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TECHNICAL NOTE

THE REMOVAL OF URANIUM FROM AQUEOUS SOLUTION

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ABSTRACT

This paper reviews the literature on uranium contamination and the removal of uranium from wastewater stemming from mining activities and nuclear power generation. After reviewing the applications of uranium in power generation, military, industry and scientific, this review discusses uranium and rare earth elements in wastewaters and the toxicity of uranium on aquatic life and humans. Further, various methods of removal of heavy metal contaminants including uranium are reviewed with special focus on the adsorption process and carbon nanotubes as a superior adsorbent.

ABSTRAK

Makalah ini membentangkan sorotan karya tentang pencemaran uranium dan pengasingan uranium daripada kumbahan hasil daripada aktiviti perlombongan dan penjanaan kuasa nuklear. Bermula dengan kegunaan uranium dalam penjanaan kuasa, ketenteraan, industri dan saintifik, sorotan ini membincangkan uranium dan elemen nadir dalam air kumbahan dan keracunan uranium kepada hidupan akuatik dan manusia. Seterusnya, beberapa kaedah pengasingan kontaminan metal berat termasuk uranium dibincangkan dengan fokus kepada proses penyerapan dan nanotiub karbon sebagai adsorbent yang terunggul.

Keywords: Uranium, heavy metal, rare earth, adsorption, carbon nanotubes.

INTRODUCTION

Most heavy metals are toxic and pose serious threat to humans, fauna and flora (Crini, 2006). These heavy metals cause alterations to the biochemical cycles of living things when discharged into the wastewater (Srivastava and Majumder, 2008). Large concentrations of heavy metals may accumulate in the human body once they enter the food chain. As a consequence, serious health disorders like kidney or liver damage (Wang et al., 2009) occur if the metals are ingested beyond the permitted concentration (Babel and Kurniawan, 2004).

Rapid development and growth of industrial, agricultural processes and mining activities have increased the concentration of toxic contamination in water and wastewater around the world, thus increasing the amounts of heavy metals directly or indirectly disposed into the environment (Shawky *et al.*, 2005). Radionuclides such as uranium possess high toxicity and radioactivity and exhibit a serious threat, even at small concentrations. Treating the wastewaters by removal of contaminants before discharge is important to avoid pollution of natural waters (Li *et al.*, 2011; Zhou, Liang and Wang, 2008). Thus, the removal of toxic and polluting heavy metal ions from industrial effluents, water supplies and mine waters has received much attention in recent years. Uranium, a toxic and weak radioactive heavy metal contaminates surface water and groundwater, where the concentrations of uranium may increase above 2µg/L. Recent realization that uranium's chemical toxicity is more dominant than its radiation hazards makes decontamination a relevant topic from the environmental point of view. Of particular interest is the projected global thrust for uranium fuel based atomic power plants (Sheppard *et al.*, 2006). Thus, this paper reviews the literature on uranium and the available removal technologies.

URANIUM DEMAND AND ITS APPLICATIONS

Power Generation

The major use of uranium in a non-military or civilian capacity is in nuclear power plants. It is used to fuel the nuclear fission reactors.With the threat of climate change and volatility of oil prices, utilities worldwide are reconsidering nuclear as an alternative and uranium is the main elements in nuclear power generation. Nuclear Malaysia (2010) reported that the Asian countries are aggressively venturing into nuclear power with more than half of the current reactors under construction are to be built in countries such as India, China, Japan and Korea. In July 2009, Malaysia decided to include nuclear energy as part of an energy option for electricity generation particularly in Peninsular Malaysia post-2020. As part of the Economic Transformation Programme (ETP) framework, deployment of nuclear for power generation was one of the key Entry Point Project announced (PEMANDU, 2010). According to the Ministry of Energy, Green Technology and Water (KETTHA), Malaysia plans to build two units of nuclear reactors that will generate 1000MW each with the first plant ready for operation by 2021 (Ting and Tengku Abdullah, 2010). Excessive amounts of uranium have reached environment through the activities of nuclear industry. In most developed and developing countries, stricter environmental regulations, with regard to contaminants discharged from industrial on-site or in-plant facilities to their own effluents and minimize the contaminant concentrations to acceptable limits prior to their discharge (Banat et al., 1996).

Military

The main application of uranium in the military sector is in projectiles (Arfsten, Still and Ritchie, 2001). Depleted uranium is used to create high-density penetrators such as those projectiles used to piece heavily armored targets (Giannardi,2003). Removable armor for vehicles such as tanks is made from hardened uranium plating. Depleted uranium is used as shielding in containers that store radioactive materials despite the fact that it itself is radioactive.Uranium is also used as the fissile explosive to create nuclear weaponry. Uranium is used to power nuclear powered submarines and ships (Bem and Bou-Rabee, 2004).

Industry

Before people knew that uranium was radioactive it was used as yellow coloring for pottery and glassware. It was also used to color kitchen and bathroom tiles.Uranium was used in photographic chemicals, lamp filaments, to improve the whiteness of dentures and in the leather and wood industry as stains and dyes. Uranium is also mined to isolate radium for use in glow in the dark paints for clock faces and aircraft dials. The radioisotopes from uranium are used in smoke detectors. Uranium metal is used as ballasts in yachts and airplanes (VanHorn and Huang, 2006).

Scientific

Uranium derivatives are used in biological science as stains to increase the contracts between virus and other cells (Kalinich and McClain, 2001). Its long half-live has also led to it being used in dating the age of the earth.

