Desalination and Water Treatment
www.deswater.com

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doi: 10.1080/19443994.2012.664680

41 (2012) 72–78
March

Taylor & Francis
Taylor & Francis Group

Potential of activated carbon adsorption processes for the remediation of nuclear effluents: a recent literature

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Received 14 December 2010; Accepted 24 January 2012

ABSTRACT

Stepping into the new globalized era, a huge revolution has been undergone by the nuclear power industries. From a source of energy, today nuclear power has demonstrated a wide variety of applications, almost in every part of the world. Lately, the development has received criticisms due to huge generation of radioactive residues, which presents a threat to the environment. Thus, the aim of this paper is to highlight the fundamental characteristics and environmental implications of the nuclear waste compound. Apart from that, historical incidents surrounding the nuclear power-generating plants were clarified, and strategies and policies that will lead to a more sustainable development of the industries were discussed. Moreover, an up-to-date literature on activated carbon adsorption process as a treatment measure for uranium and radon was outlined.

Keywords: Activated carbon; Adsorption; Nuclear; Radon; Uranium

1. Introduction

To date, the emission of the nuclear effluents, specifically released from the nuclear fission reactors, naturally occurring radioactive materials, and medical wastes remains a highly contested agenda from a global view point [1,2]. With the price of the crude petroleum oil escalating to an unprecedented height due to dwindling oil reserves in the oil-exporting countries, towards the end of 2008, the installation of the world nuclear power plants is forecasted at 439 [3]. Meanwhile, the global uranium production was recorded at 40,000 tons, denoted approximately 15% of the world electrical energy generation [4]. In the common practice, nuclear waste is disposed at the open dumps (inundated swampland, abandoned sand mines, and quarries), without taking care of the surrounding environment, and prohibit the infiltration of contaminants into the underlying water channels. Lately, environmental rules and regulations concerning the pollution from industrial waste streams are more stringent and restrictive, inevitably affecting the design, planning, and operation of the radioactive-emitting industries [5]. This has inspired a developing exploration to establish a leading selective, reliable, and durable alternative for judicious treatment of the heavily polluted nuclear residues. With the aforementioned, this bibliographic review attempts to postulate an initial platform in describing the distinct physiochemical properties, development and environmental implications of the nuclear effluents. The present work is aimed at providing an up-to-date literature of activated carbon adsorption processes for the treatment of nuclear effluents. The environmental rules and the

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future prospects were laconically elucidated, to familiarize the knowledge deficiencies in this area.

2. The origin and physiochemical properties of the nuclear compounds

In general, nuclear compound is defined as a diverse group of decaying chemical substances (unstable configurations), emitting different types, levels, and periods of ionizing radiation (alpha, beta, neutron, or gamma radiation) [6]. In the perspective, uranium is an anisotropic, strongly electropositive, dense (19 g/cm^3), and paramagnetic metallic element [7], with the oxidation states of +3 (red), +4 (green), +5 (unstable), and +6 (yellow) [3]. Uranium is dispersed at the concentration level of 2–4 ppm in crust, rocks, and soils, and naturally occurs as uranyl hydroxyl carbonate complexes (in water) [8,9]. Uranium ions are water-soluble, but the degree of solubility is highly dependent on the presence of inorganic ligands (mainly $[OH]^-$, $[HCO_3]^-$, $[CO_3]^{2-}$, $[H_2PO_4]^-$ $[HPO_4]^{2-}$, $[PO_4]^{3-}$ and $[SO_4]^{2-}$) [10].

Whereas, radon is a tasteless, odorless, radioactive, and the heaviest member of noble gas, emitted from the decaying chains of radium, uranium, and thorium, or accumulates in the confined areas (attics, caves, and basements) and tap water (form coatings on the inside of pipe work) [11,12]. It is sparingly soluble in water, but appreciably more soluble in organic solvents [13]. Radon was emitted as a by-product from the uraniferous ores processing plants, with a brilliant phosphorescence below its freezing point (202 K; −71°C; −96°F), and turns orange-red as it liquefies below 93 K (−180.1° C; −292.3°F) [14].

Historically, uranium was initially discovered by Martin Heinrich Klaprothin Berlin as early as 1789 [15]. In 1841, the first sample of uranium metal was extracted by Eugène-Melchior Péligot, a professor of analytical chemistry at the Central School of Arts and Manufactures, Paris [16]. The proof of the radioactivity from uranium was unfolded by a French physicist, Antoine Henri Becquerel, in the middle of 1896, which signified the revolution of nuclear physics [17]. The modern chemical age of nuclear compounds began with the embarkment of uranium boom (Little Boy) over the Japanese city and the sparkling of atmospheric weapon tests until the turn of the 1950s [18].

