Review

Detoxification of pesticide waste via activated carbon adsorption process

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A B S T R A C T

Concern about environmental protection has increased over the years from a global viewpoint. To date, the percolation of pesticide waste into the groundwater tables and aquifer systems remains an aesthetic issue towards the public health and food chain interference. With the renaissance of activated carbon, there has been a consistent growing interest in this research field. Confirming the assertion, this paper presents a state of art review of pesticide agrochemical practice, its fundamental characteristics, background studies and environmental implications. Moreover, the key advance of activated carbon adsorption, its major challenges together with the future expectation are summarized and discussed. Conclusively, the expanding of activated carbon adsorption represents a plausible and powerful circumstance, leading to the superior improvement of environmental preservation.

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Contents

1. Introduction

Water scarcity and pollution rank equal to climate change as the most intricate environmental turmoil for the 21st century. Over the past several decades, the exponential population and social civilization growth change affluent lifestyles and resources use, and continuing progress of the industrial and technologies has been accompanied by the sharp globalization and vast generation of agrochemical wastes, which create the most intransigent paradox abroad the nations [1]. In 1950, the world population was reported as 2.5 billion, whereas in 2000 (50 years later), the figure has risen to 9.1 billion, designated an elevation of 144% and by 2050, the record is predicted to be further strengthened, implying a steady growth of 1.5 billion [2]. Simultaneously, the departure of the pesticide practice, notably in the form of herbicides, insecticides,

fungicides, algaecides, antimicrobials, avicides, miticides, molluscicides, nematicides, rodenticides, bactericides, defoliants, piscicides and virucides, has prevailed to be a growing branch that protests against the invasive species which pose an enormous threat to the indigenous ecologies, and prevents against the transmission of diseases [3,4], thus enabling qualitative and quantitative assurance of the crops yield stability for fulfilling the pressing need of the rising food's demand [5].

In the early irrigation, it was a common practice to dispose agrochemical refuse by uncontrolled tipping or dumping, an operation in which waste is spread over the estates ground or tipped to fill in low economic value open dumps on selected pieces of land (inundated swampland, abandoned sand mines and quarries), without taking care of the surrounding environment, nor considering any precautions to compact, cover and prohibit the percolation of contaminants into the underlying waterways [6]. Lately, the enforcement of environmental rules and regulations concerning the monitoring of pollution from agrochemical effluent waste streams by regulatory agencies is becoming more stringent and restrictive, inevitably affects the design, planning, and operation of

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the agricultural industry [7]. This has inspired a growing research interest in establishing a leading selective, reliable and durable alternative for judicious treatment of the heavily polluted pesticides waste. With the aforementioned, this bibliographic review attempts to postulate an initial platform in describing the origin, distinct physiochemical properties, development and environmental impacts of the pesticide waste. The present work is aimed at providing a concise and up-to-date picture of the present status of the pesticide waste treatment technologies (activated carbon and its advanced integrated technologies). The comprehensive literature together with the challenges and future perspectives has been highlighted and outlined, to familiarize the knowledge deficiencies regarding pesticide waste treatment via activated carbon adsorption process.

2. Definition, classification and historical study of the pesticide waste

In general, pesticide is defined as a diverse group or mixture of chemical substances, biological agents, antimicrobials, disinfectants or devices which are intentionally applied for selective administration and attenuation against any pests including insects, plant pathogens, weeds, molluscs, birds, mammals, fish, nematodes (roundworms) and microbes that compete for the production, processing, storage, transport, or marketing of food, agricultural commodes, destroy property, or widespread of diseases [8–10]. More precisely, pesticide is featured by its unique chemical structures developed to mimic and substitute for specific molecules to the targeted pests, which present a deleterious disruption to the desired biological reactions [7]. The classification of pesticide substance is usually categorized to its physiological chemical classes (Fig. 1) or its targeted implications, with insecticide is corroborated to the protection of termites, ants, mosquitoes, and cockroaches while herbicides and rodenticides are linked to the weeds and rat infestations control (Table 1).

Ample ancient historical documents, the early concept regarding pesticide practice (utilizing sulphur as fumigant) has been initialed by Chinese during the 2500 B.C. in Sumeria [11], which in the 16th and 17th centuries, the invention of mixed whale oil–vinegar hindering the development of insect larvae (by weakening the cuticle) in Japan and combat between nicotine extracts from tobacco leaves, pyrethrum, lime and soap against insects and fungi in France has been witnessed [7]. Until the turn of 19th century, rotenone and pyrethrum were individually isolated from the root of *Derris eliptica* (tropical vegetables) and the flowers of chrysanthemums as organic insecticides, while the spread of the Colorado beetle was battled through an impure form of copper arsenite (Paris green) in the United States [12].
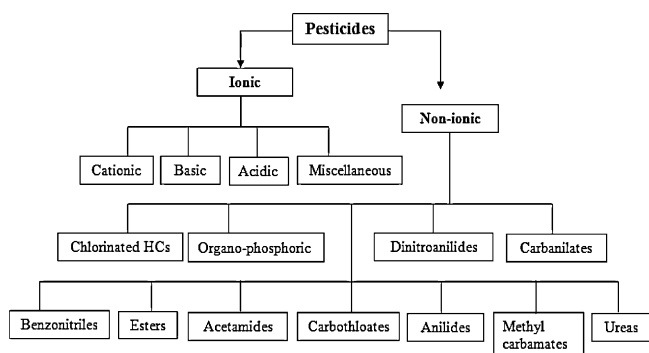

Fig. 1. The classification of pesticides on the basis of the significant physicochemical properties and behavior in water and soil [7].

The modern chemical age of pesticides began with the discovery of the insecticidal potential of dichlorodiphenyl trichloroethane (DDT) in 1939 by Paul Müller in Switzerland and the development of organophosphorus insecticides in Germany [13], which in 1945, the first soil-acting carbamate herbicides were identified in the United Kingdom and simultaneously, the organochlorine insecticide chlordane was introduced in Germany and the United States [14]. Towards the end of 1960s, the vigor of the pesticide practice has been well recognized and entrenched into both scientific literature and pest control with the embraced failings of the new organosynthetic insecticides [15,16].