Uranium metal is used as a target for high-powered x-ray machines (Gall, Mikecz, and Papp, 1988).On the other hand, rare earth elements have become essential ingredients in the production of many industrial materials including permanent magnet, catalytic cracking materials, luminescence materials, hydrogen storage materials, magnetic refrigeration materials, optical fiber, magneto-optical storage materials, giant magneto resistance materials, lasers, superconductor materials and dielectric materials.

URANIUM IN WASTEWATER

Evidently, many reports cite Malaysia as having reasonably substantial amounts of rare earth elements. In the case of the Lynas operation in Gebeng, Pahang, the Lynas operation's residues are expected to contain thorium and uranium and their decay products at concentration of about 1600ppm (Th) and 30 ppm (U) depending on minerals used in the process (IAEA, 2011). Any move to venture into rare earths would not be difficult since mining is not entirely alien to the country. The implication of this development is the potential of wastewater being affected by heavy metal such as uranium. With the increasing demand for uranium and rare earth elements, mining activities will be on the rise and consequently the waste of mine tailings will become an issue. Large quantities of uranium mill tailings have accumulated probably more than 938 million m^3 of uranium tailings produced at more than 4,000 mines (Abdelouas, 2006).One example is the mining activities and ISL operations in German severely disturbed ground water system. Approximately 2,650 million liters of mine water containing sulphuric acid and uranium (levels up one to three orders of magnitude higher than German water quality guidelines). They were circulated or trapped in the pore space of the rocks. The trapped waters have arsenic, cadmium and nickel as well. While the mine was in operation, there was little ground water contamination because the water flowing into the mine was pumped away from the acidified ore body. However, with the cessation of mining, pumping of working areas ceased, and the ground water may become contaminated because of the mixing of inflowing ground water with the remaining acid. It is dangerous as one of the water system storage will lead to the consumer. During mining operation, large volumes of contaminated water are pumped out of the mine and released to rivers and lakes, spreading into the environment (Filov et al., 1993).

Another source of environmental pollution by uranium is phosphoric fertilizers which have been reported to increase the uranium content of top soil to between 30 and 300 mg/kg dry weight (Harmsten et al., 1980; Filov et al., 1993). The main types of uranium containing liquid wastes are mine waters and effluents from hydrometallurgical (uranium ore-processing) work. Such wastes were produced in quantities of 2-2.5 m^3 per ton of processed ore and contained uranium in concentrations of the order of 1-3 mg/L. During mining operation, large volumes of contaminated water are pumped out of the mine and released to rivers and lakes, spreading into the environment (Filov et al., 1993). The poor technology of waste management leads to the contribution uranium exposure to the ground water system.

URANIUM TOXICITY

Naturally occurring radionuclides (NOR) such as uranium as well as its progenies that co-exist with rare earth minerals and other valuable minerals is technologically enhanced during the separation process. These materials posed additional health problems related to ionizing radiation or radiological risk (Ismail, 2001; Ismail, 2011). Uranium series have progenies that are gamma, alpha and beta emitters and they are considered as internal as well as external radiation hazards. Health risks related to radiation exposures from NOR are present in rare earth minerals as well associated valuable mineral are dependent on the doses received. Levels of doses received are dependent on activity of NOR in these raw minerals, rare earth concentrates and residues.

Aquatic organisms

The growth of saprophylic aquatic microflora was depressed at a uranium concentration of 0.5 mg/L and ceased completely within 8-10 days at 100 mg/L. Algae showed slowed growth at 10 mg U/L and failed to grow altogether at 100 mg U/L. For daphnids (sea fleas), uranium was toxic at concentrations of 5 and 10 mg/L and is lethal within 24 h at 100 mg/L. Guppies exposed to uranium at 50 mg/L all died within 84 days; with exposures to 5, 10, and 26 mg U/L, stillbirths were frequently observed. The biochemical oxygen demand in bodies of water began to decrease within 24 h when uranium was added at 5 mg/L and after 24 h when it was added at 1 mg/L. The lowest uranium concentration than altered the sanitary condition of water bodies was 1mg/L (Filov et al., 1993).

Humans

Uranium (VI) is a human carcinogen and its concentration level in water should not exceed 0.002 mg/L (2 ppb) according to the WHO, (Wang et al. 2009).Uranium is a general cellular poison and can potentially affect any organs and tissues. The two main hazards caused by uranium are damage due to its chemical toxicity and the injury caused by the ionizing radiation resulting from the disintegration of uranium isotopes. Normally, it depends on the solubility of the compound concerned, its route of entry into the body and isotopic composition. The chemical toxicity of uranium compounds is directly dependent on their solubility in biological media. Therefore, the rates of uranium compounds go into solution is proportional to its solubility. Disposed uranium is hazardous once it reaches the top of the food chain and penetrates into humans. The critical organ for the chemical toxicity of uranium and its soluble compounds is the kidney. Majority of uranium compounds are excreted through and accumulate in this organ, causing structural damage to its tubules and glomeruli and thus impairing its function. Besides, it brings effect like changes in pathologic and impair the function of many other organs and systems including a gastrointestinal tract, liver, lungs, thyroid, adrenals, cardiovascular system and central peripheral nervous systems.

In addition, uranium also causes disorders of water, lipid, and carbohydrate metabolism (Wang et al., 2009). Particularly toxic are compounds soluble in water and body fluids (Tarasenko, 1983; Filov et al., 1993). The staffs working at nuclear facilities or people living nearby face risks of radiation impact during normal operation or accidents (Anke et al., 2009; Zoriy et al., 2010).