Today, nuclear energy has been widened worldwide and entrenched into a multidisciplinary of development.

3. Environmental concern of nuclear effluents

Within the last few years, the percolation of nuclear waste into the waterways and atmosphere constitutes a persistent and detrimental impact towards the survival of aquatic compartments and environmental matrix. Acute exposure to nuclear substances may induce dramatic chemical damages, poisoning of forestry, inhibition of the terrestrial ecosystems and irrigation channels, resulting in food chains and ecological imbalance [19,20]. Increasingly, excessive inhalation of nuclear wastes is vulnerable to a broad variety of central nervous, specific lung defenses' failure, and reproductive sensory disorders presage as skeletal tumors, birth defects, stomach cancer (between 0.25 and 1.0% per 100,000 pCi/L in a water supply), and neurodegenerative diseases (amyotrophic lateral sclerosis) [21–24].

According to the United Sates Environmental Protection Agency, epidemiological study has indicated a significant linkage between the 80,000 additional cases of cancer (25% fatality rate) in North America with the nuclear weapons testing at Hanford [25,26]. Meanwhile, several chronic incidences of birth defects and physiological syndromes were correlated with the mishandling of nuclear accidents at Chelyabinsk, Russia, in 1957 [27], Windscale, UK, in 1958 [28], Chernobyl, Ukraine, in 1986 [29], Goiania, Brazil, in 1987 and the First Gulf War (Balkans and Iraq) in 1991 [30]. In Europe, excessive exposure to nuclear substances has been susceptible (Ahlbom and cohort studies) to adverse childhood leukemia and lymphoma at Sellafield nuclear power plant in the UK, and increased mortality rate among the uranium miners in Germany and Czechoslovakia [31]. In accordance with the threshold limits enacted under the World Health Organization, the peak tolerable daily intake of uranium level is 0.6 μg/kg of body weight per day [32], where the allowable occupational release concentration for radon is at an average value of 20 mSv per year [33].

4. Decontamination of nuclear waste via activated carbon adsorption processes

Within recent decades, activated carbon adsorption process, a surface phenomenon by which a multi-components fluid (gas or liquid) mixture is attracted to the surface of solid adsorbent and forms attachments via physical or chemical bonds, is recognized as the most promising treatment technology [34]. In particular, Dai and Wu [35] have initialed a pioneering study in 1975 intended to recover uranium metal ions onto a mixture of inorganic adsorbents (activated carbon, aluminum hydroxide, and ferric hydroxide mixture in the weight ratio 1:3:4). Similar research has been advanced by Abbas and Streat in 1994 [36] and 1998 [37] using a tri-n-butyl phosphate impregnated activated carbon. Nitric acid treated carbon sample illustrated an improving dynamic adsorption performance, with a longer breakthrough time, higher adsorption capacity, and larger Brunauer-Emmett-Teller (BET) surface area.

A year later, Park et al. [38] have evaluated the feasibility of coconut waste-based activated carbon and chitosan for removing four hydrolyzed uranyl species [UO_2^{2+}, $UO_2(OH)^+$, $(UO_2)_2(OH)_2^{2+}$ dimers and $(UO_2)_3(OH)_5^+$ trimer] with an initial concentration of 50 and 200 mg/L, respectively. Result showed a typical S-shaped breakthrough curve (maximum adsorption capacity of 175 mg/g at pH 3) with unusual trailing and plateau at higher pH regions, mainly ascribed to the presence of competitive (hydronium ions) and ion-exchange effect.

In 2003, Mahramanlioglu [39] has conducted a preliminary study to explore the potential use of waste tires (pre-impregnated with hydrochloric acid subsequent by carbon dioxide activation at 900°C) derived activated carbons for removing uranium ions from the aqueous solutions. Equilibrium data were correlated well with the empirical Freundlich, Lagergren and intraparticle mass transfer equation. Likewise, Coleman et al. [40] and Starvin and Rao [41] have performed a focus study to emphasize the versatility of hydrophobic aerogels and diarylazobisphenol modified activated carbons for removing uranium ions from the polluted wastewater. Modified activated carbons indicated a quantitative enhancement of the retention capacity (65% improvement), well described by the Langmuir ($R^2 = 0.99997$), Freundlich ($R^2 = 0.99924$) and first-order kinetic ($n = 2.87$ and $K_f = 1{,}169$) models.