Till the early 1980s, debates about the aggressiveness and improper use, manufacture, import, export and application of pesticide technologies among the world international organizations, national governments and industries have intensified the formulation of the international code of conduct and use of pesticides (code of conduct), exclusively governed by the Food and Agriculture Organization (FAO) of the United Nations (amended once in 1989 to include the prior informed consent procedure, and subsequently revised in 2002) [17,18]. Meanwhile, the early (1940s) pesticides production rate was estimated as 600,000 tonnes/year and today, its utilization has been vigorously widen, achieving an annual generation of 3 million tonnes/year (50-fold), corresponding to a market value of US$ 15,900 million [10].

3. Emergence of pesticide waste towards the public health and environmental impacts

Within the last few years, intensive wide spread contamination of atmosphere and surface water related to adverse industrial operations has attested a fastidious concern for many environmentalists. Recently, the infiltration of pesticide effluents into the waterways and aquifer systems, endorsed by the spray drift, soil leaching, tile drainage outflow, base-flow seepage, surface run-off, volatilization (diffuse-source inputs), sewer overflows, improper handling of tank mix leftovers, leaking of faulty equipments, incorrect storage of canisters, dripping from agricultural practices, accidental spillage and farmyard deposition (point source pathways) (from agriculture, horticulture, forestry, chemical industry and domestic activities) [19], constitutes an accumulative, persistent, threatening and detrimental deterioration towards the survival of aquatic compartments, flora, fauna and environmental matrix (water and soil) [20].

The real hazard setting aside aesthetic considerations is exacerbated when pesticide agents interfere with the transmission of light through water, retard photosynthetic activities, inhibit the growth of biota terrestrial ecosystems (carotenoid, aromatic and branched chain amino acid synthesis), affect the symbiotic process, damage the quality of the irrigation channels and impede the propagation of algae, thus upsetting biological processes within streams, resulting in food chains and ecological imbalance [13,21]. Increasingly, acute exposure to pesticide substances was illustrated vulnerable to a wide spectrum of immunosuppression (favorably among low-income populations, malnutrition and unsanitary conditions), neurobehavioral disorders and developmental toxicity presage as allergy, autoimmune disease, reproductive abnormalities (spermatogenetic dysfunction, mal-descent of testes and malformations of penis), multiple myeloma, leukemia, malignant lymphoma, increased kit mortality, deformities, life-threatening bleeding and myocytes with burning sensation in the mouth and throat, nausea, vomiting, sweating, hyperventilation, pain, fasciculation, myotonia, weakness and myoglobinuria [9,22,23].

Nonetheless, sporadic and excessive susceptibility to the pesticides clastogenic action has been subjected to the alterations of cardiovascular system, musculoskeletal defects and thyroid func-

Table 1
The classification of pesticide according to its targeted implications [7].

Type	Implications
Algaecides	Control algae in bodies of water, including swimming pools.
Antimicrobials	Kill microorganisms that produce disease.
Attractants	Attract specific pests using natural insect chemicals called pheromones that confuse the mating behavior of insects.
Avicides	Control pest birds.
Bio-pesticides	Naturally occurring substances with pesticidal properties.
Defoliants	Cause foliage to drop from a plant, typically to aid in harvesting process.
Desicccants	Aid in drying process of plants or insects, usually for laboratory purposes.
Fumigants	Produce vapors or gases to control air or soil-borne insects and diseases.
Fungicides	Destroy fungi that infect people, plants, and animals.
Herbicides	Control noxious weeds and other vegetation that are growing or completing with a desired species.
Insect Growth Regulators (IGRs)	Accelerate or retard the rate of growth of insects.
Insecticides	Control or eliminate insects that affect people, plants, and animals.
Miticide (Acaricides)	Kill mites that live on plants, livestock, and people.
Molluscicides	Kill snails and slugs.
Nematicide	Kill nematodes, which are microscopic wormlike organisms that live in the soil and cause damage to food crops.
Ovicides	Control insect eggs through the application of low-sulphur petroleum oils to plants and animals.
Piscicides	Control pest fish.
Plant Growth Regulators (PGRs)	Accelerate or retard the rate of growth of plant.
Predacides	Control vertebrate pests.
Repellents	Repel pests such as mosquitoes, flies, ticks, and fleas.
Rodenticides	Control mice, rats, and other rodents.

Table 2
United Nations criteria for grouping substances according to toxicity [27].

United Nations packaging group	Oral toxicityLD_{50} (mg/kg)	Dermal toxicityLD_{50} (mg/kg)	Inhalational toxicity by dusts and mists LC_{50} (mg/L) (1 h)
I	>5	>40	<0.5
II	5–50	40–200	0.5–2
III (solids)	50–200	200–1000	2–10
III (liquids)	50–500		

tion failure (acetanilide, ethylene bisdithiocarbamates, nitroanilines, organophosphates and synthetic pyrethroids) ranging from predomination of neuromuscular complaints (lethargy, irritability, poor concentration, mood swings, depression, insomnia, muscle aches and pains), hyper-stimulation of parasympathetic autonomic nervous system, profuse diarrhea, salivation, urination, lacrimation, dyspnea, wheezing (muscarinic receptors), bradycardia, tremors, convulsions, respiratory paralysis (bronchial spasm and tonic convulsions), lung edema to even death [24–26]. Tables 2 and 3 outline a typical framework for the classification of pesticides on the basis of its toxicity level, and degree of hazard control with its user groups. In the perspective, parental pesticide poisoning (clinical studies conducted by the International Agency for Cancer Research) was significantly associated to its anthropogenic carcinogenic, mutagenic and genotoxic nature with the stimulation of congenital malformations, diminished intelligence, genetic syndromes (mostly Turner's syndrome), and deoxyribonucleic acid (DNA) (or chromosome) damages [24,28,29].

4. Pesticide waste treatment via activated carbon adsorption and its integrated technologies

Throughout recent decades, the emergence of pesticide practice as a viable agricultural repellant, synergist, fumigant, defoliant, desiccant or sterilant, is interpreted as one of the most challenging tasks, which has deeply embedded with huge momentum and popularity around the world [30]. A developing research by the invention of a wide variety of treatment technologies (precipitation, coagulation–flocculation, sedimentation, flotation, filtration, membrane processes, electrochemical techniques, biological process, chemical reactions, adsorption and ion exchange) with varying levels of success has accelerated a dramatic progress in the scientific community [31–37] (Table 4). Of major interest, adsorption process, a surface phenomenon by which a multi-components fluid (gas or liquid) mixture is attracted to the surface of a solid adsorbent and forms attachments via physical or chemical bonds, is recognized as the most efficient and promising fundamental approach in the wastewater treatment processes [38].