In a study involving 130 persons chronically exposed to soluble uranium compounds, seven of them were found to have nephrosis as their urine contained uranium at levels up to 3.46 mg/L. Examination of 237 uranium mine workers revealed anemia in 31% of them, marked leucopenia in 23%, and lymphocytosis in 14%. The similar group uranium workers whom employed for about 10 years reduced body weight and pathological changes in the lungs, kidneys, and blood due to radiation exposure. On the other case, the employees of a factory producing chemical reagents from uranium salts such as uranyl nitrate, uranyl acetate, and uranium (VI) oxide were diagnosed and having "vegetative dystonia and unstable parameters of peripheral blood with tendency toward leuko-, lympho-, and thrombopenia" (Andreyeva, 1960, 1971; Filov et al., 1993). Uranium can enter the human body by oral route, inhalation, skin and mucous membranes. The daily dietary intake of uranium by inhabitants of three urban areas in the USA was estimated to be $(1.3\text{-}1.4)\times10^{-6}$ g. For the United Kingdom, an average daily intake of about 1×10^{-6} g was recorded, while intakes of up to 4.55×10^{-6} g/day were reported for areas near uranium mines in Japan. Normally, some 70% of dietary uranium is contained in meat, fresh fish, and bakery products. Drinking water containing uranium in concentrations of $(0.025\text{-}0.04)\times10^{-6}$ g/L contributes only 2-3% of the daily intake. In contrast, drinking water was the major source of uranium intake in areas where it contained high concentrations of this element (2×10^{-4} g/L), and in such areas the contribution of food was small (1-5%) (Filov et al., 1993).

WASTEWATER TREATMENT TECHNOLOGIES

The removal of uranium from aqueous solution is a crucial step due to its extreme toxicity and environmental concern (James et al., 2008). Therefore, wastewaters containing uranium are required to be treated in order to prevent radioactive contamination of the environment (Wang et al., 2009). The removal of these persistent inorganic pollutants (PIP's) has become a vital and fertile area for the present day research (James et al. 2008).

Technologies for treating industrial wastewaters can be divided into three categories: chemical methods, physical methods, and biological methods. Chemical methods include chemical precipitation, chemical oxidation or reduction, formation of an insoluble gas followed by stripping, and other chemical reactions that involve exchanging or sharing electrons between atoms. Physical treatment methods include sedimentation, flotation, filtering, stripping, ion exchange, adsorption, and other processes that accomplish removal of dissolved and undissolved substances without necessarily changing their chemical structures. Biological methods are those that involve living organisms using organic, or in some instances, inorganic, substances for food, completely changing their chemical and physical characteristics. As a general rule, biological treatment is more economical than any other type of treatment, when reasonably complete treatment is required, and whenever it can be made to work successfully (Woodard, 2001).

Removal of toxic inorganic pollutants was accomplished successfully over years by various techniques like liquid–liquid extraction (LLE), ion exchange, nano and ultra filtration, bio-sorption, co-precipitation, electro deposition and solid phase extraction (SPE), imprinted polymers-SPE (James et al. 2008). A number of different methods such as membrane process, coagulation, dialysis, adsorption, foam flotation, osmosis, photocatalytic degradation (Li et al. 2011; Wang and Zhou, 2008) and biological methods have been used for the removal of toxic pollutants from water and wastewater (Pontius, 1990).

Most of the available methods proposed cannot be used on an industrial scale for technological and especially economic reasons. The proper disposal of the resulting sludge that contains high levels of the metal and other contaminants is another major limitation for these processes. Conventional methods for removing metal ions from aqueous solution have been studied in detail, such as chemical precipitation, ion exchange, electrochemical treatment, membrane technologies, adsorption on activated carbon etc. However, chemical precipitation and electrochemical treatment are ineffective, especially when metal ion concentration in aqueous solution is as low as 1 to 100 mg/L, they also produce large amount of sludge to be treated with great difficulties. Ion exchange, membrane technologies and activated carbon adsorption process are extremely expensive, especially when treating a large amount of water and wastewater containing heavy metal in low concentration, so they cannot be used at large scale (Wang and Chen, 2006). Much has been discussed about their downside aspects in recent years (Atkinson et al., 1998; Crini, 2006), which can be summarized as expensive, not environment friendly and usually dependent on the concentration of the waste.

Several methods have been utilized to remove uranium from wastewater and process effluents. Among them are chemical coagulation–flocculation (Gafvert et al., 2002), activated carbon (Coleman et al., 2003), ion exchange (Barbette et al., 2004; Barton et al., 2004; Bryant et al., 2003; Gu et al., 2004), ultrafiltration assisted by complexation (Kryvoruchko et al., 2004) and nanofiltration (Favre-Re´guillon et al., 2008). However, many of the available processes proposed cannot be used on an industrial scale for technological and especially economic reasons. Another major limitation for these methods is the proper disposal of the resulting sludge that contains high levels of the metal and other contaminants. Other viable technologies include chemical precipitation, membrane filtration, ion exchange, liquid extraction, adsorption, etc. However, only adsorption method is the most versatile and widely used because the other methods are high cost and low feasibility for small-scale industries (Hu, 2009). It also considered to be very important due to its cost effective treatment, easy operation, narrow space for building the plant, no chemical reagents needed and no sludge produced (Anirudhan et al. 2010).

