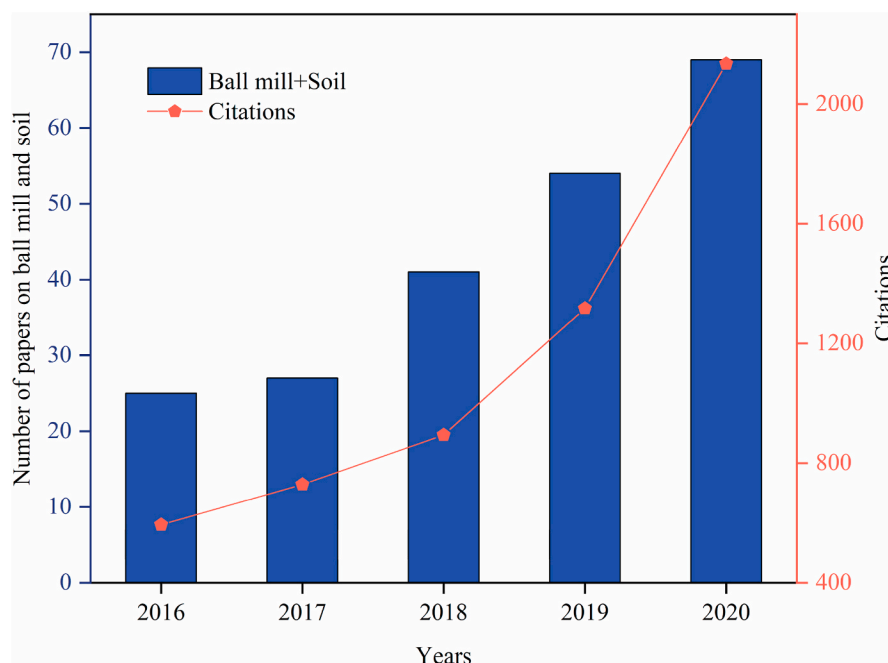

Fig. 1. The number of publications and citations of literature with the keywords of "ball mill" and "soil" (Data from Web of Science).

mechanisms are summarized, and the advantages and future directions of ball milling in this field are also analyzed.

2. Ball milling type

Recently, many studies have optimized ball milling techniques, including type, material quality improvement, technology parameter optimization, and optimization control of their processes. In the common ball milling process, the accumulated mechanical energy in a solid feed is often taken as the basic quantity to study the physicochemical transformations therein. Because ball milling has different types, discrete media (grinding balls) driven by jars transfer energy to the grinding feed in different ways (Baláž, 2008). Although raising the ball milling intensity can transfer more energy, not raising the ball milling intensity can also effectively remediate soil in an appropriate time (Tables 1 and 2). After the ball milling strength was boosted, the time to reach mechanochemical activation was not significantly shortened. Santhanam et al. simulations have shown that the key to the occurrence of a mechanochemical reaction is the energy delivered to the milled material, not the type of ball milling (Santhanam and Dreizin, 2012). Hence, ball milling only needs a reasonable time and milling strength to effectively remove soil contaminants.

The ball milling technique enables the remediation of contaminated soils by applying impact and shear effects to soil particles, inducing

chemical reactions or disrupting its lattice organization, altering its physicochemical properties (Baláž, 2008). After being applied with mechanical energy, soil triggers physical changes such as deformation, fracture, cracking, and volume loss, while its internal crystal structure, physicochemical properties, and reaction activities change to a corresponding extent. Generally, different types such as horizontal, stirring, and planetary were used for mechanochemical activation (Tan and Li, 2015) (Fig. 2).

The type of ball mill affects the efficiency of soil remediation, which is mostly planetary and horizontal. In previous studies, planetary ball mill has been widely applied due to its excellent applicability and effectiveness (Tables 1 and 2). Planetary ball mill by combining revolution and autorotation, which delivers activation energy to the material, which can be reduced to a few microns (He et al., 2020). While horizontal ball mill is simpler, the grinding jar is horizontally placed and rotated to bring the grinding balls rolling inside the jar to remediate contaminated soil. Previous studies have confirmed that the reaction rate of a planetary ball mill is faster than that of a horizontal ball mill (Cagnetta et al., 2018). Consequently, the ball milling time (BMT) for solid materials to get equivalent energy is shorter. Planetary ball milling increases the energy density by revolution, but its energy utilization is low and mechanical energy is not delivered efficiently (Gao et al., 2021; He et al., 2020). For scale-up applications, the jars of planetary ball mill must have led to elevated energy consumption due to volume and mass

Table 1
Technical parameters and conditions of ball milling for different inorganic pollutants and soil characteristics.

Pollutants (Concentration (mg kg^{-1}))	Soil characteristics (wt%)	Ball mill conditions						Effects (Leaching concentrations (mg L^{-1}))	References
		Ball mill type	Reagents (reagent–to–soil weight ratio (wt %))	Ball–to–soil weight ratio	Rotation speed (rpm)	Ball milling time (h)	Grinding ball texture, number, and diameter (mm)		
Pb (10439.35)	–	Planetary ball mill (Fritsch Pulverisette 7)	Ca(H$_2$PO$_4$)$_2$ (10)	14	550	2	Zirconia (20)	4.36	Zhang et al. (2020)
Pb (8975.58) Cu (4225.18) Zn (3787.31)	–	Planetary ball mill (Fritsch Pulverisette 7)	–	12	550	2	(15)	Pb/Cu/Zn 0.027/0.59/0.16	Yuan et al. (2019b)
Cr(VI) (2360)	–	Planetary ball mill (Retsch PM100)	Ca(S$_x$) (3)	9	500	2	(20)	0.51	Yuan et al. (2019a)
Cr(VI) (13806.4)	–	Planetary ball mill (FRITSCH P7)	Na$_2$S (5)	14	500	2	(20)	0.84	Yuan et al. (2018)
Cs (200)	SiO$_2$/Al$_2$O$_3$/ Na$_2$O/Fe$_2$O$_3$/ CaO/TiO$_2$/MgO/ OM 74.1/13.0/3.2/ 2.7/1.7/0.86/ 0.20/0.16/4.08	Planetary ball mill (Retsch PM–100)	Nano-metallic Ca/ CaO–NaH$_2$PO$_4$ 1-1 (5)	6.1	500	1	Stainless steel 20 (10)	0.004	Mallampati et al. (2012)
^{134}Cs/^{137}Cs (6201 Bq• kg^{-1})	Mica/Fibrolite	Tumbling ball mill	Nano-Fe/Ca/CaO/ [NaH$_2$PO$_4$] 11/11/28/50 (10)	–	100–150	2	Ceramic 10 (10)	n.d. [d]	Mallampati et al. (2015)
As/Hg/Pb/Se (221.0/144.2/ 17582.9/249.9)	–	Attritor Mill (01HD/ 01HDDM)	–	4	4200	10	Stainless steel 2 (10)	As/Hg/Pb/Se >0.1/>0.1/ >0.1/>0.05	Montinaro et al. (2009)
Cd/Pb/Zn (20000–24000/ 2875–107857/ 10000–28000)	Synthetic sandy soil [a] Synthetic bentonitic soil [b] Synthetic kaolinitic soil [c]	Impact mill (Spex Mixer/ Mill mod. 8000)	–	4	870	5	Stainless steel 2 (10)	Cd/Pb/Zn >0.2[c]/>70[a]/>5[c]	Montinaro et al. (2008)

[a] SiO$_2$/Bentonite/CaCO$_3$/Fe$_2$O$_3$/MnO$_2$/HA: 78%/20%/0.5%/0.25%/0.25%/1%.
[b] SiO$_2$/Bentonite/CaCO$_3$/Fe$_2$O$_3$/MnO$_2$/HA: 33.5%/60%/5%/0.25%/0.25%/1%.
[c] SiO$_2$/Kaolin/CaCO$_3$/Fe$_2$O$_3$/MnO$_2$/HA: 33.5%/60%/5%/0.25%/0.25%/1%.
[d] n.d. represents "not detected", which means concentration is lower than Sample Detection Limit (SDL).

Table 2
Technical parameters and conditions of ball milling for different organic pollutants and soil characteristics.