A notable trend in the development of activated carbon, an adsorbent with its large porous surface area, controllable pore structure, thermo-stability and low acid/base reactivity has been witnessed [39], mainly hinges on its superior ability for removing a broad type of organic and inorganic pollutants dissolved in aqueous media, even from gaseous environment [40]. In particular, the pioneering works regarding the recycling and polishing of wastewater comprising trace quantities of isopropyl-N-(3-chlorophenyl) carbamate have been firstly advocated by Schwartz in 1967, signifying a maximum adsorption capacity of 334 mg/g corresponding to the

Table 3
The use and classification in the control of supply and use of pesticides [27].

Class	Simple criteriaAcute oral LD_{50} (mg/kg)	Degree of hazard and control	User group
I	<20	Severe occupational hazard. Permit required.	Professional pesticide applicator.
II	20–200	High occupational hazard. Control sales.	As above plus licensed farmers.
III	200–2000	Moderate occupational hazard. General sales.	General farmer use.
IV	>2000	Slight hazard.	General pubic home and garden use.

Table 4
Technical advantages and drawbacks of existing pesticide treatment technologies.

Treatment	Advantages	Drawbacks
Fenton process	Rapid decomposition of organic and inorganic pesticides (high removal rates). Ability in removing a broad range of pesticides (biodegradable and non-biodegradable). Ease of handling. Low operational cost.	Production of residual sludge. Require large usage of chemicals reagents. Mismanagement can lead to accumulation of ion particles or free radicals.
Biological treatment	Capable for treating a wide range of pesticides which cannot be treated by chemical or membrane technologies.	Require large surface areas for implantation of treatment and biomass separation units. Hampered by the presence of bio-refractory organics (humic substance or surfactants).
		Limited suitability in treating stabilized pesticide (less biodegradable) due to the recalcitrant characteristics of its organic carbon. Low digestion rates (days or weeks). Require suitable cultured condition and constantly fed bacteria.
Advanced oxidation treatment	Particularly useful for cleaning biologically toxic or non-degradable pesticides. Non-selective pathway allowing treatment of multiple pesticides in a single step. Accelerating pesticides removal via solar, UV–vis-rays or ultrasonic radiation.	Deteriorated by potentiality of chlorine oxidation, resulting in the formation of chlorine or hypochlorite. Poor economic acceptability for large-scaling processes. Extra energy sources (solar, UV–vis-rays or ultrasonic) are involved.
Membrane technologies	Operated without phase changes or chemical conditioning. Only low energy consumption (pressure) is needed.	Subjected to the fouling effect by a wide spectrum of constituents (resulting in short lifetime of membranes and decreases of process productivity). Huge generation volume of concentrate (which is unusable and require further treatments).
Ion exchange treatment	Exhausted resins are regenerable. Low operational cost. Low energy consumption.	Different resins are fabricated to selectively remove preferable types of pesticides. Improper for treating chlorinated pesticides. The presence of microorganisms can reduce the exchange capacity (irreversible) leading to a reduction in treated water quality. Disposal of the treated pesticides is usually a problem to the loaded waste treatment systems (after the regeneration step).

nominal activation energy of 3.4 kcal/mole [41]. Seven years later, similar research has been advanced by Parkash [42] with varying dosage of powder activated carbons in an agitating system, accomplishing a peak removal of 36% (paraquat). The extent of effort has proliferated for the next 35 years, and today, a variety of scientific publications and its manifestations have been widely developed (Tables 5 and 6) (Figs. 2 and 3).

In 2004, Ayranci and Hoda [50] have endeavored to abound a tentative study for the treatment of ametryn, aldicarb, dinoseb and diuron using a commercial activated carbon cloth, denoting a competent removal of 85% with an adsorption capacity 421.58 mg/g.

Dispersive interaction between the delocalized π electrons in the pesticide structure and carbon cloth was presumed to be the primary attraction force, ascribed to the attendance of branched alkyl substituent and aromatic rings which instigated an aggressive hydrophobicity across the surfaces [30]. pH reduction augmented protonation of acidic groups along the edges of graphitic layers and facilitated minimization of the electrostatic energy, thereby resulting in a drastic rise of the adsorption capacity [59]. Relatively, a substantial investigation has been undertaken to examine the prominent role of natural organic matters (NOM) in assessing an activated carbon adsorption system (by spiking water containing
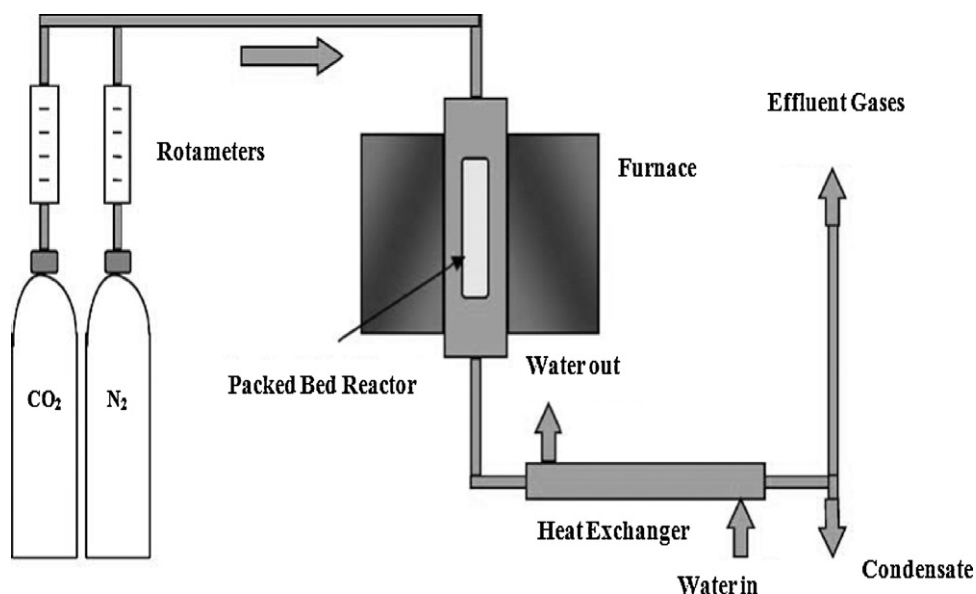

Fig. 2. Schematic diagram of pyrolysis and activation process for the preparation of activated carbon derived from used tires in the adsorption of paraquat dichloride [49].