ADSORPTION PROCESS

Adsorption process is considered most appropriate as it can remove both organic and inorganic pollutants and the operation of the process is convenient (Bhatnagar and Sillanpaa, 2009). Over the last few decades adsorption has gained importance as an effective purification and separation technique used in wastewater treatment and low cost adsorbents are becoming the focus of many investigations on the removal of heavy metals from aqueous solutions (Tran et al., 2010). Adsorption techniques are widely used to remove certain classes of pollutants from waters, especially those that are not easily biodegradable (Crini, 2006). In accordance with the very abundant literature data, liquid-phase adsorption is one of the most popular methods for the removal of pollutants from wastewater since proper design of the adsorption process will produce a high-quality treated effluent. This process provides an attractive alternative for the treatment of contaminated waters, especially if the sorbent is inexpensive and does not require an additional pre-treatment step before its application. Adsorption is a well-known equilibrium separation process and an effective method for water decontamination applications (Dabrowski, 2001).

Adsorption has been found to be superior to other techniques for water re-use in terms of initial cost, flexibility and simplicity of design, ease of operation and insensitivity to toxic pollutants. Adsorption also does not result in the formation of harmful substances (Crini, 2006). Many other factors can also affect the adsorption capacity in the same sorption conditions such as surface chemistry (heteroatom content), surface charge and pore structure. A suitable carbon should possess not only a porous texture, but also high surface area. Recently, Guo et al. (2003) showed that the adsorption does not always increase with surface area. The adsorption capacity depends also on the accessibility of the pollutants to the inner surface of the adsorbent, which depends on their size.

Crini (2006) reported that the adsorption has advantages that make them excellent materials for environmental purposes, such as high capacity and rate of adsorption, high selectivity for different concentrations, and also rapid kinetics. The effectiveness of treatment depends not only on the properties of the adsorbent and adsorbate, but also on the following environmental conditions and variables used for the adsorption process: pH, ionic strength, temperature, existence of competing organic or inorganic ligands in solution, contact time and adsorbent concentration (Crini, 2006). Despite the fact that industrial effluents contain several pollutants simultaneously, little attention has been given to adsorption of pollutants from mixtures (Aksu, 2005). The development of the adsorption process requires further investigation in the direction of testing. Adsorption has been found to be superior to other techniques for water treatment due to low cost, flexibility and simplicity of design, ease of operation and insensitivity to toxic pollutants (Tan et al., 2008).

Adsorbent

For adsorption of uranium (VI) from wastewater, many researchers have used various solids, which are natural, organic and inorganic, for example, coir pith, zeolite, bentonite (Majdan et al., 2010) cork biomass, goethite, activated carbon (Kutahyali and Eral, 2004), and cement (Yusan and Akyil, 2008). Recently, numerous approaches have been studied for the development of cheaper and effective adsorbents. Many methods have been developed and used to remove metal ions from wastewater such as granulated activated carbon (Sekar, 2004), fly ash (Mckay, 1999), peat (Chu, 1999), resins (Arias, 2002), kaolinite (Sublet, 2003), manganese oxides (Biskup, 2004), zeolite (Li, 2004), and biomaterials (Ekmekyapar, 2007; Anirudhan et al., 2010). However, some of these processes are often associated with many problems which make their use ineffective. For example, high operational costs and the inability of these processes to remove the heavy metals to the acceptable levels are the most common shortcomings of some of these processes. These methods have also not been very efficient for the removal of low concentrations of heavy metals.

The adsorption processes are found to be the most promising techniques for the removal of heavy metals from water with functionalized carbon nanotubes as superior adsorbents. However, apart from the study undertaken by Dobrowolski and Stefaniak (Dobrowolski, 2000), most studies that have reported the uptake of metal ions by carbon nanotubes and other adsorbents have focused on the uptake of such ions in the parts per million concentration range (usually above 100mg/L) (Mohan, 2006) and not for trace concentrations, i.e. the parts per billion concentration range.

Carbon nanotubes (CNTs) known as “materials of the 21st century” due to their unique properties such as functional mechanical, thermal, electrical and optoelectronic properties which depend on atomic arrangement (how the sheets of graphite are ‘rolled’), the diameter and length of the tubes, and the morphology, or nanostructure (Avouris, 2002). Due to their surface functional groups and hydrophobic surfaces, CNTs show strong interactions with both heavy metal ions and organic compounds. CNTs are relatively new adsorbents and hold interesting positions in carbon-based adsorptive materials for many reasons. On one hand, they provide chemically inert surfaces for physical adsorption, and their high specific surface areas stand comparison with those of activated carbons (ACs). On the other hand, CNTs are essentially different from ACs in the aspect that their structure at the atomic scale is far more well-defined and uniform. Due to their highly porous and hollow structure, large specific surface area, light mass density and strong interaction between CNTs and pollutant molecules (Long, 2001), the applications of CNTs for the removal of hazardous pollutants from gas streams (Agnihotri, 2005) and from large volumes of aqueous solutions have being studied extensively.

Recent advancements in the synthesis, processing and understanding of nanostructured materials have generated considerable interest in the materials research community (Sandoval et al., 2005). Since, the discovery of carbon nanotubes (CNTs), researchers has been investigating methods for exploiting their extraordinary properties (Sandoval and Wicker, 2006).There is one promising methods to synthesis CNTs using castor oil under microwave radiation at a comparatively low temperature. The characterized result is shown in the Fig 1. CNTs exhibit a more compact stacking morphology due to cohesive force generated from the introduction of oxygen containing functional groups.This method have several benefits due to the presence of inexpensive and natural oil serving as raw material, causes no leakage and requires no vacuum conditions during synthesis. In addition it is free from resource depletion and is friendly to the environment. Natural resources present in the natural world and recycled raw materials will come to be employed from environmental considerations. In addition, oils that are consumed via combustion are required to be fixed as CNTs, from the viewpoint of reduction in amount of carbon dioxide emitted to the atmosphere (Ahmad Zaidei, Abdul Rashid and Normanbhay, 2011).