Pollutants (Concentration (mg kg^{-1}))	Soil characteristics (wt%)	Ball mill conditions						Effects (Degradation (%))	References
		Ball mill type	Reagents (reagent–to–soil weight ratio (wt %))	Ball–to–soil weight ratio	Rotation speed (rpm)	Ball milling time (h)	Grinding ball texture, number, and diameter (mm)		
PFOS (5)	Clay	Planetary ball mill (Retsch PM 100 (D))	KOH (0.25)	33	275	6	Stainless steel 90/10 (10/15)	96%	Turner et al. (2021)
HCB (200)	Sand/Silt/Clay/Organic carbon 51.16/47.14/1.70/1.00	Planetary ball mill (QM–3SP04)	CaO/Na$_2$S$_2$O$_8$ (0.25)	30	350	2	(8)	85%	Fan et al. (2020)
Chlorpyrifos (10000) Glyphosate (10000)	Synthetic sandy soil [a] Synthetic kaolinitic soil [b]	Planetary ball mill (Fritsch Pulverisette 4)	–	11	200	5	Zirconia (10)	Chlorpyrifos/Glyphosate >95%/>95%	Hu et al. (2018)
HCB (783)	SiO$_2$/Al$_2$O$_3$/Fe$_2$O$_3$/K$_2$O/TiO$_2$ 65.85/19.58/7.64/3.98	Planetary ball mill (QM–3SP2)	Aluminum powder (0.5&1) or HA (0.5&1)	30	550	3.5	Stainless steel	>95%	Deng et al. (2018)
PCBs (1000)	SiO$_2$/Al$_2$O$_3$/Fe$_2$O$_3$/K$_2$O/TiO$_2$ 67.89/16.77/9.45/2.16/1.34	Planetary ball mill (QM–3SP2)	–	45	275	4	Stainless steel (5/10)	98%	Wang et al. (2017)
PCBs (100–5000)	SiO$_2$/Al$_2$O$_3$/Fe$_2$O$_3$/CaO/CuO 9.8/17.3/4.2/2.8/0.0022	Stirring ball milling	–	~0.4–0.67	300–500	1	Steel (5)	94.7%	Lu et al. (2017)
Hexabromocyclododecane (500)	Sand/Silt/Clay/Organic carbon/Hematite 20.1/43.2/36.7/1.46/8.35	Planetary ball mill (QM–3SP2)	CaO (25)	~40	275	1.5	Stainless steel (9.6)	98%	Zhang et al. (2014)
Phenanthrene (500) Pyrene (500)	Sandy loam	Ball mill (Retsch model MM2)	Magnetite (Particle Diameter <5 μm) (~5)	–	2000	1	Agate 5 (7)	Phenanthrene/Pyrene 10%/16%	Joseph-Ezra et al. (2014a)
Lindane (94.5)	OM content 2.30	Planetary ball mill (QM–3SP04)	Na$_2$S$_2$O$_8$/NaOH 0.35/0.15 (~0.09)	30	550	2	Zirconia (8)	95%	Zhang et al. (2021)
Phenanthrene (150)	Illite/Kaolinite/Illite or smectite mixed-layer/Quartz/Calcite/Plagioclase and hematite 53/21/19/3/2/1	Planetary ball mill (Fritsch Pulverisette 5)	Synthetic birnessite (δ–MnO$_2$) (0.15)	–	700	~0.17 (10 min)	Agate 7	~20%	Napola et al. (2006)
Decabromodiphenyl ether (8.9 ± 0.5)	–	Planetary ball mill (DSP–LBPBM06C)	NaBH$_4$ (0.0125)	5	550	2	Stainless steel	100%	Yi et al. (2021)
Petroleum	–	Planetary ball mill (XQM–4L)	–	30	500	4	Grinding balls ratio: 2:5:3 (15/8/3)	81.09%	Wang et al. (2022)

[a] SiO$_2$/Kaolinite/CaO/Fe$_2$O$_3$/HA: 78%/20%/0.5%/0.5%/1%.
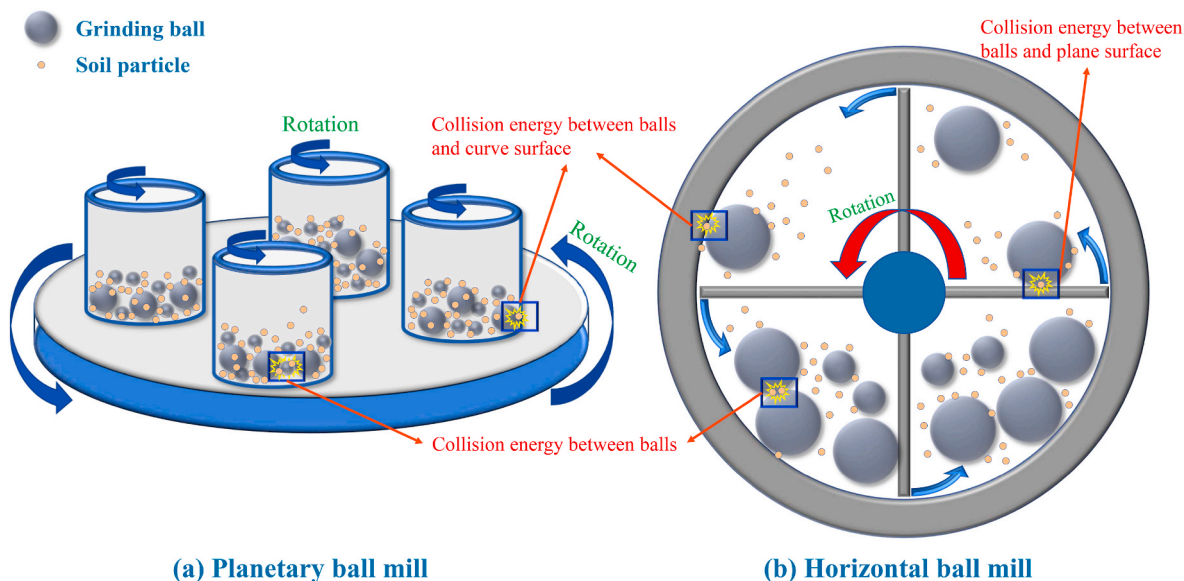[b] SiO$_2$/Kaolinite/CaO/Fe$_2$O$_3$/HA: 33.5%/60%/5%/0.5%/1%.

Fig. 2. Schematic representations of planetary ball mill (a) and horizontal ball mill (b).

gains. Additionally, the planetary ball mill uses inertial forces generated by rotation to drive the grinding ball, but centripetal forces tangent to gravity make part of the gravitational potential energy wasted. Therefore, it may not be suitable for industrialized applications (Fokina et al., 2004). Gao et al. designed and optimized a horizontal ball mill using computer simulation software, which has multiple sets of welded blades inside the jar, and the ball is driven by the blades, maximizing the effect of gravitational potential energy (Gao et al., 2021). The contaminated soils treated by ball milling in the laboratory mostly range from a few to tens of grams, and there are currently some examples of pilot-scale applications (Concas et al., 2020a; Lu et al., 2017).

Although the ball milling technology may be energy-intensive, it still shows a promising aspect in the remediation of contaminated soil (Cagnetta et al., 2018). After the working mode and operating conditions of ball milling were optimized, its mechanochemical reactions and energy utilization efficiency were enhanced, which is conducive to the large-scale popularization and application of ball milling technology.

3. Influencing factors on soil remediation by ball milling

Different process parameters and conditions may have different effects on the soil remediation efficiency of the ball milling technique. The
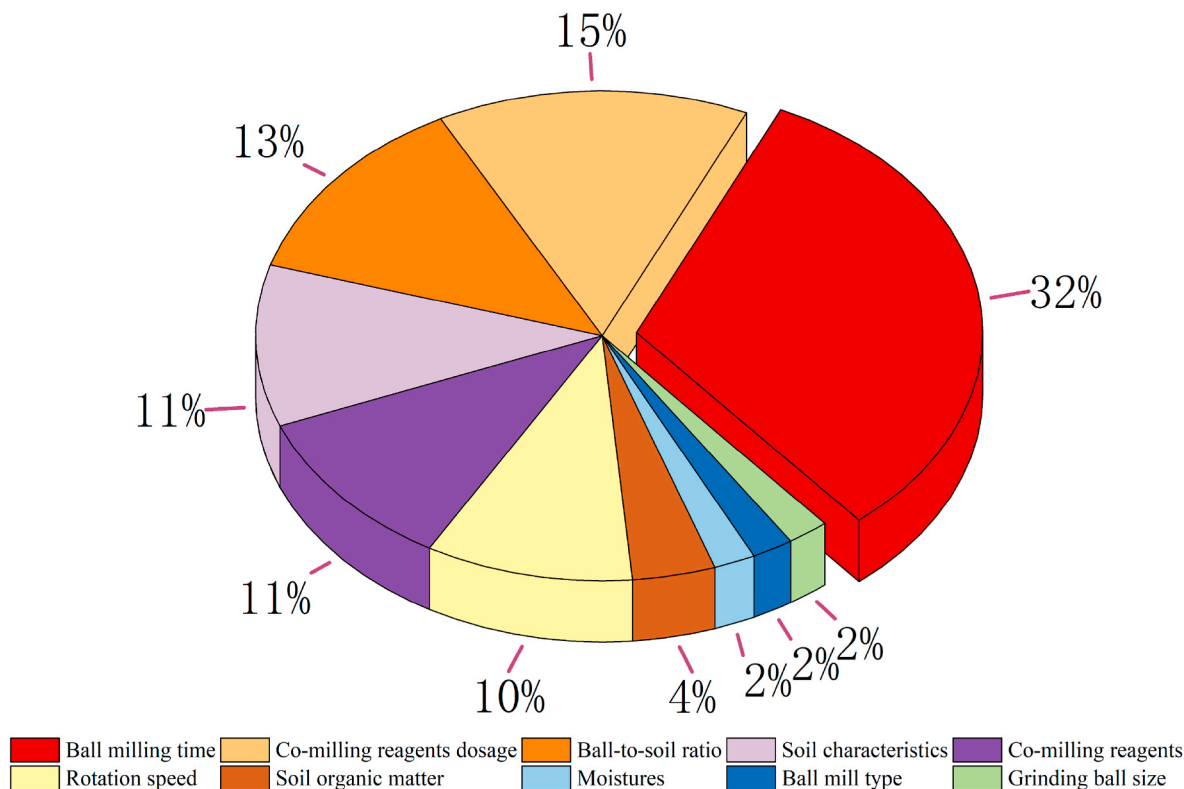

Fig. 3. The proportion of factors influencing ball milling in soil remediation (Data from 26 literatures).

optimal parameters and conditions can enhance the mechanochemical reaction rate, shorten the remediation time, and overcome the issue of low operability. Through literature statistical analysis, the BMT, co-milling reagents (including soil components), rotation speed, and ball-to-soil weight ratio are considered to be the key factors affecting the ball milling performance (Fig. 3).

