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**Organic Material in Clay-Based Buffer Materials
and Its Potential Impact on Radionuclide
Transport**

**Matières organiques dans les matériaux tampons
à base d'argile et leurs effets possibles sur la
migration des radionucléides**

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ABSTRACT

AECL has submitted an Environmental Impact Statement (EIS) to evaluate the concept of nuclear fuel disposal at depth in crystalline rock of the Canadian Shield. In this disposal concept used fuel would be emplaced in corrosion-resistant containers which would be surrounded by clay-based buffer and backfill materials. Once groundwater is able to penetrate the buffer and corrosion-resistant container, radionuclides could be transported from the waste form to the surrounding geosphere, and eventually to the biosphere. The release of radionuclides from the waste form and their subsequent transport would be determined by the geochemistry of the disposal vault and surrounding geosphere. Organic substances affect the geochemistry of radionuclides through complexation reactions that increase solubility and alter mobility, by affecting the redox of certain radionuclides and by providing food for microbes. The purpose of this study was to determine whether the buffer and backfill materials proposed for use in a disposal vault contain organics that could be leached by groundwater in large enough quantities to complex with radionuclides and affect their mobility within the disposal vault and surrounding geosphere.

Buffer material, made from a mixture of 50 wt.% Avonlea sodium bentonite and 50 wt.% silica sand, was extracted with deionized water to determine the release of dissolved organic carbon, humic acid and fulvic acid. The effect of radiation and heat from the used fuel was simulated by treating samples of buffer before leaching to various amounts of heat (60°C and 90°C) for periods of 2, 4 and 6 weeks, and to ionizing radiation with doses of 25 kGy and 50 kGy. Humic substances were isolated from the leachates to determine the concentrations of humic and fulvic acids and to determine their functional group content by acid-base titrations. The results showed that groundwater would leach significant amounts of organics that would complex with radionuclides such as the actinides, affecting their solubility and transport within the disposal vault and possibly the surrounding geosphere. Heating and radiation affect the amount and nature of leachable organics. However, one of the key parameters that needs to be addressed is the mobility of organic molecules through an intact buffer and backfill.

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MATIÈRES ORGANIQUES DANS LES MATÉRIAUX TAMPONS À BASE D'ARGILE
ET LEURS EFFETS POSSIBLES SUR LA MIGRATION DES RADIONUCLÉIDES

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RÉSUMÉ

EACL a présenté une Étude d'impact sur l'environnement (EIE) pour évaluer le concept de stockage permanent du combustible nucléaire à grande profondeur dans des formations de roche cristalline du Bouclier canadien. Selon ce concept, le combustible irradié serait mis en place dans des conteneurs résistant à la corrosion qui seraient entourés de matériau tampon à base d'argile et de remblai. Lorsque l'eau souterraine arrive à s'infiltrer dans le tampon et à pénétrer dans ce conteneur, les radionucléides pourraient être transportés depuis la forme de déchets jusqu'à la géosphère environnante, et, finalement, jusqu'à la biosphère. La libération des radionucléides de la forme de déchets puis leur migration seraient déterminées par la géochimie du dépôt et de la géosphère environnante. Les substances organiques influent sur la géochimie des radionucléides par des réactions de complexation qui augmentent la solubilité et modifient la mobilité, en agissant sur le redox de certains radionucléides et en fournissant des éléments nutritifs aux microbes. L'objet de la présente étude était de déterminer si les matériaux tampons et le remblai que l'on propose d'utiliser dans le dépôt contiennent des matières organiques qui pourraient être lixiviées par les eaux souterraines en quantités suffisamment grandes pour former des complexes avec les radionucléides et influencer sur leur mobilité dans le dépôt et la géosphère environnante.