Table 5
List of researches for the pesticide treatment via activated carbon adsorption process during the last 40 years.

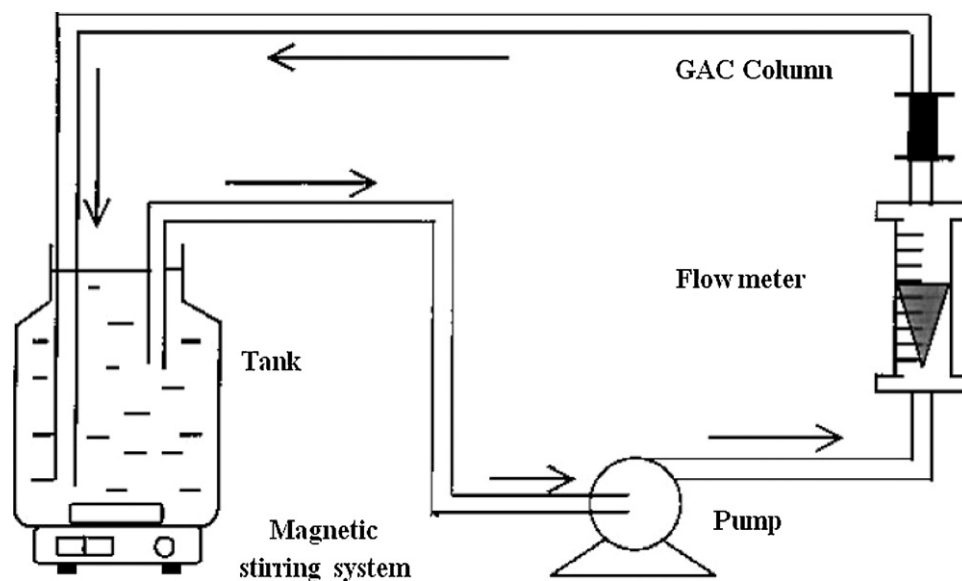| Activated carbon type/precursor	Pesticide type	Maximum adsorption capacity (mg/g)	Percentage removal (%)	Reference
Filtrasorb 400 (commercial GAC)	2,4-Dichlorophenoxyacetic acid	411.13	–	[5]
	2-Methyl-4-chlorophenoxyacetic acid	389.20		
	Chlorophenoxyacetic acid	516.85		
Spectracarb 2225 (commercial activated carbon cloth)	Ametryn	354.61	–	[30]
	Aldicarb	421.58		
	Dinoseb	301.84		
	Diuron	213.06		
Nuchar C-190 (commercial PAC)	Isopropyl-N-(3-chlorophenyl) carbamate	334	–	[41]
PAC 313W (commercial)	Lindane	412.0	–	[43]
Charcoal based PAC (commercial)	4-(4-Chloro-o-tolyloxy)butyric acid	125.77	–	[44]
	Imazalil	208.03		
Filtrasorb 400 (commercial GAC)	Lindane	181	–	[45]
Filtrasorb 400 (commercial GAC)	Lindane	183	–	[46]
	Alachlor	151		
Wood-based PAC (commercial)	Asulam	–	80	[47]
	Simazine		90	
	Simetryn		100	
Darco G-60 (commercial AC)	Bromoxynil	500	–	[48]
	Diuron	300		
Filtrasorb 400 (commercial GAC)	Paraquat dichloride	68.0	–	[49]
Used tires	Paraquat dichloride	27.8		
Spectracarb 2225 (commercial activated carbon cloth)	Bentazon	151	–	[50]
	Propanil	114		
Activated carbon fiber (commercial)	Atrazine	238.1	–	[51]
	Simazine	370.4		
Commercial PAC	Carbofuran	190	–	[52]
Powder activated charcoal (commercial)	Isoproturon	104.21	–	[53]
Activated carbon (commercial)	Atrazine	0.356	–	[54]
	Propoxur	0.240		
	Methidathion	0.456		
Sewage sludge	Triadimenol	111.80	–	[55]
Date stones	2,4-Dichlorophenoxyacetic acid	238.10	91.83	[56]
NFEN 12915 (commercial GAC)	Metribuzin	210.00	–	[57]

Fig. 3. Differential batch column reactor for the adsorption of pesticides atrazine, bromoxynil and diuron [58].

Table 6
Isotherm and kinetic studies of pesticide treatment via activated carbon adsorption process.

Activated carbon type/precursor	Pesticide type	Applicable isotherm models	Applicable kinetic models	Reference
Filtrasorb 400 (commercial GAC)	2,4-Dichlorophenoxyacetic acid	Langmuir	Pore diffusion	[5]
	2-Methyl-4-chlorophenoxyacetic acid	Sips	Surface diffusion	
	Chlorophenoxyacetic acid			
Spectracarb 2225 (commercial activated carbon cloth)	Ametryn	Freundlich	Pseudo-first order	[30]
	Aldicarb	Langmuir	Pseudo-second order	
	Dinoseb			
	Diuron			
Nuchar C-190 (commercial PAC)	Isopropyl-N-(3-chlorophenyl) carbamate	Freundlich	–	[41]
PAC 313W (commercial)	Lindane	Freundlich	–	[43]
		Langmuir		
Charcoal based PAC (commercial)	4-(4-Chloro-o-tolyloxy) butyric acid	Freundlich	–	[44]
	Imazalil			
Filtrasorb 400 (commercial GAC)	Lindane	Dubinin–Radushkevich	Surface diffusion	[45]
		Radke–Prausnitz		
Filtrasorb 400 (commercial GAC)	Lindane	Freundlich	Homogeneous pore diffusion	[46]
	Alachlor			
Filtrasorb 400 (commercial GAC) Used tires	Paraquat dichloride	Langmuir	Pseudo-second order	[49]
Spectracarb 2225 (commercial activated carbon cloth)	Bentazon	Langmuir	Pseudo-first order	[50]
	Propanil	Freundlich		
Activated carbon fiber (commercial)	Atrazine	Freundlich	Surface diffusion	[51]
	Simazine			
Commercial PAC	Carbofuran	Freundlich	–	[52]
Powder activated charcoal (commercial)	Isoproturon	Langmuir	–	[53]
		Freundlich		
Sewage sludge	Triadimenol	BET	–	[55]
Date stones	2,4-Dichlorophenoxyacetic acid	Langmuir	Pseudo-first order	[56]

natural organic matters into the adsorption streams) [47,60,61]. Result anticipated an adverse depression of the adsorption capacity, attributed to the changes of the surface polarity distribution, displacement effect, pore blockage, pore constriction and direct competition for the adsorption sites (Fig. 4).