3.1. Co-milling reagents

To shorten the BMT and improve the remediation performance, co-milling reagents (additives) play an important role. When a co-milling reagent (frictional material) is ball milled, it can generate surface plasmons, and undergo chemical bond cleavage with the emission of high-energy particles at the fracture site (Kaupp, 2009). Surface plasmons initiate mineralization and decomposition of organic pollutants in soils, which are constituted by very reactive radical centers. Co-milling reagents vary in their efficiency because of their abilities to supply free electrons and capture contaminants. A variety of reagents have been used in ball milling tests, including KOH, CaO, MgO, SiO$_2$, Fe/SiO$_2$ mixture, Ca(H$_2$PO$_4$)$_2$ (Tables 1 and 2). They differ in their advantages and disadvantages, with CaO being low cost and highly efficient, while SiO$_2$ can decompose the agglomerates, prevent agglomeration during ball milling and improve the treatment effect (Wang et al., 2020). Wang et al. compared the remediate effects of multiple co-milling reagents on PCBs and found that CaO had the best treatment effect with degradation efficiency above 96% after treatment for 1 h (Wang et al., 2017). Numerous studies have highlighted the effect of co-milling reagents in ball milling treatment (Hu et al., 2018; Yi et al., 2021; Yuan et al., 2018; Zhang et al., 2020). Nevertheless, most experiments focused on co-milling reagents have been restricted to laboratory research with limited practical applications (Wang et al., 2017).

Combination co-milling reagents may work better than single-reagent treatments. Zhang et al. added Fe and SiO$_2$ co-milling reagents, which achieved nearly 99.9% degradation of hexachlorobenzene (HCB) after ball milling and 84.9% using CaO (Zhang et al., 2014). Wang et al. used Al$_2$O$_3$ and K$_2$S$_2$O$_8$ as co-milling reagents to degrade per-fluorooctanoic acid (PFOA) in kaolin soil, and the degradation efficiency was 85%, but the efficiency of using Al$_2$O$_3$ or K$_2$S$_2$O$_8$ alone was 10% and 27%, respectively (Wang et al., 2019). Bimetallic co-milling reagents such as Fe/Zn are superior to previous Fe, CaO, and Fe$_2$O$_3$ co-milling reagents in the degradation of POPs by ball milling (Song et al., 2019).

Generally, to explore the role of co-milling reagents in ball milling, the main reagents and auxiliary reagents can be divided. Co-milling auxiliary reagents have particularly poor or even no degradation effect when used alone for ball milling. However, there is an obvious elevating remediation effect when it is used with co-milling main reagents. There are two general classes of co-milling main reagents: (1) compounds such as CaO (Hu et al., 2018), KOH (Turner et al., 2021), and persulfate (Zhang et al., 2021); (2) pure metals such as Zn, Fe, Mg, and Al (Hu et al., 2019; Turner et al., 2021). The co-milling auxiliary reagents are mainly neutral species (e.g., SiO$_2$) (Wang et al., 2017), metal oxides (e,g., Fe$_3$O$_4$ and Al$_2$O$_3$ (Wang et al., 2019; Yi et al., 2021) as well as hydrogen donors such as n-butylamine (Birke et al., 2009). For example, zero-valent metals combine some organic molecules (e.g., alcohols, amines or amides, and ethers), which are used as auxiliary reagents to provide hydrogen for the reaction system. This combination facilitated the complete dehalogenation of organic pollutants and reduced the potential for the generation of toxic by-products. Birke et al. confirmed that Mg combined with n-butylamine could degrade PCBs, pentachlorophenol (PCP), and dichlorobenzene (DCB) by ball milling method (Birke et al., 2009). In bimetallic systems, zero-valent metals (e.g., Zn, Al, Fe, and Mg) are electron donors, and transition metals (e.g., Pd and Ni) are catalysts that facilitate the corrosion and hydrogenation processes of metals. Yi et al. found higher removal efficiency of Fe/Ni than CaO for the removal of decabromodiphenyl ether from the soil (Yi et al., 2021). Besides, the main reagent was also sometimes used as an auxiliary

reagent. Because they act as an active reagent to provide free electrons for the main reagents in the reaction system. For instance, CaO is widely used in ball milling processes but was served as an auxiliary reagent in some studies. Fan et al. used CaO as a co-milling auxiliary reagent to activate persulfate and effectively degraded HCB in contaminated soil (Fan et al., 2020). In soil remediation, the ratio between the main and the auxiliary agent should be determined based on the contaminant and soil characteristics. At 550 rpm ball milling speed, the mineralization rate of lindane was 100% after being treated for 4 h when the ratios of Na$_2$S$_2$O$_8$, NaOH, and soil are 0.35:0.15:5, respectively (Zhang et al., 2021).

The ratio of co-milling reagents to contaminated soil [reagents-to-soil weight ratio, RR (wt%)] is another important influence factor. After increasing RR values, the contact of both can be raised to enhance soil remediation effects. In previous studies, the RR value was 0.05 minimum and 50 maximum (Tables 1 and 2). Therefore, to save energy and maximize remediation efficiency, the optimum ratio of co-milling reagents to soil should be selected properly. For example, when the RR value of Ca(H$_2$PO$_4$)$_2$ with Pb contaminated soil increased from 4 to 10 wt%, just ball milling for 2 h, the leaching concentration of Pb has been below the regulatory limit of 5 mg L^{-1} (Zhang et al., 2020). Therefore, RR is an important indicator for the remediation of both organic and inorganic contaminated soils.

The particle size and hardness of co-milling reagents have certain effects on soil remediation efficiency. The particle size is too small, the energy delivered by ball milling is not enough to initiate a mechanochemical reaction. If the particle size of SiO$_2$ is small, it cannot be easily decomposed, it is difficult to form free radicals so that the dehalogenation efficiency of pollutants decreases (Wang et al., 2020). Compared with 0.750 mm and 0.375 mm SiO$_2$, 0.375 mm is more effective, but it is almost ineffective when it is less than 38 μm (Zhang et al., 2014).

Co-milling reagents mainly include reductants, alkaline earth metal oxides, neutral substances, and oxidants (Cagnetta et al., 2016). Reducing reagents generally act as electron donors and low acidic hydrogen sources, such as base metals and hydrogen donors. Zero-valent metals are strong electron-donors, which means they also have excellent performance when used alone (Sui et al., 2018). In addition, the combination of hydrogen donors with pure metal has a synergistic effect (Birke et al., 2009). After ball milling, calcium polysulfide [Ca(S$_x$)] was utilized to achieve the reduction and immobilization of Cr(VI) simultaneously, facilitating the leaching concentration reduction of Cr(VI) (Yuan et al., 2019a).

Alkaline earth metal oxides such as MgO, Al$_2$O$_3$, and La$_2$O$_3$ combined with ball milling can effectively degrade mono-chlorobiphenyl (Tanaka et al., 2005). These oxides have been used together with neutral species, such as SiO$_2$, to enhance their remediation capacity. Recently, highly efficient oxidants, such as persulfate, have been used in many studies as co-milling reagents. With the aid of the energy applied by ball milling, persulfate can produce strong oxidizing radicals (SO$_4^{\bullet-}$ and hydroxyl radicals, etc.) to degrade and mineralize organic pollutants (Wang et al., 2019). CaO and Na$_2$S$_2$O$_8$ were selected as the co-milling reagents, HCB was converted to inorganic matter through dechlorination reaction and achieved an 85% degradation rate after 2 h ball milling (Fan et al., 2020).

The co-milling reagents need to be selected according to the soil composition, and the suitable co-milling reagents and BMT must be determined by pre-experiment before practical applications. For the lightly contaminated soil, it can also achieve a better remediation effect without the addition of co-milling reagents, while controlling the cost and alleviating its effect on soil characteristics. The heavily contaminated soil can be added with a suitable amount of co-milling reagents, which can guarantee the remediation effect of the contaminated soil. By ball milling hexabromocyclododecane contaminated kaolin and red soil, respectively, Fe/SiO$_2$ enhanced the remediation effect in contaminated kaolin, while CaO performed better than Fe/SiO$_2$ in red soil (Zhang et al., 2014). More energy consumption is necessary to achieve

equivalent results when co-milling reagents are not added.

Co-milling reagents play an important role in shortening the BMT as well as enhancing the treatment effect, but their impacts on soil texture are currently unknown. Previous studies demonstrate that ball milling can activate the catalytic properties and adsorption capability of clay and oxide components in soils (Hu et al., 2018). These activated minerals may trigger the rapid degradation of organic pollutants in soil without the need for additional co-milling reagents. Researchers should make full use of the natural components of the soil, which have been shown to be effective in remediation by previous studies (Cagnetta et al., 2016; Concas et al., 2020b; Zhou and Haynes, 2010). The excess co-milling reagent rather wastes the volume of the ball mill jar, even if recycled there is still residual unreacted reagent. For example, the excessive addition of CaO may increase soil alkalinity, which is not conducive to plant growth and soil reclamation. Wang et al. conducted a ball milling test with the addition of CaO or SiO_2, and after 4 h of ball milling, the degradation rate of PCBs was not greatly promoted compared to that without co-milling reagents (Wang et al., 2017).

There are large differences in the physicochemical characteristics of different co-milling reagents, and careful selection of co-milling reagents is required for soil remediation for different aims and applications. Considering the use effectiveness and economic cost, it is necessary to explore co-milling reagents with rich sources and similar properties to soils, such as natural minerals and harmless solid wastes. Magnetite, for example, has been reported to facilitate the remediation of ball milling in soils contaminated with polycyclic aromatic hydrocarbons (Joseph-Ezra et al., 2014a).