Meanwhile, Kütahyalı and Eral [42] and Mellah et al. [43] have examined the influences of contact time, pH, concentration, and temperature on the batch adsorption systems. The optimum operating conditions were reported at an initial concentration of 50 ppm, 4 h of shaking time, and pH 5; and at an initial concentration of 100 mg/L, 4 h of shaking time, and pH 3, respectively. Accordingly, Kütahyalı and Eral [44] have attempted to prepare an olive-stone-based activated carbon via $ZnCl_2$ chemical activation for the sorption of uranium and thorium from aqueous solutions. The sorption of uranium and thorium was studied as a function of shaking time, pH, initial metal ion concentration, temperature, and adsorbent concentration in a batch system. The total sorption capacities were found to be 0.171 and 0.087 mmol/g for uranium and thorium, respectively. The positive values of enthalpy change, $\Delta H°$, indicated the endothermic nature of the process for both metals, and decrease in Gibbs energy change, $\Delta G°$, with the rises of temperature validated the sorption is more favorable at high temperatures.

In China, Zhao et al. [45] have developed a new solid extractant, benzoylthiourea-anchored activated carbon for selective removal of uranium (VI) ions. The maximum sorption capacity was obtained at 82 mg/g, while the rate constant for the uranium sorption by the as-synthesized extractant was 0.441 min^{-1} from the first-order rate equation. Thermodynamic parameters ($\Delta H° = -46.2$ kJ/mol; $\Delta S° = -98.0$ J/mol K; $\Delta G° = -17.5$ kJ/mol) showed that the adsorption is exothermic and of spontaneous nature, respectively. Additional studies indicated that the benzoylthiourea-anchored activated carbon selectively sorbed uranyl ions in the presence of competing ions, Na^+, Co^{2+}, Sr^{2+}, Cs^+, and La^{3+}.

Until the turn of 2011, Morsy and Hussein [46] have carried out a batch sorption experiment using a commercial grade granular activated carbon (GAC) for removing uranium ion from crude phosphoric acid. It was found that nitric acid treatment oxidized the surface of activated carbon and significantly increased the adsorption capacity for uranium in acidic solutions. The finding is in agreement with the previous studies [36,37]. Equilibrium data were well fitted to the Langmuir isotherm model, showing a monolayer capacity of 45.24 mg/g. On the contrary, Jung et al. [47] have performed a pitch-based activated carbon fiber (ACF) assisted electrosorption study to treat lagoon sludge containing 100 mg/L uranium and high concentration of chemical salts composed of 3.8% $NaNO_3$, 19.8% NH_4NO_3, and 1.9% $Ca(NO_3)_2$. At a potential of −0.9 V (vs. Ag/AgCl) and pH 4, uranium concentration in the effluent was reduced to less than 1 mg in 30 min and maintained for 50 h. It is equivalent to the cumulative amount of uranium of about 600 mg/g. Cyclic electrodesorption by simple potential reversing allowed more than 99% of adsorbed uranium (VI) ion to be recovered at +1.2 V and pH 3. No change in adsorption performance and physical structure of ACF electrode was observed after a continuous five cycling, indicating it is a reversible process. Thus, it is likely that electrosorption process for the removal of uranium ions would be economically attractive due to the high electrosorption efficiency and the possibility for regeneration.

In the same vein, the ordinary concept of radon gas adsorption process has been first proposed by Rutherford [48], an England chemist who surprisingly discovered the feasibility of activated carbons for reducing the emanation of radon in 1906; and during the 1960s and 1970s, the invention of activated carbon analytical techniques for specific detection of radon concentration in slurries and groundwater (hydro-geological investigations) has been witnessed [49,50]. Towards the end of the 1980s, George [51] and Scarpitta and Harley [52] have alleged an early attempt for adequate determination of the indoor radon concentration within the exposure intervals from one to seven days; while a three-year laboratory column study on the removal of radon from water supplies has been undertaken by Lowry and Brandow [53] in 1985, which recorded a competent removal of 85%.