In Jordan, a comparative evaluation encompassing multi-walled carbon nanotubes, activated carbons and silica for discrimination of atra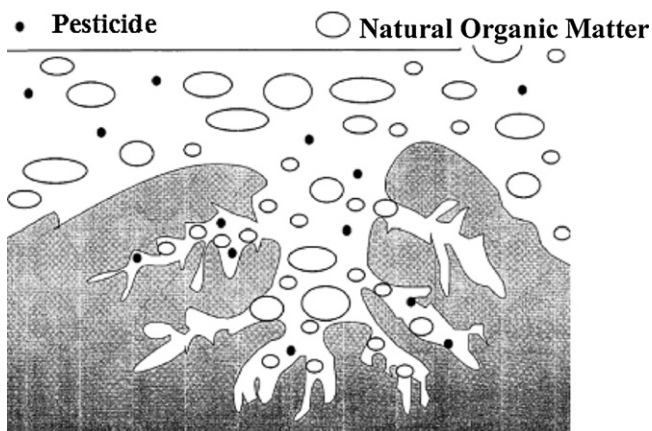zine, propoxur and methidathion in the multi-residue solid phase extraction process has been performed by El-Sheikh et al. [54]. Activated carbons appeared to be the most versatile and suitable candidate, owning to its low initial cost, simplicity of design, insensitivity to toxic substances, high adsorption capability and regenerability. In the case of a binary or multi-components system, adsorption capacities of each individual pesticide were noted proportionally less with another, indicating the existence of competition for available sites, with non-equal adversary to the heterogeneity of carbon surface. Accordingly, pesticide with the greatest ionic potential saturation value may effectively occupy most of the active sites hindering further adsorption of other pesticides by possessing a lower affinity [51].

Coinciding in coping the temporal fluctuations in varying strengths and compositions of pesticide waste, and ameliorating the single step adsorption process, recently, the development of collaborated multistage treatments, which combine adsorption process with numerous complementary approaches have received extensive stern attention and encourages worldwide. As early as 1995, the first attempt inoculating *Rhodococcus rhodochrous* and *Acenetobacter junji* bacteria in the granular activated carbon system has been proposed by Jones et al. [62] in a pilot study, suggesting a maximum adsorption capacity of 58.9 μg/g with an initial influent concentration of 20 μg/L. Subsequently, a separated coagulation–adsorption and coagulation–adsorption–nanofiltration treatment has been regulated by Kouras et al. [43] and Sarkar et al. [53], for the selective removal of pesticide lindane and isoproturon from the contami-
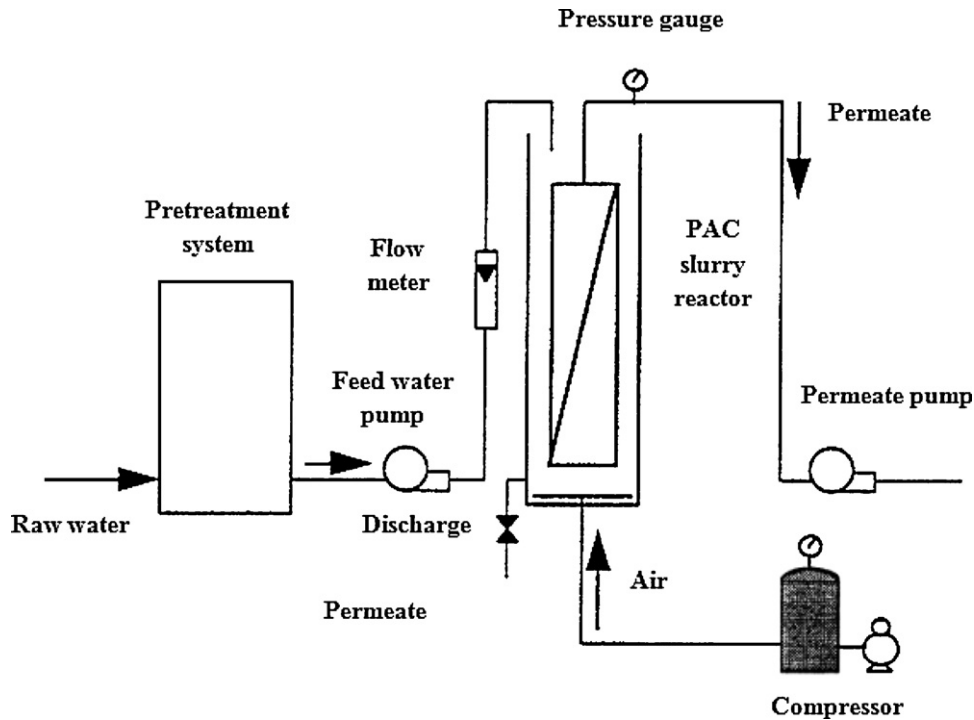

Fig. 4. Schematic presentation of the impact of natural organic matter on pesticide adsorption [61].

Fig. 5. Powder activated carbon/microfiltration pilot systems for treating of atrazine [64].

nated lake and river water. An apparent performance enhancement was exhibited; provoked by the formation of metal complexes (with coagulants ferric chloride and basic polyaluminium chloro-sulphate), reduction of repulsive electrostatic forces and removal of larger fulvic acid molecules (that cannot penetrate the porous carbon) [63]. In the same vein, a series of powdered activated carbon-microfiltration (Fig. 5), up-flow anaerobic sludge blanket-activated carbon, ion exchange resin-powdered activated carbon and oxidation-activated carbon auxiliary system has been individually conducted by Lebeau et al. [64], Ghosh et al. [65], Humbert et

al. [66] and Ormad et al. [67], elucidating a successive improvement of pesticide uptake rate.