3.2. Ball milling time

Increased BMT promotes the immobilization and degradation of contaminants in the soil. An appropriate BMT can ensure adequate contact of co-milling reagents with contaminants and adequate energy transfer. After extending the BMT, the metals gradually transformed from a weak acid-soluble fraction to a relatively stable fraction, and the content of the residual fraction increased (Yuan et al., 2019b). The D_{50} (median grain size) decreased with prolonged BMT, and the newly fractured soil particles would have a larger specific surface area (SSA) (Yuan et al., 2018). These new surfaces provide many reactive sites that immobilize or degrade soil heavy metals and organic pollutants (Plescia et al., 2003). Soil particles were dominated by large particle sizes (~100 μm) in the initial stage of ball milling, decreasing to the nanoscale after prolonged BMT (Montinaro et al., 2009). This process leads to aggregation and agglomeration of soil particles, resulting in heavy metals adsorbed on the particles entrapped in the crushed particles (Concas et al., 2007). The degree of aggregate with a compact structure formed by soil particles was also elevated after extending the BMT, thus reducing the toxicity of pollutants. Yuan et al. found that the leaching concentration of Cu decreased below regulatory limits after 15 min with prolonged BMT, making it easier for Pb to chelate with soil organic matter (SOM) and thereby promoting heavy metals transferring mobile fractions to stable fractions (Yuan et al., 2019b). When the BMT increased from 0.5 to 4 h, the leaching concentration of Cr(VI) decreased from above 60 mg L^{-1} to below 5 mg L^{-1} (Yuan et al., 2019a). Similarly, the degradation efficiency of PCBs was approximately 98% after ball milling for 4 h (Wang et al., 2017).

The relative collision between the grinding ball and soil increases with increasing the BMT, thereby facilitating energy transfer between them. Rosenkranz et al. used a discrete-element simulation model to study the grinding ball motion of planetary ball mill and pointed out that the BMT is positively correlated with the transfer of stress-energy (Rosenkranz et al., 2011). In previous studies, most of the BMT was from 1 to 5 h (Tables 1 and 2). Accordingly, to guarantee the remediation effect of ball milling on the contaminated soil, an appropriate BMT needs to be selected to ensure remediation efficiency.

3.3. Filling ratio and ball-to-soil ratio

The amount of filling of the grinding balls inside the jar depends on the ball mill jar volume, and the amount of filling ratio further affects the ball milling strength. The filling ratio determines the effective number of impingements of the grinding ball on the soil and, in turn, the energy density transferred to the soil. A small filling ratio results in a low amount of energy accumulated within the system. When too much soil and grinding balls are in the jar, the new surface of the soil increases rapidly and the jar is quickly filled. The effective impact of the grinding ball is simultaneously reduced, weakening the remediation efficiency of the contaminated soil. Wang et al. set a filling ratio of 1/3 in the ball milling test with only 0.3% remaining dechlorane plus after 2 h of treatment at a high mass ratio of dechlorane plus (Wang et al., 2016). Moreover, the grinding balls interfere with each other when they are in excessive numbers, making the effective collision energy drop. When the mill jar filling ratio was at 30–40 vol%, the effective collision energy of multiple grinding balls was an approximately linear accumulation of a single grinding ball. An appropriate filling ratio can ensure adequate contact between them and promote co-milling reagents and soil components to function.

Increasing the ball-to-soil weight ratio in jars can reduce the mean free path of grinding ball collision and enhance the collision frequency. This allows the contaminated soils to gain more mechanical energy, which promotes remediation efficiency. Concas et al. defined the cumulative energy in mechanochemical energy transfer that, after ball milling for a given time, acts on the total energy change per unit mass of soil (Concas et al., 2020b). The resulting calculation formula suggests that the ball-to-soil ratio has a positive effect on the cumulative energy (Concas et al., 2020b). Mio et al. used a computer simulation of the grinding ball movement in a ball mill jar and found that the resulting impact energy could also be improved as the number of balls increased (Mio et al., 2002). The energy density of a unit mass ball is in a certain range, and the mechanochemical reaction can initiate the contaminant conversion. The mass fraction that is converted depends on the total energy delivered by ball milling (Delogu et al., 2004).

An elevated ball-to-soil ratio increases the number of collisions between the pellet and the soil while generating high energy to promote soil remediation (Wang et al., 2022). The low ball-to-soil weight ratio leads to an insufficient collision between the co-milling reagents, grinding balls, and soil. Yuan et al. improved the ball-to-soil ratio by increasing the number of grinding balls, and the leaching concentration of Cr(VI)-contaminated soil decreased to 5 mg L^{-1} when the ratio was above 10 under other conditions optimum (Yuan et al., 2018). After the ball-to-soil ratio is raised to a certain value, the promotion of soil remediation is limited, so an appropriate ball-to-soil ratio should be selected. Tables 1 and 2 summarize the ball-to-soil ratios reported in the literature, and the ratios range from 4 to 45. The total masses of the grinding balls are all larger than those of the contaminated soils, and sufficient ball numbers are available to provide the collision energy required for soil remediation.

The ball-to-soil weight ratio is higher for remediation of organic contaminated soil, mostly 30–45, and 4–14 for inorganic contaminated soil (Tables 1 and 2). For the effective remediation of soils and to avoid excessive energy consumption, the optimal ball-to-soil ratio should be determined.

3.4. Rotation speed

Ball milling rotation speed, which involves the interaction between mechanical energy and chemical energy. The higher the rotation speed, the better the soil remediation effect. 94% of the energy produced by ball milling is derived from collisions and about 6% is provided by friction and shearing (Chattopadhyay et al., 2001). For planetary ball milling, varying the rotational speeds (revolution and autorotation speeds) of rotary disk and jar, the collision and friction components can

be adjusted, i.e., compressive and shear forces resulting from grinding ball impingement. They can affect the collision frequency and speed of the grinding ball, ultimately modifying the energy transfer efficiency. When the rotation speed is too high, the soil may be U-shaped adherent to the inner wall of the jar difficult to detach, which affects the remediation effect. Santhanam and Dreizin verified the energy exchange and rotation speed dependence using the discrete element model (Santhanam and Dreizin, 2012). The rotation speed of ball milling is 200 rpm, the leaching concentration of Cr(VI) in the soil is about 100 mg L^{-1}, which increases above 400 rpm, and its concentration decreases to 5 mg L^{-1} (Yuan et al., 2018). It is known from Tables 1 and 2 that the rotation speed upon ball milling is the most at 200–500 rpm. Soil remediation involves both economic costs and environmental benefits. To meet the soil environmental quality standards, higher rotation speeds (such as 500 rpm or even higher) are required, but suitable rotation speeds should be chosen to avoid unnecessary energy costs.

3.5. Grinding ball size

Different grinding ball sizes also contribute to ball milling remediation of contaminated soils. The small size of the ball is beneficial for reducing soil adhesion to the grinding ball because it is more prone to shear forces, and it generates lower collision energy due to its low weight. The large size of the grinding ball is heavier and it generates more collision energy, and its number is limited by the jar volume resulting in a lower collision frequency. In addition, the large size of the grinding ball easily produces a collision dead angle in the jar. The grinding ball diameters used for soil remediation ranged from 5 to 40, with 10 mm being the most selected grinding ball and 10 mm paired with 5 mm or 15 mm the most when combined (Tables 1 and 2). Gao et al. used ball milling to remediate DDT contaminated soils, and found that diameters of 15 mm and 19.6 mm were more effective than 8 mm or 10 mm (Gao et al., 2021). Sometimes only a fraction of the energy generated by a collision can be transferred to the soil, and the key to initiating a mechanochemical reaction is the single collision energy whose reaction rate is controlled by effective collision power. When soil remediation by horizontal ball milling, the ball diameter should be no less than 30 mm to meet the requirement of single collision energy (Gao et al., 2021).

In summary, the analysis of the current study is still unable to clarify the relationship between the grinding ball size and the remediation effect of contaminated soils, nor to directly select the optimal grinding ball match ratio or grinding ball size. Yet, the effects of ball size and weight on the remediation of soils by ball milling still need to be considered. It is recommended that the grinding ball size be optimized in the future, such as computer models and actual experiments.

3.6. Other influencing factors

SOM exhibits two different effects on soil remediation: (1) Competition with pollutants for active radicals hinders the degradation of pollutants; (2) It can increase the mutual contact of individual reactants and improve the remediation efficiency of soil pollutants (Fan et al., 2020; Montinaro et al., 2008). The addition of H_2O_2 to the soil, or heating to 600 °C, increased the HCB degradation efficiency by 13% after heating, confirming the negative impact of SOM on ball milling (Fan et al., 2020).

When the soil moisture content is excessive, the energy generated by ball milling is first delivered to the liquid phase rather than the solid phase, and the contaminated soil receives less energy. Moisture can hinder the generation of active sites and eventually lead to a decline in remediation efficiency. Similarly, ball milling can be blocked or interrupted by moisture. Moisture can promote the formation of soil agglomerates, causing the soil in the jar to produce the sticky wall cake. Furthermore, CaO absorbs moisture from the environment in long-time ball milling, which easily produces sticky walls. Therefore, co-milling

reagents should guarantee absolute dryness. Turner et al. conducted a ball milling test of PFOA and perfluorooctanesulfonic acid (PFOS) in sands, the degradation rate of PFOA in sand containing 100% saturated water decreased by about 40% compared with that in dry sand (Turner et al., 2021). Co-milling reagents might eliminate the effect of water. For example, KOH promoted the removal of PFOA and PFOS from sands with a water saturation of 100%, and the destruction percentage is 92% and 99%, respectively (Turner et al., 2021).