In Columbia, Hassan et al. [54] have conducted a comparative experiment assessing the consistent measurement of radon concentration onto three commercially available adsorbents, BPL activated carbon, silica gel (grade 40), and molecular sieve type 13X. Isotherm equilibrium data were correlated well with type III adsorption isotherm as defined by the BET classification, and modified semi-empirical Freundlich equation written as:

$$q = k'(P/P_0)^n \tag{1}$$

or

$$\ln q = \ln k' + n \ln(P/P_0) \tag{2}$$

where q, P, $P_{0,}$ T, and n are the adsorption capacity (pCi/g), adsorbate pressure (mmHg), reference pressure (mmHg), temperature (K), and adsorption intensity (dimensionless),respectively; and k', the temperature-dependent adsorption rate constant (pCi/g) is given by the expression:

$$k' = 2.10 \times 10^{-11} - 6.58 \times 10^{-14}T \tag{3}$$

In this respect, the adsorption capacity (pCi/g) and concentration (pCi/L) of radon onto BPL activated carbon were derived as:

$$q = (2.10 \times 10^{-11} - 6.58 \times 10^{-14}T)/(P/10^{-14})^{1.75} \tag{4}$$

$$C = \frac{9.1 \times 10^{-21} q^{0.571} T^{-1}}{(2.10 \times 10^{-11} - 6.58 \times 10^{-14}T)^{0.571}} \tag{5}$$

Whereas, López and Canoba [55] have adopted the applicability of Henry's Law and parabolic second-order differential equation for describing the sorption rate of radon gas given by:

$$Q(t) = k(t)C \tag{6}$$

where $Q(t)$, $k(t)$, and C are the functions of adsorbed activity (Bq/kg), adsorption coefficient (m^3/kg), and concentration (Bq/m^3), respectively.

In Hungary, Espinosa and Silva [56] have revealed the reliability for measuring indoor radon concentrations using a commercially available air-purifying respirator filter. The method is based on the diffusion of radon gas onto the carbon filter, and the measurement of the radioactive daughters resulting from radon decay. The photopeaks of the ^{214}Bi daughter gamma rays (0.609 MeV) were analyzed with a Hyper-Pure Germanium detector and a multichannel system. This procedure resulted in highly reproducible and reliable measurements of indoor radon levels. A similar result was reported by Chege et al. [57] who investigated the effects of meteorological parameters (outdoor temperature, rainfall, and wind speed) on the indoor radon concentration in four types of traditional hut in Kenya, by performing short-term (about 48 h) measurements with activated charcoal canisters. Yasuoka and his co-workers [58] who evaluated the effects of deodorizing activated carbon (carbon-filter) for radon mitigation have indicated the similar findings. The results showed that activated carbon respiratory filters may potentially be used as personal, mobile radon monitors, environmental monitors, or as aids in the investigation of radiological accidents.

Meanwhile, Alabdula'aly and Maghrawy [59] have carried out laboratory and pilot plant studies using three different types of activated carbons (F-300, F-400, and HD-4000) to remove radon from the water supply. From the experimental kinetic study, the data indicated that at least 6 h are needed to attain the equilibrium between radon activity adsorbed onto carbon and its concentration in the aqueous phase. The γ-exposure rate (mR/h) was monitored at different depths throughout the whole pilot plant run (60 days). It can be found that the exposure rate of F-300 and F-400 reached maximum values (0.2 mR/h) at 36 cm depth after 14 days of operation. However, for HD-4000, the corresponding value was 0.23 mR/h at 15 cm depth after the same time period. At steady-state performance, the maximum dose for both F-300 and F-400 was 0.12 mR/h at 56 cm depth, while the maximum value for HD-4000 was 0.14 mR/h corresponding to 15 cm depth. The result is useful for designing a GAC adsorption system for the removal of medium level radon concentration from water supplies. Relatively, Cooper et al. [60] has introduced a temperature calibration formula based on the gas adsorption theory to evaluate the radon concentration in air from the average temperature, collection time, and liquid scintillation count rate. Experimental results showed that the adsorption efficiency of the detectors can vary by nearly a factor of two for temperature values in the range 8.5–31°C. The derivation is expected to be useful for establishing standardized protocols for optimized radon assessment in dwellings and workplaces (Table 1) [38–47,53,54,59,61,62].

5. Management rules and future prospects

For the past two decades, the transition to the nuclear fusion energy has been one of the rapidly advancing dilemmas to the natural environment. Numerous technologies have been addressed and confronted to utilize activated carbon (or its integrated technologies) for the adsorptive treatment of nuclear waste [5,63–65]. Although there have been some successful industrial-scale applications, generally the industry is still facing various constrictions and sustainable resources management. Amidst these shortcomings, there has been a developing innovation of microwave technology for the preparation of activated carbons. Compared with conventional heating techniques, microwave irradiation has additional advantages as higher heating rates, selective heating, greater control of the heating process, no direct contact between the heating source and heated materials, and reduced equipment size and waste [66].