Explicitly, incorporation of electrochemical approach, electrosorption, an attractive combination of the high adsorption capacity and electrical conductivity of the graphitic carbons, primarily operated in the presence of an applied low direct current voltages without high-pressure pumps, thermal heaters, membranes, distillation columns or chemicals, has embarked a huge revolution of the pesticide treatment industry (Fig. 6) [68]. Schematic representative of the possible adsorption sites of ben-
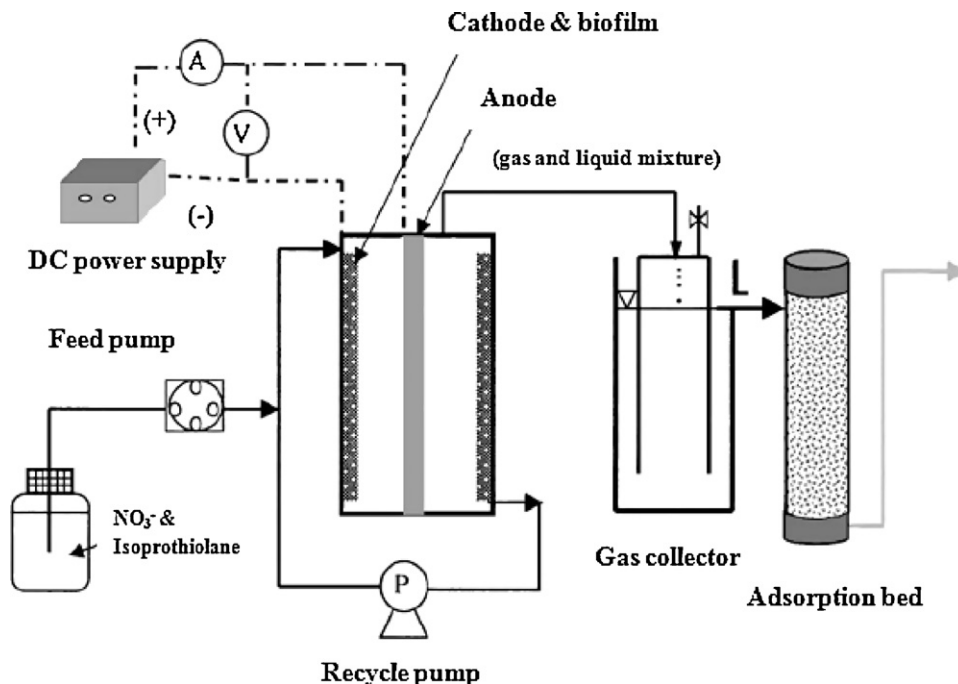

Fig. 6. Schematic of experimental apparatus for electrochemical reactor coupled with activated carbon adsorber for the removal of nitrate and inhibitory pesticide [69].
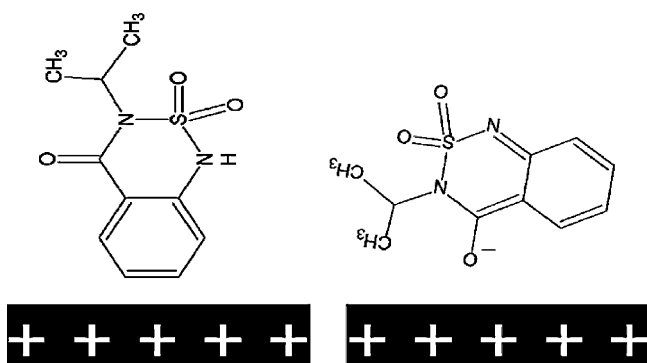
Fig. 7. Schematic representation of possible adsorption sites of bentazone onto activated carbon cloth under anodic polarization: (a) through aromatic ring and (b) ionic interaction through negative charge on the bentazone molecule [59].

Fig. 9. Fourier Transformed Infra-red (FTIR) spectrum of the activated sludge derived activated carbon with sulphuric acid activation [55].

tazone onto activated carbon cloth under anodic polarization is shown in Fig. 7. In contrast to activated carbon adsorption processes, preliminary studies have revealed its promising role in escalating the extents of impurities removal and possibility of elevating adsorption rate or capacity by manipulating the interfacial potential of the conductive electrodes (activated carbon) [70,71]. Additionally, it provides a convenient means of controlling adsorption and desorption processes, riding a direction for in situ restoration of the exhausted activated carbon into the purge stream through reversed potential [72].

Morphological studies of the date stone prepared activated carbon, waste tire derived activated carbon, rayon based activated carbon fibers and commercial activated carbon fibers (Kuraray FT-20) are presented in Fig. 8(a), (b), (c) and (d) respectively.

Typically, these textural structures are characterized by a well-developed mesoporous graphite layer in nature, with spaces between t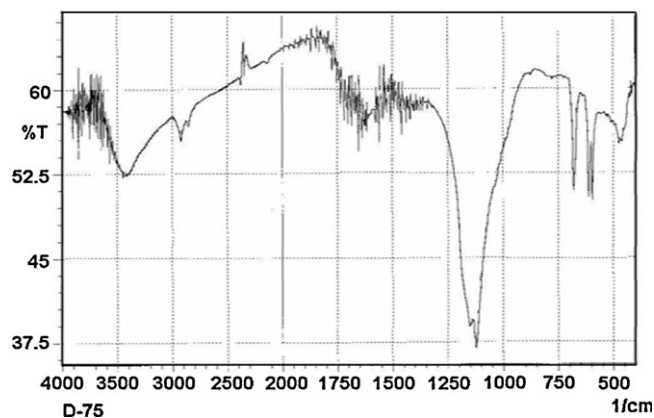he carbon layer planes and gaps between the stacks forming an interconnected network of slit-shaped pores, of which its surface chemistry is ministered strongly by the heteroatoms (hydrogen, oxygen, nitrogen, sulphur and phosphorus) bonded at the edges of aromatic sheets, or adopted within the carbon matrix along the heterocyclic ring system.