Because the internal impact force is extremely strong when ball milling runs, which easily causes local high temperature, at the collision point of the grinding ball with the soil, the temperature is much larger than that in the jar, possibly reaching 1000 K (Monagheddu et al., 1999; Urakaev and Boldyrev, 2000). The risk of generating dioxins and their precursor substances, such as PCBs and polybrominated diphenyl ethers (PBDEs), needs to be carefully evaluated in ball mill large-scale treatment of POPs. When dioxin precursors are present in contaminated soils, the local high temperature inside the ball mill may drive polybrominated and polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PXDD/Fs) formation. These highly toxic substances are easily formed due to the influence of temperature during ball milling (around 300 °C) and large amounts of iron elements (PXDD/Fs catalysts) in the grinding ball and jar. Therefore, there is a need to avoid the generation of PXDD/Fs, which can be operated intermittently, or to add a cooling system, always controlling the temperature of the ball milling system below 60 °C (Lu et al., 2017).

In conclusion, various factors affect the remediation process of contaminated soils by ball milling techniques (Fig. 3). Therefore, future research is warranted to ascertain the remediation effect changes in ball milling techniques under different parameters and conditions in soil remediation.

4. Mechanisms of the ball milling technique for the treatment of contaminated soils

4.1. Inorganic pollutants

Soil is a complex and heterogeneous system, and ball milling can trigger the activation of multiple components in the soil. Low-intensity ball milling is capable of comminuting soil particles. Comminuting can increase the SSA of soil components and enhance the adsorption capacity significantly, and these surfaces are extremely reactive. Meanwhile, the stratified structure of the soil is destroyed and the crystallinity of mineral components decreases, thus leading to structural cracking of soil minerals (i.e., lattice deformation), and heavy metals are more likely to diffuse into the deformed lattice network structure. For instance, ball milling can cause delamination of aluminosilicate structures due to the friction forces when exfoliating clay minerals (Di Leo et al., 2019). The collision can comminute the clay particles and increase their SSA. High-intensity ball milling leads to deformation of the crystal structure of the clay, exposing silicon-oxygen tetrahedrons on the outer surface with easier breakage of their edge chemical bonds (Di Leo et al., 2019). Clay minerals and oxides are considered the main soil inorganic components responsible for chelation, complexation, adsorption, and precipitation of inorganic contaminants in soils (Uddin, 2017; Zhou and Haynes, 2010). SOM similarly has an important role in the chelating immobilization of heavy metals in soils. In addition, the high-energy impact of ball milling on soil components not only produces a transient high temperature but also causes the rotation, alignment, and reorientation of molecules (Wang et al., 2019). Thus, sustained ball milling resulted in the transformation of soil components while enhancing the ability of soil components to stabilize heavy metals (Fig. 4a).

4.1.1. Adsorption

The new surface formed after the soil particles were comminuted by ball milling had abundant electrons and was extremely chemically
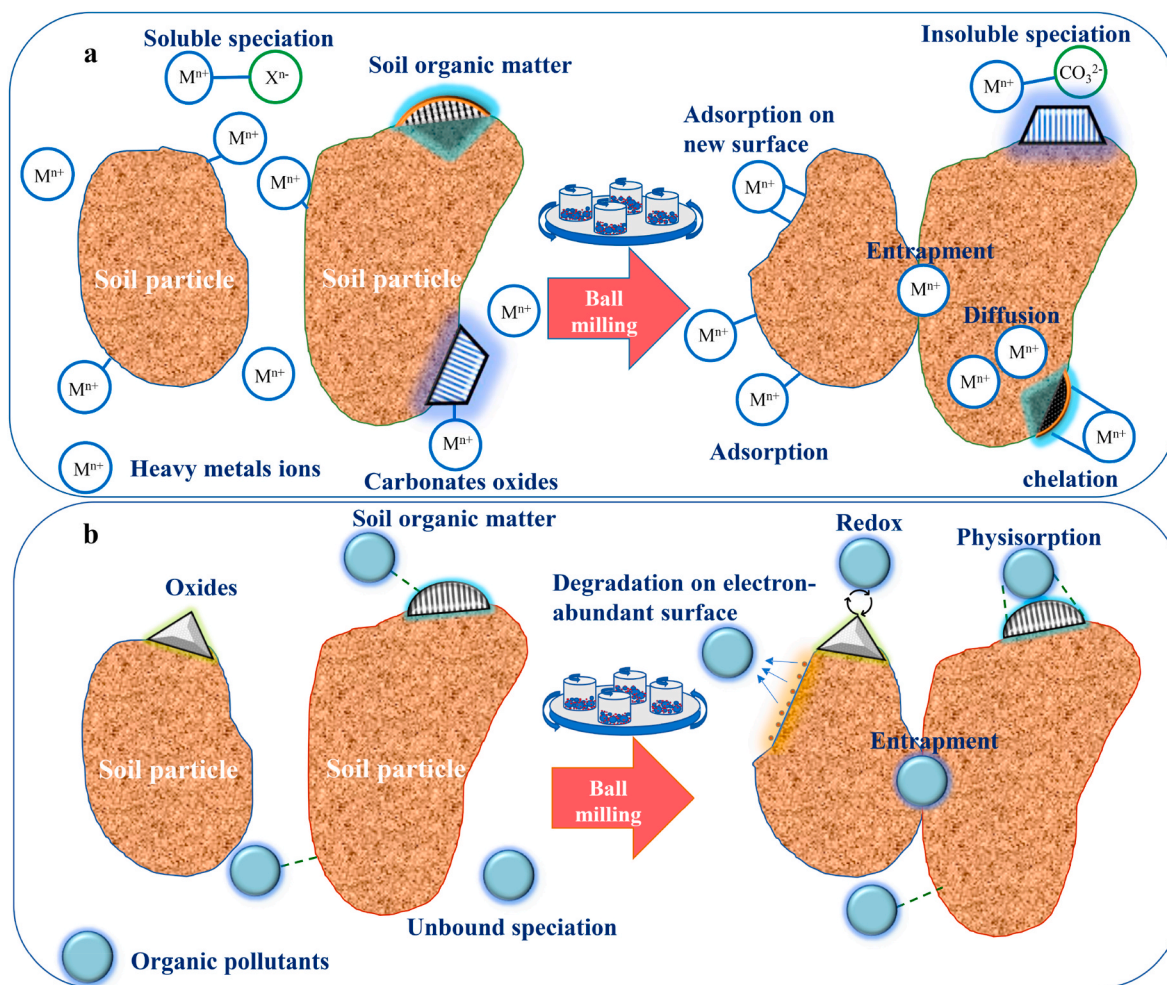
Fig. 4. Remediation of possible mechanochemical interactions in inorganic (a) and organic (b) contaminated soils by ball milling.

active. It has a large number of highly reactive dangling bonds, thus initiating chemical reactions to adsorb contaminants. Heavy metals undergo chemisorption on these surfaces, which is one of the main mechanisms by which ball milling immobilizes metals in soils (Montinaro et al., 2007).

Ball milling can enhance the immobilization and adsorption capacities of soil clay components for heavy metals, making them very active. Clays are aluminosilicates with very small particles and possess a series of tetrahedral sheets (SiO_4), which, by combining reactive oxygen species with octahedral (AlO_6), constitute a basic structure unit layer (Fig. 5). Different clay mineral components make up different proportions of the basic structure unit layers. In the interlayer spaces of clay minerals, physical adsorption of heavy metals occurs through cation exchange. While at the unit layer surface of clay minerals, there are many dangling bonds for the complexation with metals to undergo chemisorption.

Non-clay inorganic components in soils, such as birnessite and ferrihydrite, positively affect the immobilization of metals after ball milling. They have unique surface electrochemistry, cation exchangeability, and redox properties. Especially, octahedral vacancies in the structure of birnessite, which has been considered as strong adsorption sites for heavy metals (Manceau et al., 2002). After contact of manganese oxide with water, due to hydration of surface functional groups (e.g., –OH), a hydrated surface is generated and its surface charge changes. Ball
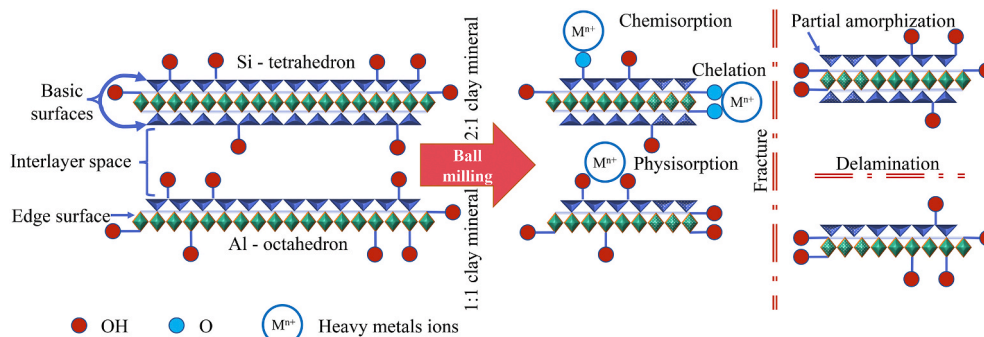

Fig. 5. Possible mechanochemical reactions of clay minerals and the corresponding heavy metals stabilization process during ball milling.

milling can drive this hydration, facilitating the entry of water molecules into the mineral lattice (Boldyrev, 2002). In brief, heavy metals could be adsorbed on the surface of soil particles during ball milling as schematically shown in Fig. 4a.