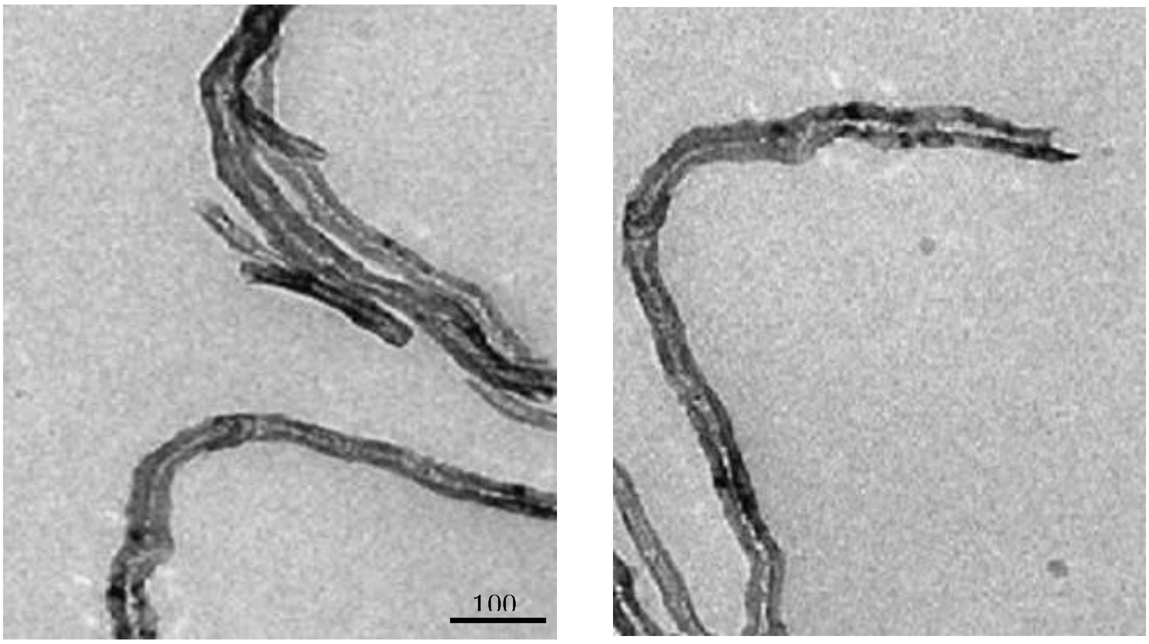

Figure 1: Philips EM 400 transmission electron microscope at an accelerating voltage of 40kV.

(*Source: Ahmad Zaidei, Abdul Rashid and Normanbhay (2011).*

DISCUSSIONS

CNTs include single-walled carbon nanotubes (SWCNTs) and multiwalled carbon nanotubes (MWCNTs) depending on the number of layers comprising them. The methods for preparing of MWCNTs are easy and can be produced in large scale. Multi-walled carbon nanotubes (MWNTs) consist of concentric cylinders around a common central hollow with a constant separation between the layers close to the graphite interlayer spacing (0.34 nm). Each individual cylinder can be characterised by a different helical angle and has a diameter ranging from 2 to 25 nm and a length of several microns (Thostenson et al., 2001 and can reach diameters of up to 100nm (Balasubramanian and Burghard, 2005). MWCNTs are efficient and suitable candidate for the preconcentration and solidification of pollutants from large volumes of wastewater (Wang et al., 2005; Fugetsu et al., 2004; Chen and Wang , 2006; Yang et al., 2006) .MWCNTs could be modified via acid (H2SO4/HNO3 = 3:1 in volume) treatment at 608C for 4 h. Through acid treatment, carboxylic groups (-COOH) could be successfully introduced onto the surface of MWCNTs (Jiang et al., 2011).

The oxidized MWCNTs exhibit excellent regeneration capacity, high sorption capacity with a wide range in pH, recycle and environmental friendliness for the disposal of U(VI)-bearing wastewater. In particular, it is worth mentioning here is that the toxicity of the MWCNTs occurred in air pollution-related cardiopulmonary diseases due to its fine particle matter (Lam et al., 2006).

The removal of U(VI) by oxidized MWCNTs was strongly dependent on pH and ionic strength. The presence of HA/FA enhanced U(VI) removal on oxidized MWCNTs at low pH while inhibited U(VI) sorption at high pH. The mechanism of U(VI) sorption on oxidized MWCNTs was assumed to be cation exchange/outer-sphere surface complexation in acidic pH and to form precipitation under circum neutral conditions. The oxidized MWCNTs exhibit higher sorption capacity and stronger chemical affinity than pristine MWCNTs.

Table 1: The study of uranium uptake by different adsorbents

Adsorbents	Conditions	Uranium uptake, qmax (µmol /g)	Reference
Hematite	pH = 4.0, I = 0.1 M $NaNO_3$	1.0	Bargar et al. (2000)
Montmorillonite	pH = 7.0, I = 1.0 M $NaNO_3$	5.0	Catalano and Brown (2005)
Goethite	pH = 4.5, I = 0.1 M $NaNO_3$	8.0	Hsi and Langmuir (1985)
Ferrihydrite	pH = 4.5, I = 0.1 M $NaNO_3$	10.0	Waite et al. (1994)
Carbon nanotube	pH = 5.0, I = 0.01 M $NaClO_4$	80.0	Shao et al. (2009)
Oxidized MWCNTs	pH = 5.0, I = 0.01 M $NaClO_4$	140.0	Sun et al (2012)

Source: (Sun et al., 2012)

CONCLUSIONS

This paper highlights uranium contamination and the removal of uranium in wastewaters from mining activities and nuclear power generation. This concern is very timely as the increasing demand for, and shift to nuclear energy production worldwide increases uranium mining activities and may precipitate uranium seepage and leach out of uranium ore. Even though the radiation level of uranium is not high, its toxicity is hazardous for aquatic life and humans. The authors further review various methods of removal of heavy metal contaminants such as uranium. Given existing adsorption techniques, carbon nanotubes can potentially be a superior adsorbent. It is observed that the modified MWCNT, which is economical and environmental friendly, may prove to be a superior adsorbent for the removal of uranium at trace levels.