Fig. 9 exhibits Fourier Transformed Infra-red (FTIR) spectrum of the activated sludge derived activated carbon with sulphuric acid activation. In most cases, there are four major adsorption bands at 2900–3500, 1300–1750, 1000–1250 and 450–750 cm^{-1}. The region between 3200 and 3650 cm^{-1} is related to the hydrogen-bonded –OH groups of alcohols and phenols on the adsorbent surface and an intensive peak at 3450 cm^{-1}, as well as at band 2930 cm^{-1}, is assigned to the free hydroxyl groups (or adsorbed water molecules) and C–H interactions within the carbon surfaces [73]. Similarly, the presence of amides functional groups, in the form of ammonia (peak 1640 cm^{-1}) and primary amines (peak
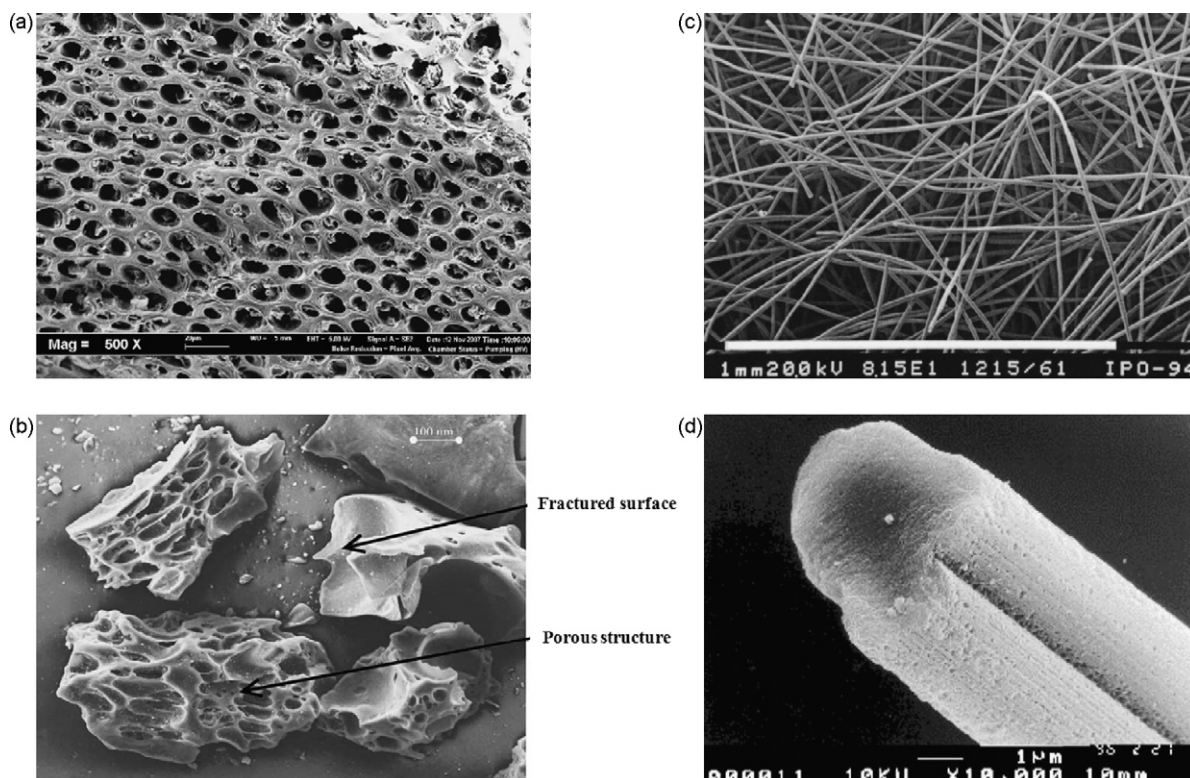

Fig. 8. Scanning electron micrographs (SEM) of the date stone prepared activated carbon (a) [56], waste tire derived activated carbon (b) [49], rayon based activated carbon fibers (c) [61] and commercial activated carbon fibers (Kuraray FT-20) [51].

$1450 \, cm^{-1}$), shows a signal between 1300 and $1750 \, cm^{-1}$ and peak around $1500 \, cm^{-1}$ is an indication of the aromatic carbon groups. Meanwhile, the IR spectra indicated weak and broad peaks about $1125 \, cm^{-1}$ corresponding to the Si–O and C–O groups stretching from alcohols, ethers to hydroxyls, and the transmittance in the $450–750 \, cm^{-1}$ region is ascribed to the vibration of in-plane and out-of-plane aromatic ring deformation, with peaks at 598 and $680 \, cm^{-1}$ are associated to the out-of-plane C–H bending mode (due to alkaline groups of cyclic ketones and their derivatives added during activation), while a band at $1150 \, cm^{-1}$ may be attributed to the anti-symmetrical Si–O–Si and ether C–O symmetric (or asymmetric) structures, resulting from the existing alumina and silica containing minerals within the adsorbents samples [74,75].

Fig. 10 displays X-ray diffraction (XRD) analysis of the waste tire [Fig. 10(a)] and activated sludge [Fig. 10(b)] derived activated carbons, reflecting amorphous structures of the waste tire and quantitative presence of crystalline minerals onto the activated sludge, induced by vaporization, melting, crystallization, vitrification, condensation and precipitation processes upon the pyrolysis stage [49,55]. In this respect, Zeolite X–Y was found at an intensity of 158 cps at $2\theta = 29.4°$, which a marginal amount of faujasite, mullite, hematite, quartz, sodalite, analcime and sodium silicates was detected at $2\theta = 26.5°, 32.9°, 35.9°, 39.4°, 43.1°, 47.5°$ and $48.5°$ respectively. Acids treatment elucidated a rapid mineral leaching effect while neutralization with sodium hydroxide was coupled with the concurrent reaction of alumina–silicates as described by Eqs. (1) and (2) (in the yielding of zeolite) [76].
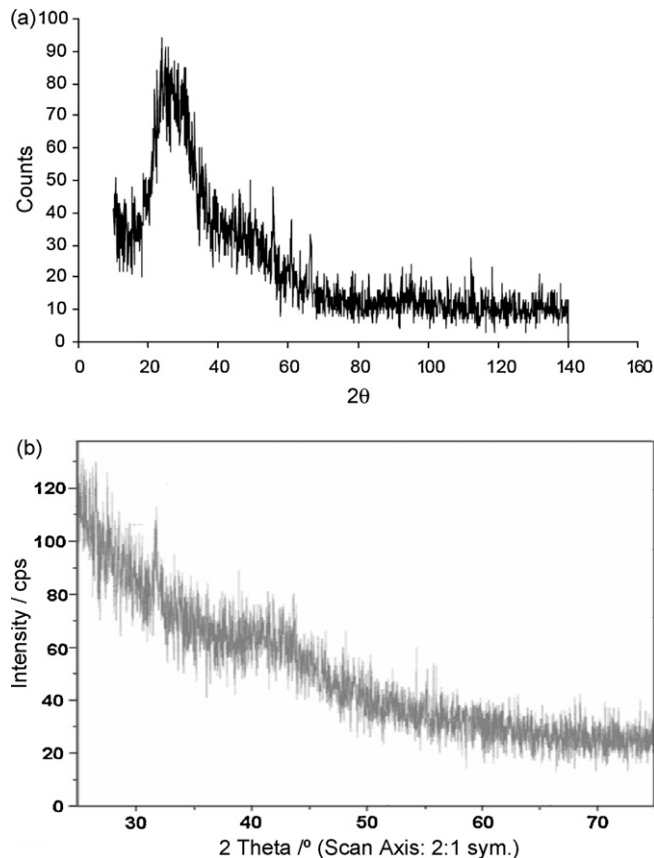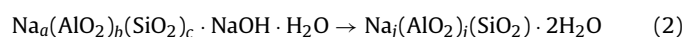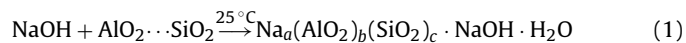
$$NaOH + AlO_2 \cdots SiO_2 \xrightarrow{25\,°C} Na_a(AlO_2)_b(SiO_2)_c \cdot NaOH \cdot H_2O \qquad (1)$$