4.1.2. Aggregation

Another major mechanism involved in ball milling the treatment of contaminated soils is the agglomeration and aggregation of soil particles (Montinaro et al., 2007). First, heavy metals in contaminated soils are adsorbed by soil particles through surface coordination. The loose soil produced stable aggregates of the compact structure after ball milling. Meanwhile, ball milling leads to the fragmentation of soil particles and the constant formation of crystal defects and partial amorphization, producing excessive free volume at the grain boundary structure (Kuru et al., 2009; Schaefer et al., 1988; Van Petegem et al., 2003). After the soil particles are comminuted to the micrometer scale, they have high surface energy and forms agglomerates under the influence of van der Waals adhesion force and electrostatic interaction. These aggregates can entrap metals between the particles. Soil particle agglomeration and aggregation may be responsible for this phenomenon. Although the adsorbed metals are only entrapped between the mineral surfaces of the two soil particles, it is sufficient to prevent metal leaching (Concas et al., 2007). Additionally, heavy metals complexes diffuse into the crystalline reticulum of soil particles, thereby enhancing the immobilization of metals (Montinaro et al., 2007).

4.1.3. Complexation

Highly active dangling bonds occur on partially amorphous surfaces, and the hydroxyl groups corresponding to dangling bonds have an extremely strong affinity for metal cations in soils, both of which can be combined by complexation. Hence, the formation of new surfaces, described above, is very important for ball milling strengthening heavy metals immobilization. Just a few hours of ball milling allowed heavy metals to be persistently immobilized on soil particles (Table 1). Therefore, delamination and comminuting of clay minerals often allow dangling bonds to form on their surface, favoring the immobilization heavy metals. The energy produced by ball milling can rupture SiO_2, which can form É center ($\equiv Si\bullet$) and non-bridging oxygen hole centers ($\equiv Si-O\bullet$), eventually combine into very stable Si–O–Me bonds (Li et al., 2020).

SOM binds to metals through different mechanisms, including chelation, weak electrostatic effects, and complexation, reducing its bioavailability. In particular, humic acid (HA) has the chelating ability to immobilize toxic metals in soil (Yang and van den Berg, 2009). Because there are a large number of reactive oxygen-containing groups such as –OH, –COOH, and quinone groups in HA. Metal ions are coordinated, complexed, or chelated to these functional groups through ionic and coordination bonds. Changes in pH can affect the surface charge and adsorption capacity of SOM. Whether HA is proton donors or heavy metals ion acceptors depends on it (Dobranskyte et al., 2006). Ball milling can cause chemical transformation of SOM, increase the fraction of aromatic fragments, and enhance the degree of branching of hydrocarbon chains. Briefly, the breakage of the C–C bond makes the length of the hydrocarbon chain shorten into an active structure. After undergoing hydrolytic oxidation, the content of oxygen-containing functional groups such as –OH increased (Savel'eva et al., 2016). HA may be transformed into various functional groups, such as methoxy, phenol, carboxyl groups, and quinoid fragments under the impact of ball milling (Savel'eva and Yudina, 2014).

4.1.4. Precipitation

The immobilization of metal species in soils also occurs by precipitation, some soluble metals undergo precipitation reactions with various inorganic substances in soils. For example, the dissolution-precipitation process of calcium carbonate with metal ions (Zeng et al., 2020):

$$CaCO_3 \rightarrow Ca^{2+} + CO_3^{2-} \tag{1}$$

$$Me^{2+} + CO_3^{2-} \rightarrow MeCO_3 \tag{2}$$

Previous studies have confirmed its immobilization on metals (Genç-Fuhrman et al., 2004; Lee et al., 2007). Surface precipitates are more likely to form when one metal is close to the ionic radius of calcium ions. Thus metals are less easily desorbed when their ionic radii are similar (Zachara et al., 1991). In general, calcium carbonate is very stable and very difficult to dissolve in solution, so it is not easy to chemically react with metals in soils. But the collision pressure in ball milling is extremely high (3.30–6.18 Gpa). This favors the creation and diffusion of carbonate lattice defects (Gaffet et al., 1999), with some fractions being amorphous. Constantly apply mechanical forces (e.g., collisions, shearing, and extrusion) to $CaCO_3$, allowing it to be continuously activated (Xiong et al., 2020). This can provide a lot of active sites for precipitation reaction. After ball milling, the lattice of $CaCO_3$ was distorted and continually grew up, making the free volume of the crystal increase. Therefore, larger metal cations are more easily trapped by the crystal lattice (Concas et al., 2007).

The processes that may be involved in the mechanochemical remediation of inorganic contaminated soils have been explored, and the mechanisms of these processes have not been explained in detail. For the phenomenon resulting from ball milling, only some of the above hypotheses are currently proposed. Therefore, these mechanisms require further investigation.

4.2. Organic pollutants

Different from inorganic pollutants, organic pollutants were reduced to amorphous carbon, light hydrocarbons, and graphite after ball milling, and finally achieved dechlorination and even complete mineralization (Wang et al., 2017). This transformation is an energy-initiated degradation reaction provided by the collision in ball milling. Therefore, for the complete mineralization process of organic waste, the study of mechanochemical activation is one of the important parts. Ball milling can fragment and activate soil components and co-milling reagents, facilitating their full contact (Rowlands et al., 1994). The generation of free electrons or highly reactive radicals by co-milling agents, including soil components, initiates subsequent degradation reactions, such as fragmentation/polymerization (Nomura et al., 2005), (de)hydrogenation (Tanaka et al., 2003), redox (Turner et al., 2021; Wang et al., 2019), oligomerization (Nasser and Mingelgrin, 2014), and de(hydro)halogenation (Aresta et al., 2003; Birke et al., 2004; Monagheddu et al., 1999). Upon accumulation of sufficient energy, halogen atoms detach from the organic pollutant to form inorganic water-soluble halide ions (X^-). This section combs mainly from the interactions of soil components with organic pollutants resulting from ball milling (Fig. 4b), which initiate the transformation of organic pollutants.

Generally, it is difficult for soil to react with organic pollutants upon ordinary grinding and simple mixing, while the degradation of organic pollutants can be accelerated by the mechanical energy provided by ball milling. Because, under the effect of mechanochemistry, the surface of soil components is activated, and the produced reactive species (free electrons and free radicals) act with organic pollutants adsorbed on the surface of soil components, and destroy them, ultimately generating nontoxic products (Hu et al., 2018). The basic reaction processes of the ball milling method for the remediation of organic pollutants are as follows:

(1) Adsorption and mechanochemical activation. Ball milling enabled soil components and organic pollutants to mix well and brought solid to close contact while producing mechanochemical effects to activate the surface activities of soil components.

(2) Degradation reactions. Free electrons and/or highly reactive radicals generated by soil components react with organic pollutants, thereby initiating a series of reactions (e.g., dehalogenation/halogenation, (de)hydrogenation, redox, fragmentation/polymerization).

(3) Mineralization. After accumulating enough energy, usually, organic pollutants are reduced to a mixture of amorphous carbon, graphite, and light hydrocarbons; Dehalogenation of halogen atoms from organic pollution to form inorganic water-soluble X^- culminates in achieving dehalogenation and complete mineralization of the target contaminants.

The activation effect of ball milling on soil components is fundamental to achieving the degradation of organic pollutants, and the resulting reactive species and a series of reactions between pollutants are the key processes that lead to the destruction of organic pollutants. Although the mechanism of ball milling degradation of organic pollutants has not been fully elucidated yet, certain research progress has been made. This section focuses on the changes in soil composition caused by ball milling and a series of processes for the adsorption and/or transformation of organic pollutants.

4.2.1. Adsorption and mechanochemical activation

Ball milling can enhance the adsorption capacity of clay components in soils for organic pollutants. In the process that organic pollutants began to decrease at the beginning of ball milling, physisorption played a major role. The effectiveness of ball milling depends on the repeated impact of the ball milling tool (generally a ball). They can increase the SSA of the clay particles and make adequate contact with the particles and organic molecules (Nasser and Mingelgrin, 2012). Hydrophobic organic compounds and clays are mainly bound to the external surface of the clay through weak interactions (van der Waals type), and hydrophilic molecules form inner-sphere surface complexes (binding via hydrogen bonding with oxygen atoms in the clay mineral interlayer space). Under mechanochemical action, the delamination of clay minerals elevates the SSA, thus creating more adsorption sites (i.e., broken bonds at the surface). On the other hand, the clay mineral crystal structure produces partial defects which elevate the adsorption capacity.

SOM includes stable humic substances, plant residue, dissolved OM, and the most active microbial biomass. They are related to the physicochemical and biological properties of soils, and especially their adsorption capacity can be enhanced by ball milling. The addition of HA to birnessite promoted the degradation of tetrabromobisphenol A due to the covalent coupling of oxidized HA with organics (Tong et al., 2016). Meanwhile, ball milling allowed the dissociation of acidic groups from HA, improving its surface activity (Skybová et al., 2007). As the OM in the soil increases, the degradation efficiency may decrease. There is mutual competition between OM and pollutants for active sites, and the active radicals involved in degradation are consumed by it (Zhou et al., 2019).