REFERENCES

Abdelouas, A. (2006). Uranium mill tailings: geochemistry, mineralogy, and environmental impact. *Elements* 2 (6), 335–341.

Agnihotri S., Rood M. J., Rostam-Abadi M., (2005). Adsorption equilibrium of organic vapors on single-walled carbon nanotubes, *Carbon* 43, 2379–2388.

Ahmad Zaidei, R., Abdul Rashid, J., and Normanbhay, S. (2011). A rapid microwave synthesis and partial characterization of carbon nanotubes. ICPE 4 international Conference on postgraduate education.

Aksu, Z. (2005). Application of biosorption for the removal of organic pollutants: a review. Proc. Biochem. 40, 997–1026.

Anirudhan T. S., Bringle C. D. and Rijith, S. (2010). Removal of uranium (VI) from aqueous solutions and nuclear industry effluents using humic acid-immobilized zirconium-pillared clay*, J.Environ. Radioact.* 101,267-276

Anke, M., et al., 2009. Uranium transfer in the food chain from soil to plants, animals and man. Chem. Erde-Geochem. 69, 75-90. Zoriy, P., Ostapczuk, P., Dederichs, H., Hobig, J., Lennartz, R., and Zoriy, M. (2010) Biomonitoring of environmental pollution by thorium and uranium in selected regions of the Republic of Kazakhstan, *J. Environ. Radioactiv.*, 101, 414–420.

Arfsten, D. P., Still, K. R., & Ritchie, G. D. (2001). A review of the effects of uranium and depleted uranium exposure on reproduction and fetal development. *Toxicology and Industrial Health 17*, 180-191.

Arias M., Barral M. T., and Mejuto J. C. (2002). Enhancement of copper and cadmium adsorption on kaolin by the presence of humic acids. *Chemosphere*, 48(10),1081–1088.

Atkinson R., Juras A., Popper T., Stritih J., Ivanova O. (1999). Assessment of the environmental impact of military activities during the Yugoslavia conflict.The Regional Environmental Center for Central and Eastern Europe, Szentendre, Hungary.

Avouris, P. (2002). Molecular electronics with carbon nanotubes Acc. Chem. Res. 35, 1026-1034

Babel, S., and Kurniawan, T.A. (2004). Cr(VI) removal from synthetic wastewater using coconutshell charcoal and commercial activated carbon modified with oxidizing agents and/or chitosan. *Chemosphere*, 54(7), 951-967.

Balasubramanian, K. and Burghard, M. (2005), "Chemically functionalized carbon nanotubes", *Small,* 1(2), 180-92.

Banat IM, Nigam P, Mc Mullan G, Marchant R, Singh D (1996). Microbial decolorization of textile dye containing effluents. A Review. *Biores Technol* 58, 217-27.

Barbette, F.R. (2004). Extraction of uranyl ions from aqueous solutions using silica-gel-bound. *Analytica Chimica*, 179–187.

Barton, C.S., Stewart, D.I., Morris, K., Bryant, D.E., (2004). Performance of three resin-based materials for treating uranium contaminated groundwater within a PRB. *J. Hazard. Mater.* 116, 191–204.

Bargar, J. R., Reitmeyer, R., Lenhart, J. J., and Davis, J. A. (2000). Characterization of U(VI)-carbonatoternary complexes on hematite: EXAFS and electrophoretic mobility measurements. *Geochim. Cosmochim.*64, 2737-2749.

Bem, H., & Bou-Rabee, F. (2004). Environmental and health consequences of depleted uranium use in the 1991 Gulf War. *Environment International 30*, 123– 134.

Bhatnagar, A., Sillanpää, M., (2009). Applications of chitin and chitosan derivatives for the detoxification of water and wastewater – a short review. *Adv. Colloid Interface* 152, 26–38.

Biskup B., and Subotic B. (2004). Removal of heavy metal ions from solutions using zeolites. Influence of sodium ion concentration in the liquid phase on the kinetics of exchange processes between cadmium ions from solution and sodium ions from zeolite. *Separation Science and Technology*, 39(4), 925–940.

Bryant, D.E., Stewart, D.I., Kee, T.P., Barton, C.S., (2003). Development of a functionalized polymer-coated silica for the removal of uranium from groundwater. *Environ. Sci. Technol.* 37, 4011–4016.

Catalano, J. G. and Brown, G. E. (2005). Uranyl adsorption onto montmorillonite: evaluation of binding sites and carbonate complexation. *Geochim. Cosmochim.* 69, 2995-3005.

Chen, C.L., and Wang, X.K. (2006). Adsorption of Ni(II) from aqueous solution using oxidized multi-walled carbon nanotubes. *Ind. Eng. Chem. Res.,* 45,9144–9149Chu W. (1999). Lead metal removal by recycled alum sludge. *Water Research*, 33(13), 3019–3025.

Coleman NT, McClung AC, Moore DP. Formation constants for Cu(II)–peat complexes. Science 1956;123:330

Crini, G. (2006). Non-conventional low-cost adsorbents for dye removal: *A review. Bioresource Technology*, 97, 1061–1085.