$$Na_a(AlO_2)_b(SiO_2)_c \cdot NaOH \cdot H_2O \rightarrow Na_j(AlO_2)_j(SiO_2)_j \cdot 2H_2O \qquad (2)$$

Fig. 10. X-ray diffraction (XRD) analysis of the waste tire (a) [49] and activated sludge (b) derived activated carbons [55].

Accordingly, in 2009, Rietveld and de Vet [77] have properly alleged a conceptual and stimulation modeling study (granular activated carbon filtration in a pseudo-moving-bed configuration, where two filters are operated in series) which incorporates all essential phenomenological operative complexes and unidentified background interferences, for accurate evaluation of input parameters and better economic appraisal of the technology. The model was calibrated and validated on data of the pilot plants and bench scale applications, and evidenced more flexible with varying influent concentrations, effluent quality, reactor sizes, or different operation modes of the reactors, thus uplifting the reliability and efficiency of the system design.

5. Major challenges and future prospects

The world is currently facing the worst environmental crisis in its entire history. For the past two decades, the enthusiasm of agrochemical waste production and the terminology of deliberate pesticide exposure have been one of the rapidly advancing dilemmas, which have focused greatest public considerations and spectacular attention towards the recovery of contamination resources. Arising from the steep development of giant plantations and growing food demand in the Indonesia, India and China [78], numerous mitigating tactics and imperative technologies have drastically been addressed and confronted to utilize activated carbon (or its integrated technologies) contemplated mainly for pesticide waste treatment, in accruing worldwide environmental benefit and shaping the national economy [6,79]. Although there have been some successful industrial-scale applications and implications, generally the industry is still facing various constrictions, the availability of economically viable technology, sophisticated and sustainable natural resources management (cost-prohibitive adsorbent and difficulties associated with regeneration), and proper market strategies under competitive markets.

Amidst these shortcomings, the developing exploration in evaluating the adequacy and suitability of natural, renewable and low cost materials (palm shell, pall fiber, palm stone, bamboo dust, peat, chitosan, lignite, fungi, moss, bark husk, chitin, coir pith, maize cob, pinewood sawdust, rice husk, sugar cane bagasse, tea leaves and sago waste) as alternative precursors has been executed tremendously [80,81]. Fluctuating upon the alterations of time, place and context, environmental effectiveness, technological feasibility, social acceptability and economical affordability (chemicals, energy consumption, treatment facilities, labor, transportation, collection and maintenance) are usually the key drivers deciding its capability, reliability and sustainable manner. Parallel to the central principles of waste management hierarchy, the paradigm shift of individual and groups recycling, recovering, reuse and reduction (quantity, weight, volume and toxicity) throughout the waste chain has seen a panacea and new menu to the waste minimization strategy [6].

Accordingly, the urgency of conceiving and administering of strategic, corrective and transparent policies, mandates and standards governing the collection, transportation, disposal, prevention, recycling, monitoring, designing and supervision of the pesticide waste management ought to be prudently pointed out and well-planned. Increasingly, the sound professional knowledge of creating environmental awareness for adequate financial provisions, engineering and operating standards, responsibilities sharing, public participation, regular opinion survey, site rehabilitation and aftercare maintenance needs to be properly assigned and counteracted [82,83].

In Europe, an initiative on persistent organic pollutants within the framework of Convention on Long Range Transport of Air Pollution (CLRTAP) and Global Stockholm Convention has been

established in June 1998 and May 2001 [22], while several North American Regional Action Plans (NARAPs) under the North American Free Trade Area (NAFTA) have been declared for virtual handling, control and vigilant destruction of polychlorinated biphenyls throughout their life cycles, with special emphasis given to its trans-boundary shipment [7]. Moreover, the Safe Drinking Water Act 1986, Convention Concerning Safety in the Use of Chemicals at Work, Convention on Biological Diversity and the Agenda 21st of the United Nations Conference on Environment and Development have been emended as an early warning and notification towards the requirement of pesticide hazards limit, prior approval for pesticide recipient country and to promulgate maximum contaminant levels (MCLs) for several water-soluble pesticides within three years from the date of enactment [84,85]. Ultimately, full co-operation and joint venture between different parties (nations, states, local government, private sector and communities) from upstream till the bottom line with compatible technologies is a directive motivation for the race to the end line.

6. Conclusion

Over the years, the world's accessibility food supply is gradually depleting, driving towards the overwhelming researches dealing with agrochemical implementations. Predictions for the next 20 years indicate an ascending trend in the pesticide practice and, subsequently in pesticide waste generation. Today, the growing discrepancy and limited success of remediation in field applications has raised apprehensions over the use of activated carbon (or its integrated technologies) as a measure to the environmental pollution control. The evolution has turned from an interesting alternative approach into a powerful standard technique by offering a number of advantages. Although it is still in the infancy, a widespread and great progress in this area can be expected in the future.

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