In the process of ball milling, various oxides in soil are extremely active, and they are an important component of the remediation process of ball milling. There are oxides with compositions such as alkaline-earth metals, metalloids, and transition metals. For example, oxides such as SiO_2, CaO, MgO, and MnO_2. They are also considered highly efficient co-milling reagents. Ball milling elevates their contact area and reaction rate, which in turn provides removal conditions for contaminants. Ball milling causes the accumulation of crystal structure defects, amorphization, and fragmentation of crystal particles in silica (metalloid oxide) (Aman and Tomas, 2004; Damm and Peukert, 2009; Delogu, 2011; Kosobudskii et al., 2015). The simultaneous covalent bond homolysis occurred and a large number of unpaired electrons were generated on its surface (Fig. 6). The generated free radicals and free electrons are particularly active, initiating pollutant degradation reactions by attacking C–X^- bonds on organic contaminants (Kaupp, 2009). There are also alkaline-earth metal oxides like CaO and MgO. In the solid-solid reaction system, the contaminants come into contact with alkaline-earth metal oxides, and the contaminants are adsorbed on their
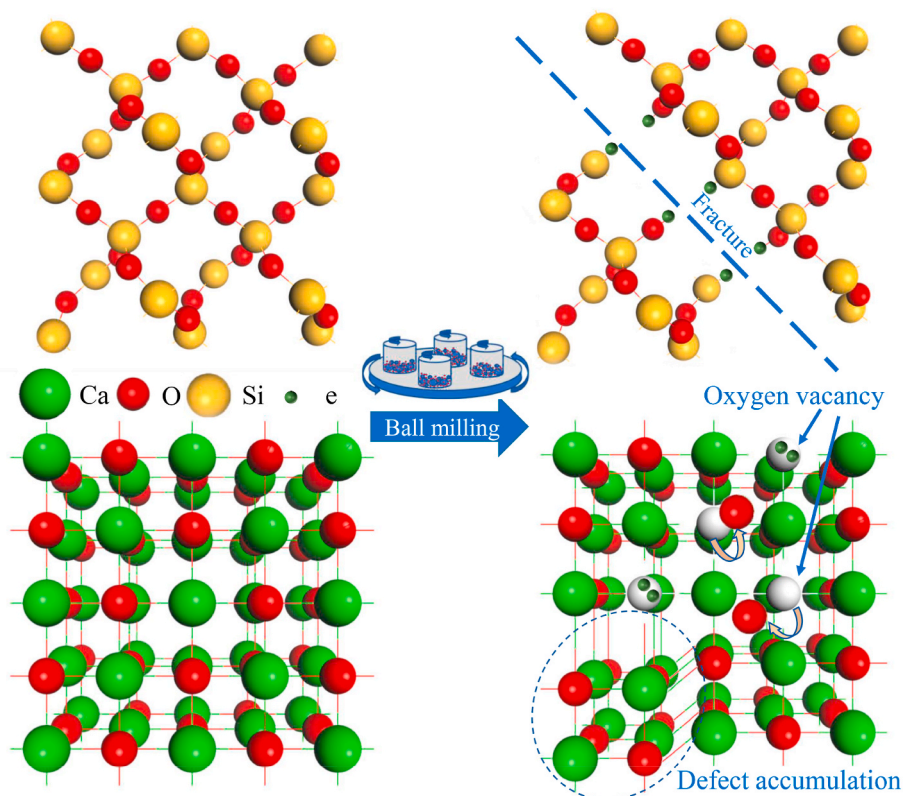

Fig. 6. Possible mechanochemical reactions of silica and calcium oxide during ball milling.

surface. CaO is activated by mechanochemical action and reactive sites (oxygen vacancies) are formed in the crystal (Fig. 6). Cagnetta et al. observed the formation of oxygen vacancies by CaO during ball milling, and the internal free electrons were released (Cagnetta et al., 2017).

4.2.2. Degradation reactions

The clay surface was acidic after ball milling treatment, which could enhance the degradation reaction of organic contaminants (Nasser and Mingelgrin, 2012). For example, on the clay's structural unit layer edge or interlayer space, the exposed aluminum ions are treated as Lewis acids, so the acidic surface of kaolinite can promote the catalytic oxidation of organic pollutants. Copper ions (transition metal center) on the surface of montmorillonite facilitates the catalytic degradation of pyrene, which can be degraded thoroughly through oxidation and dimerization (Joseph-Ezra et al., 2014b). By ball milling, organic contaminants undergo stepwise and multistage dehydrohalogenation, which are then cleaved to low molecular compounds and inorganic halides. Some other organic contaminants, such as DDT, undergo dehydrochlorination and dechlorination, carbon-carbon bond rupture, and migration of chlorine atoms and phenyl groups (Song et al., 2019).

Transition metal oxides are different from alkaline-earth metal oxides, although it also forms oxygen vacancies and releases a large number of free electrons upon the ball milling process. But there is a high-valent cation in transition metal oxides, which can trap the generated electrons and disfavor the aggregation of free electrons on the particle surface. Birnessite (δ–MnO_2) during ball milling, the MnO_6 layer would change, Mn^{4+} in the octahedral center of MnO_6 layer formed an inner-sphere surface complex with organics, and the oxygen bridges combined Mn^{4+} with the phenolic groups of pentachlorophenol (PCP) (Di Leo et al., 2013). Then, Mn^{4+} accepts electrons from PCP, and phenoxyl radicals are formed. This illustrates that Mn^{4+} is decreased after reduction while generating Mn^{3+} and new vacancies. Immediately afterward, organic contaminants reduce Mn^{3+} to Mn^{2+}, and Mn^{2+} is released. The reduction of $Mn^{3+} \rightarrow Mn^{2+}$ is more important for the mineralization at the initial stage of organic contaminants. Because before the degradation efficiency is less than 50%, Mn^{3+} accepts electrons of organic contaminants more easily than Mn^{4+}, initiating the destruction reaction (Chai et al., 2018). Upon consumption of Mn^{3+} to a certain amount, the structure of MnO_2 is distorted and the reaction of $Mn^{4+} \rightarrow Mn^{3+}$ starts to dominate. Concomitant with the above redox reactions, birnessite releases lattice oxygen, which is the major source of reactive oxygen species for the mineralization of organic pollutants (Chai et al., 2018). This degradation mechanism illustrates that the released surface lattice oxygen is important for the mineralization of organic pollutants.

4.2.3. Mineralization

Since halogen atoms are electronegative, but also the dissociation energy of the chemical bond in which the halogen atom is located is very strong. Halogen atoms are detached from the organic molecules with the simultaneous production of organic radicals and oxygen radicals. Most halogens are eventually trapped at the surface lattice of the oxide, accompanied by the generation of halides. For example, chlorine detached from organic pollutants binds to Ca^{2+}, such as $CaCl_2$ (Hall et al., 1996; Zhang et al., 2010). After dehalogenation, high molecular weight inorganic carbon (graphite and amorphous carbon) is formed through reactions such as splicing, condensation, and dehydrogenation, which can effectively destroy organic pollutants (Hall et al., 1996; Song et al., 2019). In addition, Ca and Fe(III) oxides produce electrons at a faster rate than quartz and kaolinite (Hu et al., 2018), Upon ball milling, gaseous oxygen is released to form oxygen vacancies with the concomitant generation of free electrons:

$$O^{2-} \leftrightarrow V_O + 2e^- + \frac{1}{2}O_2\uparrow \tag{3}$$

Fe_2O_3 may be the source of active unpaired electrons and oxidizing species involved in electron transfer (Samara et al., 2016). Previous studies have also observed this reaction, including pyrene on Cu-montmorillonite (Joseph-Ezra et al., 2014b) and 2,4-dichlorophenol on manganese oxides (birnessite) (Nasser and Mingelgrin, 2014). There is also cations (e.g., Fe^{3+}) that oxidize organic molecules and together with oxygen vacancies initiate the destruction of organic contaminants (Samara et al., 2016). After the addition of Fe/Fe_3O_4 to the ball milling of HCB and hexachloroethane, the contaminants are finally mineralized through dechlorination and polymerization (Hu et al., 2019). One of the important functions of ball milling is to promote the contact between contaminants and mechanochemical activated components. Through a series of chemical reactions such as free-electron transfer, free radical action, and nucleophilic substitution, organic contaminants are finally completely mineralized. The mechanism of ball milling for the remediation of organically contaminated soils needs to be fully elucidated in the future. For instance, the scavenging of electrons and radicals by reagents was utilized to elucidate the remediation pathway.

5. Application of ball milling techniques for soil remediation

5.1. Remediation of inorganic contaminated soils

Heavy metals, metalloids, and nuclides accumulated in soils through the food chain cause many human health problems (Bian et al., 2013; Bolan et al., 2014). It is necessary to search for a remediation technique that uses few chemical reagents and simple operating conditions. Ball mill is an excellent immobilization technique. The bioavailability of soil inorganic contaminants can be minimized by using ball milling to remediate contaminated soils with an appropriate extension of the BMT after performing simple operations (Montinaro et al., 2008).

In general, ball milling provides sufficient energy to ground materials to promote physicochemical reactions, thereby activating soil components to immobilize metals, reaching the effect of remediation of contaminated soils. Montinaro et al. obtained about 100% immobilization of heavy metals (Cd, Pb, Zn, As, and Ni) after a certain time ball milling treatment, obtained the same remediation effect in simulated and real contaminated soils (Montinaro et al., 2009). Concas et al. used two leaching methods to corroborate that the ball milling treatment has achieved soil remediation in multi-metal contaminated mining areas (Concas et al., 2020a). Results show that the leaching concentrations of Al, As, Pb, and Hg was below the drinking water regulatory thresholds for the corresponding leaching methods after ball milling treatment. Changes in soil pH may cause heavy metals to be released again, however, ball milling can enhance the anti-acid ability of soils. After ball milling, the leaching concentrations of Pb, Cu, and Zn were reduced to 0.027 mg L^{-1}, 0.59 mg L^{-1}, and 0.16 mg L^{-1}, respectively, and their pH-dependent leaching tests indicated that the soil had improved anti-acid ability (Yuan et al., 2019b). Ball milling can also promote the transformation of heavy metals to more stable fractions, and the contents of the residual fraction gradually increase after prolonging the BMT, reducing the bioavailability of toxic metals. Yuan et al. observed that ball milling could promote the transformation of heavy metals in soils to reducible, oxidizable, and residual fractions, and the residual fractions increased from approximately 0.4%–13.3% after 4 h of ball milling (Yuan et al., 2019b).