Dabrowski, A., (2001). Adsorption, from theory to practice. *Adv. Colloid Int. Sci.* 93, 135–224.

Dobrowolski, R., and Stefaniak, E. (2000). Study of chromium(VI) adsorption from aqueous solution on to activated carbon. *Adsorption Science and Technology*, 18(2), 97–106

Ekmekyapar F., Aslan A., Bayhan Y. K., and Cakici A. (2006). Biosorption of copper(II) by nonliving lichen biomass of Cladonia rangiformis hoffm. *Journal of Hazardous Materials*, 137(1),293–298.

Favre-Re´guillon A., Lebuzit G., Murat D., Foos J., Mansour C., Draye M. (2008). Selective removal of dissolved uranium in drinking water by nanofiltration. *Water Research*, 42, 1160 – 1166

Filov, V. A., Ivin, B. A., & Bandman, A. L. (1993). Harmful chemical substances. New York: E Horwood.

Fugetsu, B., Satoh, S., Shiba, T., Mizutani, T., Lin, Y.B., Terui, N., Nodasaka, Y., Sasa, K., Shimizu, K., Akasaka, T., Yokoyama, A., Mori, M., Tanaka, K., Sato, Y., Tohji, K., Tanaka, S., Nishi, N., and Watari, F. (2004). Caged multiwalled carbon nanotubes as the adsorbents for affinity-based elimination of ionic dynes. *Environ. Sci. Technol.* 38, 6890–6896.

Gafvert, T., Ellmark, C., Holm, E., (2002). Removal of radionuclides at a waterworks. *J. Environ. Radioactivity* 63, 105–115.

Gall, L.F., Mikecz, P., and Papp, T. (1988). Preparation of uranium targets by a chemical method. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 270(2–3), 248–250

Giannardi, C., and Dominici, D. (2003). Military use of depleted uranium: assessment of prolonged population exposure. *Journal of Environmental Radioactivity 64*, 227-236.

Gu, B., Ku, Y.-K., Jardine, P.M., 2004. Sorption and binary exchange of nitrate, sulfate, and uranium Antonina P. Kryvoruchko, Lyudmila Yu. Yurlova, Irina D. Atamanenko, Boris Yu. Kornilovich (2004). Ultrafiltration removal of U(VI) from contaminated water. *Desalination*, 162, 229-236 on an anion-exchange resin. *Enviorn. Sci. Technol.* 38, 3184–3188.

Guo, Z.J., Li, Y., and Wu, W.S. (2009). Sorption of U(VI) on goethite: effects of pH, ionic strength, phosphate, carbonate and fulvic acid. Appl. Radiat. Isot. 67, 996-1000.

Harmsten, K. F., & De Haan, A. M. (1980). Occurrence and behavior of uranium and thorium in soil and water. Netherlands Journal of Agricultural Science, 28, 40–62.

Hsi, C.K.D., Langmuir, D., (1985). Adsorption of uranyl onto ferric oxyhydroxides- application of the surface complexation site-binding model. *Geochim. Cosmochim*, 49, 1931-1941.

Hu, J., Chen, C., Zhu, X., and Wang, X. (2009). Removal of chromium from aqueous solution by using oxidized multiwalled carbon nanotubes. *Journal of Hazardous Materials, 162*, 1542–1550.

IAEA, 2011. Report of the International Review Mission on the Radiation Safety Aspects of a Proposed Rare earth processing Facility (the Lynas Project).

Ismail, B., Redzuwan, Y., Chua, R.S. and Shafiee, W. (2001). Radiological impacts of the *amang* processing industry on neighbouring residents. *Applied Radiation and Isotopes.* 54, 393-397.

Ismail B., Teng, I.Y and Muhamad, S.Y., (2011). Relative radiological risks serived from different TENORM wastes in Malaysia. *Radiation Protection Dosimetry.* 1-8

James, D., Venkateswaran, G., and Rao, T.P. (2009), Removal of uranium from mining industry feed simulant solutions using trapped amidoxime functionality within a mesoporous imprinted polymer material, *Microporous and Mesoporous Materials,* 119, 165–170

Jiang, X., Gu, J., Lin, L., and Zhang Y. (2011). Investigation on the modification to polyurethane by multi-walled carbon nanotubes. *Pigment & Resin Technology,* 40(4), 240–246.

Kalinich, J.F., and McClain, D.E. (2001). Staining of intracellular deposits of uranium in cultured murine macrophages. *Biotech Histochem.* 76(5-6), 247-52.

Kingsnorth, D., IMCOA: "Rare Earths: Facing New Challenges in the New Decade" presented by Clinton Cox SME Annual Meeting 2010, 28 Feb – 03 March 2010, Phoenix, Arizona.

Kutahyali, C., and Eral, M. (2010) Sorption studies of uranium and thorium on activated carbon prepared from olive stones: Kinetic and thermodynamic aspects. *Journal of Nuclear Materials,* 396, 251–256.

Lam, C. W., James, J. T., McCluskey, R., Arepalli, S., and Hunter, R. L. (2006). A review of carbon nanotubes toxicity and assessment of potential occupational and environmental health risks. *Crit. Rev. Toxicol.* 36, 189-217.

Li, H.,Chao, L., Xu, F., and Liang Y. (2004). Preparation of the novel nanocomposite $CO(OH)_2$/Ultra –stable Y zeolite and Its application as a supercapacitor with high energy density. *Adv. Mater,* 16(20), 1853-1857.