Accordingly, the urgency to administer strategic and transparent policies, which govern the collection, design, and supervision of the nuclear waste management, ought to be prudently well planned. Increasingly, the sound professional knowledge of creating environmental awareness for adequate operating standards, responsibilities sharing and public participations needs to be properly assigned and counteracted. In Europe, an initiative on persistent radioactive pollutants within the framework of the International Atomic Energy Agency (IAEA), European Union (EU), and Nuclear Energy Agency of the Organization for Economic Co-operation and Development (NEA/OECD) has been established. Meanwhile, several waste management regulations and Specified Radioactive Waste Final Disposal Act under the Nuclear Waste Management Organization of Japan (NUMO) and Nuclear and Industrial Safety Agency (NISA) have been declared for virtual handling, control, and vigilant destruction of the nuclear progenies throughout their life cycles, with special emphasis given to its transboundary shipment [33,67,68].

In United States, the enactment of the Public Law 95-604, Nuclear Waste Policy Act 1982, Safe Drinking Water Act Amendments 1986; and subsequent formation of the Pan American Health Organization (PAHO), Atomic Energy Commission (AEC), and Global Nuclear Energy Partnership (GNEP) was amended in 2006 to include the submission of Environmental Impact Assessment (EIA) reports for approval [19,69–71].

Table 1
Lists of researches for nuclear waste treatment via activated carbon adsorption process during the last 25 years

Nuclear effluents	Precursors/type of activated carbons	Adsorption capacity	Removal (%)	Operational parameters	Reference
Uranium	Coconut	175.00 mg/g	≈100.0	$C_0 = 50\text{–}500$ mg/L; pH = 5.0; $T = 30$°C	[38]
Uranium	Used tires	216.03 mg/g	≈100.0	$C_0 = 238\text{–}714$ mg/L; pH = 3.5; $T = 20$°C	[39]
Uranium	Commercial	2.00 mg/g	–	$C_0 = 0.1$ mg/L; pH = 7.0; T = room temperature	[40]
Uranium	Diarylazobisphenol modified	18.35 mg/g	99.8	$C_0 = 60$ mg/L; pH = 4.5; T = room temperature	[41]
Uranium	Charcoal	50.54 mg/g	96.0	$C_0 = 50$ mg/L; pH = 5.0; $T = 30$°C	[42]
Uranium	Commercial	28.30 mg/g	≈100.0	$C_0 = 100\text{–}300$ mg/L; pH = 3.0; $T = 20$°C	[43]
Uranium	Olive stones	40.70 mg/g	–	$C_0 = 25\text{–}300$ mg/L; pH = 6.0; $T = 30$°C	[44]
Uranium	Benzoylthiourea anchored	82.00 mg/g	–	$C_0 = 20\text{–}200$ mg/L; pH = 5.0; $T = 20$°C	[45]
Uranium	Commercial	45.25 mg/g	–	$C_0 = 200$ mg/L; pH = 5.0; $T = 25$°C	[46]
Uranium	Activated carbon fibers	600.00 mg/g	≈99.0	$C_0 = 100$ mg/L; pH = 4.0; V = –0.9 V	[47]
Uranium	Commercial	0.03 mg/g	–	$C_0 = 0.1$ mg/L; pH = 5.0	[61]
Radon	Commercial	–	85.0	$C_0 = 55.50\text{–}27{,}750$ Bq/L; $T = 10$°C	[53]
Radon	Commercial	21.56 Bq/g	–	$C_0 = 547.60$ Bq/L; $T = 15\text{–}35$°C	[54]
Radon	Commercial	–	80.6	$C_0 = 111$ Bq/L; $T = 23$°C	[59]
Radon	Commercial	0.34 Bq/g	≈100.0	$C_0 = 910\text{–}6{,}000$ Bq/L	[62]

Ultimately, full cooperation and joint venture between different parties from upstream till the bottom line is a directive motivation for the race to the end line.

6. Conclusion

Predictions for the next 20 years indicate an extensive use of the nuclear power energy, and subsequent in nuclear effluents' generation. Limited and improper management has risen over the use of activated carbon adsorption as a measure for the field remediation. The feasibility of the process has been well proven by a number of laboratory and pilot-scale studies. With the advancement of microwave irradiation which involves shorter processing time, energy savings, selective heating, and reduced equipment size and waste, activated carbon process will certainly be a major contributor for treatment of nuclear effluents. Although it is still in the infancy, a great progress in this area can be expected in the future.

Acknowledgment

The authors acknowledge the financial support provided by Universiti Sains Malaysia under the Research University (RU) scheme (Project No. 1001/PJKIMIA/814072) and RU-PRGS grant scheme (Project 465 No. 8043030).

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