Bioavailability, toxicity, and mobilization of inorganic pollutants in soils depend on their biogeochemical processes such as anions metal (loids) (e.g., Sb, Cr, and As). To reduce the toxicity of metals and promote the formation of residual fractions, co-milling reagents can be added, resulting in rapid and effective soil remediation. For example, Cr (VI) in soils exhibits higher mobility and solubility. Ball milling enables its simultaneous immobilization and reduction. With the addition of the Na_2S reductant in the ball milling test, ball milling increased the utilization of reducing reagents, but also shortened the time (several hours or less) to complete the remediation (Yuan et al., 2018). Likewise, $Ca(S_x)$ was also chosen as the co-milling reagent, and the Cr(VI) leaching

concentration was reduced to 0.51 mg L^{-1}, and the co-milling reagent could ensure long-term stability of the remediation (Yuan et al., 2019a). Mallampati et al. conducted practical remediation of radioactive Cs contaminated soils by separating magnetic soils after adding nano Fe/Ca/CaO/[PO$_4$] ball milling for 2 h, which resulted in the immobilization efficiency of ^{134}Cs and ^{137}Cs either in the magnetic or non-magnetic soil fractions reaching 100% (Mallampati et al., 2015). Pilot-scale applications of ball milling on inorganic contaminated soils have been developed. Concas et al. developed equipment capable of scale applications by amplifying filling ratios (Concas et al., 2020a). This equipment achieves the remediation of various inorganic pollutants in the soil of the mining area, and the leaching concentration of all heavy metals except Pb is below the regulatory limit. Thus, real-scale applications can be achieved by enhancing the remediation effects of soils using more optimal parameters of ball milling. In addition, Montinaro et al. proposed a schematic flowsheet for the real scale application of ball milling remediation technology (Montinaro et al., 2008). In terms of eco-environmental protection and economic cost, the ball milling method has the characteristics of low-risk and no waste liquid. Consequently, ball milling technology has a promising application prospect and potential in the remediation of heavy metals contaminated soils.

5.2. Remediation of organic contaminated soils

POPs in soils are cumulative and highly toxic to living organisms and even have carcinogenic, teratogenic, and mutagenic effects. They pose a serious threat to the environment and human health and require suitable methods to remediate contaminated soils, prevent their transfer into drinking water, and accumulate in crops and feeds. The ball milling method has obvious advantages in the transformation or mineralization of toxic organic pollutants, especially the difficult degradable organic pollutants in soils (Fan et al., 2020; Turner et al., 2021).

Organic contaminants in soils can be entrapped by soil minerals, which are not easily remediable. The ball milling method facilitates the desorption of organic contaminants in soils while degrading and mineralizing them by mechanochemistry. Therefore, ball milling can remediate soils polluted by substances like HCB. Fan et al. used ball milling to treat HCB-contaminated soil found that HCB was degraded by dechlorination, polymerization, and ring-opening (Fan et al., 2020). HCB formed PCBs upon release of chlorine atoms and occurrence of dimerization reaction, which finally transformed them into graphitic or amorphous carbon after prolonging the BMT. The effect of ball milling also causes HCB to undergo a ring-opening reaction to form alkanes or alkenes and carbon-oxygen compounds. It is visible that ball milling can degrade and mineralize pollutants such as HCB and PCBs in soils. In addition, the use of NaBH$_4$ in the ball milling treatment promoted reductive debromination, and the removal efficiency of deca-bromodiphenyl ether in contaminated soil was about 100% (Yi et al., 2021). Wang et al. successfully treated real petroleum-contaminated soil by ball milling, with the highest removal rate of total petroleum hydrocarbons reaching 95.9% (Wang et al., 2022). Additionally, the self-purification of soils can be enhanced by ball milling. The highly active soil components formed after ball milling can effectively degrade most organic pollutants and even completely mineralize to inorganic forms. Wang et al. confirmed that quartz and oxides (e.g., Fe$_2$O$_3$, Al$_2$O$_3$, and CaO) from soils exhibited higher reactivity after ball milling to dechlorinate PCBs and destroy the aromatic hydrocarbon skeleton, which reduced PCBs to the limiting value of 50 mg kg^{-1} (Wang et al., 2017). Four soil common components, CaO, SiO$_2$, Fe$_2$O$_3$, and kaolinite, were used as co-milling reagents, and glyphosate and chlorpyrifos in the soil were finally mineralized into safe inorganic form (Hu et al., 2018). In conclusion, ball milling for remediation of organically contaminated soils has a positive effect when co-milling reagents are not used.

Since the carbon-fluorine (C–F) bond is not easily disrupted in organic fluoride, soils contaminated by substances such as per- and polyfluoroalkyl substances (PFAS) are difficult to treat with conventional remediation techniques. While the ball milling method can achieve the complete defluorination of organic fluoride. Generally, PFAS destroyed starting from the head groups after the formation of free electrons and reactive radical species under ball milling and then promoted the degradation reaction of PFAS by nucleophilic attack on C–F or C–C bonds (often at the functional groups). Whether from PFOA contaminated soil, or PFOS compounds (defluorination by reductive reaction only), the ball milling method remediates them effectively. Ball milling of soil containing PFOA and PFOS with the simultaneous addition of KOH resulted in a PFOS destruction rate of 96% in clay after 6 h (Turner et al., 2021). The practical application of the ball milling method has been carried out in several fields for many years. As a promising technology, a variety of ball mill types and models are available on the market, including large trucks fitted with ball mills that can directly reach the site of contaminated soils (Rowlands et al., 1994). For example, Lu et al. used a multi-level high-energy ball milling for pilot-scale remediation of organic contaminated soils, and the destruction efficiency of PCBs and PBDEs exceeded 94% (Lu et al., 2017). Cagnetta et al. conducted on the design of a generic scale treatment plant and briefly discussed its configuration and cost (Cagnetta et al., 2018). The extension of ball milling remediation processes and their feasibility need to be further evaluated in future studies.

6. Conclusions and future perspectives

In this review, the research progress of the ball milling on various contaminated soils remediation has been summarized, basically focusing on the introduction of different ball milling types and their remediation effects, influencing factors, and removal mechanisms. Compared with horizontal ball milling, planetary ball milling has a better remediation effect on contaminated soils. The effect of ball milling on the decontamination of pollutants from soils can be greatly improved through optimizing the conditions of ball milling. Ball milling can be combined with other soil remediation techniques to achieve sustainable remediation outcomes, such as combining with thermal treatment, magnetic separation, or bioremediation to enhance the performance of ball milling. Nevertheless, the ball milling method still faces some problems and challenges regardless of the theoretical and practical application aspects, which warrant the following further research:

The first concern relates to the co-milling reagents. How to synthesize or introduce efficient and low-cost co-milling reagents, and reduce the residual co-milling reagents in the soil to decrease the effect on soil texture are issues to be considered. At present, the selection of industrial wastes (red mud, phosphorus gypsum, coal gangue, fly ash and tailing, etc.), nanomaterials, natural minerals, etc. as co-milling reagents, could be a future promising research direction.

The second issue is the environmental risk. Is it possible that some pollutants are generated during the process of ball milling? For example, polychlorinated dibenzo-para-dioxins and dibenzofurans as well as their corresponding polybromides may be unintentionally produced when using the ball milling to remediate soil containing chlorinated and brominated dioxin precursors (Lu et al., 2017). Currently, controlling the temperature through the cooling system, prolonging the BMT, or completely removing the halogen source may be some feasible methods to solve the problem. In the future, more feasible methods need to be developed to reduce the generation of potential pollutants during the ball milling process.

The third is on the optimization and expansion of the ball milling method. Currently, the study of ball milling conditions only performs single factor tests under a given few conditions. Therefore, it is necessary to further study the soil remediation by ball milling technology under more parameters and different conditions. In terms of expansion of the ball milling method, the removal of emerging pollutants from some soils could be a future development direction. Emerging pollutants are very complex organic substances, which often include antibiotics, endocrine disruptors, and caffeine, among others. It is suggested that the

combination of the experimental verification and the theoretical model in the future would not only reduce the number of tests but also obtain relatively better ball milling conditions in the larger research scope.

The fourth is the reuse of remediated soil. The main components of soil are clay minerals and metal oxides, which are considered excellent adsorbents. Many studies have used ball milling to enhance their adsorption performance. Therefore, soil obtained after successful remediation by ball milling can be used as an adsorbent material. For example, petroleum-contaminated soils are completely mineralized by ball milling to form ternary composites (including clay minerals/metal oxides/carbons) (Wang et al., 2022), which may be used as adsorbents for environmental applications. Contaminated soils can be green efficient remedied by ball milling and high-value reuse of remediated soils.

Fifth, the combined effects of the environmental factors are much more complicated under real environmental conditions. Therefore, there are still some immobilization and degradation or mineralization mechanisms induced by ball milling that need to be further clarified. Which factor plays the dominant role? How to evaluate the contribution? Elucidating the above issues is of particular importance for improving the remediation efficiency and environmental applications of the ball milling method. Future research priorities should revolve around the issues outlined above. In summary, the ball milling technology shows promising potential for research and development in soil remediation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Further reading

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Update

Journal of Cleaner Production

Corrigendum to "Recent advances in the treatment of contaminated soils by ball milling technology: Classification, mechanisms, and applications" [J. Clean. Prod. 340 (15 March 2022) 130821]

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The authors did use figures in their article that are similar in content to the figures that are used in figures in Cagnetta et al. (2018), CREST. The Editors recognize there is indeed overlap in the intellectual content, making the same points. Via this corrigendum, the editors would like to acknowledge the earlier article: Giovanni Cagnetta, Jun Huang & Gang Yu (2018) A mini-review on mechanochemical treatment of contaminated soil: From laboratory to large-scale.

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