Li, P., Chen, B., Husain, T. (2011). IRFAM: an integrated rule-based fuzzy adaptive resonance theory mapping system for watershed modeling. ASCE. *J. Hydrol. Eng.* 16(1), 21-32

Long, R.Q., and Yang, R.T. (2001). Carbon nanotubes as a superior sorbent for nitrogen oxides, *Ind. Eng. Chem. Res.* 40, 4288–4291.

Majdan, M., Pikus, S., Gajowiak, A., Sternik, D., and Zeba, E. (2010). Uranium sorption on bentonite modified by octadecyltrimethylammonium bromide *Journal of Hazardous Materials* 184, 662–670.

McKay, G., and Ho, Y.S. (1999). The sorption of lead(II) ions on peat. *Water Research,* 33(16), 578–584.

Mohan D., and Pittman, C.U. (2006).Activated carbons and low cost adsorbents for the remediation of tri- and hexavalent chromium from water. *Journal of Hazardous Materials,* 137, 762–811.

Nuclear Malaysia (2010). History. Retrieved from http://www.nuclearmalaysia.gov.my/.

Performance Management and Delivery Unit (PEMANDU) (2010). Economic transformation Programme. Kuala Lumpur: Prime Minister Deparment.

Pontius, F.W. (1990) Water quality and treatment. 4th Edition. New York: McGraw-Hill Inc.

Sekar, M., Sakthi, V., and Rengaraj, S. (2004) "Kinetics and equilibrium adsorption study of lead(II) onto activated carbon prepared from coconut shell," *Journal of Colloid and Interface Science,* 279(2), 307–313.

Sublet, R., Simonnot, M.O., Boireau, A., and Sardin, M. (2003). Selection of an adsorbent for lead removal from drinking water by a point-of-use treatment device. *Water Research,* 37(20), 4904–4912.

Sun, Y., Yang, S., Sheng, G., Guo, Z., and Wang, X. (2012). The removal of U(VI) from aqueous solution by oxidized multiwalled carbon. *Journal of Environmental Radioactivity* , 40-47.

Tan, X., Fang, M., Chen, C.,Yu, S., and Wang, X. (2008). Counterion effects of nickel and sodium dodecylbenzene sulfonate adsorption to multiwalled carbon nanotubes in aqueous solution. *CARBON 46* , 1741-1750.

Thostenson, E.T., Ren, Z. and Chou, T.W. (2001). Advances in the science and technology of carbon

nanotubes and their composites: a review. *Composites Sci. Technol.,* 61(13), 1899-912.

TNB, (2011). History. Retrieved from http://www.tnb.com.my/faq.html

Sandoval, J. H. and Wicker, R. B. (2006). "Functionalizing stereolithography resins: effects of dispersed multi-walled carbon nanotubes on physical properties", Rapid Prototyping Journal, Vol. 12 Iss: 5, pp.292 – 303

Shawky, S., Galeel, A.M., and Aly, A. (2005). Sorption of uranium by non living water hyacinth roots. *Journal Radioanalytical and Nuclear Chemistry,* 26(1), 81-84.

Sheppard, S.C., Sheppard, M.I., Gallerand, M.O., and Sanipelli, B., (2005). Derivation of ecotoxicity thresholds for uranium*, J. Environ. Radioactivity,* 79, 55–83.

Srivastava, N.K., and Majumder, C.V. (2008). Novel biofiltration methods for the treatment of heavy metals. *Journal of Hazardous Materials,* 1–8.

Tarasenko, M., Promin, O., and Silayev, A. (1977). Barium compounds as industrial poisons (an experimental study). *J. Hyg. Epidem. Microbiol. Immunol.* 21, 361-373.

Tran, H. V., Tran, L. D., and Nguyen, T. N. (2010). Preparation of chitosan/magnetite composite beads and their application for removal of Pb(II) and Ni(II) from aqueous solution *Materials Science and Engineering C* 30, 304–310.

Waite, T.D., Davis, J.A., Payne, T.E., Waychunas, G.A., Xu, N., (1994). Uranium (VI) adsorption to ferrihydrite e application of a surface complexation model. *Geochim. Cosmochim,* 58, 5465-5478.

Wang, J., and Chen, C. (2009). Biosorbents for heavy metals removal and their future. *Biotechnology Advances*, 27, 195–226.

Wang, X.K., Chen, C.L., Hu, W.P., Ding, A.P., Xu, D., and Zhou, X. (2005). Sorption of 243Am(III) to multiwall carbon nanotubes. *Environ. Sci. Technol.* 39, 2856-2860.

Wang, X.K., Chen, C.L., Nagatsu, M., (2009). Europium adsorption on multiwall carbon nanotube/iron oxide magnetic composite in the presence of polyacrylic acid. *Environ. Sci. Technol.* 43, 2362-2367

Woodard, F. (2001) Industrial waste treatment handbook. Boston: Butterwoth-Heinemann.

VanHorn J.D., Huang H. (2006). Uranium(VI) bio-coordination chemistry from biochemical, solution and protein structural data *Coord. Chem. Rev.*, 250 (7-8), pp. 765–775

Yang, K., Wang, X., Zhu, L., and Xing B. (2006) Competitive sorption of pyrene, phenanthrene, and naphthalene on multiwalled carbon nanotubes. *Environ. Sci. Technol.* 40, 5804–5810.

(Doyurum) Yusan, S., and Akyil, S. (*2008*). “Sorption of Uranium(VI) from aqueous solutions by akaganeite”, *Journal of Hazardous Materials*, 160, 388–395.

Zhou, Y., Liang, Z., Wang, Y. (2008). Decolorization and COD removal of secondary yeast wastewater effluents by coagulation using aluminum sulphate. *Desalination,* 225, 301–311.