Un matériau tampon, constitué d'un mélange de 50 % en poids d'argile bentonitique Avonlea (contenant du sodium) et de 50 % en poids de sable siliceux, a été traité à l'eau désionisée en vue de déterminer la quantité libérée de carbone organique, d'acide humique et d'acide fulvique dissous. L'effet des rayonnements et de la chaleur du combustible irradié a été simulé en soumettant les échantillons de tampon avant la lixiviation à diverses quantités de chaleurs (60 °C et 90 °C) pendant des périodes de 2, 4 et 6 semaines, et à des doses de rayonnements ionisants de 25 kGy et 50 kGy. Les substances humiques ont été isolées des lixiviats afin de déterminer les concentrations d'acides humiques et fulviques et de déterminer leur teneur en groupements fonctionnels par titrage acide-base. Les résultats ont montré que l'eau souterraine lixivierait des quantités considérables de matières organiques qui formeraient des complexes avec les radionucléides comme les actinides, influant sur leur solubilité et leur migration à l'intérieur du dépôt et éventuellement dans la géosphère environnante. La chaleur et les rayonnements influent sur la quantité et la nature des matières organiques lixiviables. Toutefois, un des principaux paramètres qui doit être pris en compte touche la mobilité des molécules organiques dans un tampon et un remblai intacts.

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1. INTRODUCTION

Atomic Energy of Canada Limited has assessed the concept of nuclear fuel waste disposal in an engineered vault at a depth of 500 to 1000 m in plutonic rock of the Canadian Shield (Dormuth and Nuttall 1987, Torgerson 1990, AECL 1994). Radionuclide transport in groundwater is the only likely path for radionuclide migration to the biosphere through the mass of rock surrounding a disposal vault. The transport of radionuclides in groundwater systems can be modeled as a chromatographic process in which radionuclides are partitioned between a mobile fluid phase and an immobile solid phase (Freeze and Cherry 1979). The interaction of dissolved contaminants with the immobile rock surfaces is controlled by contaminant solubility and sorption reactions with solid surfaces. An element's solubility and sorption behavior may be affected by pH, redox conditions, ionic strength, and by complexation with inorganic anions and organic ligands. If radionuclides are complexed by organic ligands in solution their mobility will be increased, while a reduction in mobility will be achieved if radionuclides are complexed to organics attached to mineral surfaces. Organics have not been specifically included in performance assessment calculations in granite-based high-level radioactive waste programs throughout the world because of the perception that organic concentrations in granite are too low for significant complexation with radionuclides (Karlsson et al. 1995).

The complexing capacity of naturally occurring organic ligands in groundwater and their potential for facilitating radionuclide transport by complexation with natural organics has recently been addressed (Vilks and Bachinski, 1996, Vilks et al. 1996). The term "facilitated transport" is used to describe any process which may speed up the migration of a contaminant beyond the velocity predicted solely by idealized Darcian flow and sorptive equilibrium with rock surfaces (Huling 1989). Although surface waters in the Canadian Shield can have high concentrations of complexing organic substances, studies of groundwaters from the Whiteshell Research Area (WRA) and the Atikokan Research Area, have indicated that organic concentrations in deep groundwaters of the Canadian Shield are too low to have an impact on radionuclide mobility. However, concern has been expressed over the possibility that organics could be introduced from the emplacement of the waste and engineered barriers (Zuidema 1995).

In the Canadian disposal concept fuel wastes would be emplaced in excavated rooms within plutonic rock and isolated in corrosion-resistant Ti or Cu containers which would be surrounded by a highly compacted clay-based buffer material, consisting of 50 wt.% Na-bentonite and 50 wt.% silica sand. The space between the buffer and the room walls would be filled with a backfill material consisting of 75 wt.% crushed host rock and 25 wt.% glacial lake clay. These clays are known to contain organic matter (Oscarson 1994), and therefore, could be a source of organic matter for the near field and the geosphere in close proximity to the buffer and backfill. The purpose of this report is to evaluate the nature and amount of organic material which could be leached from these clays.

The Na-bentonite that is used in the reference buffer material of the Canadian Nuclear Fuel Waste Management Program (CNFWMP) is Avonlea bentonite, a commercial product produced by Avonlea Mineral Industries Ltd., Regina, Saskatchewan (Oscarson 1994). This clay was

formed as a sedimentary deposit from volcanic ash within the Bearpaw Formation of Upper Cretaceous age (Oscarson and Dixon 1989). The mineralogical composition consists of ~80% smectite (montmorillonite), 10% illite, 5% quartz, with minor amounts of gypsum, feldspar and carbonate. The organic content of this clay is 0.31 wt.% organic carbon (Oscarson 1994).

The clay which is used in the reference backfill material of the CNFWMP is Lake Agassiz clay, which is mined by Kildonan Concrete Products, Ltd., Winnipeg, Manitoba. This clay was formed as a freshwater sediment in glacial Lake Agassiz of Pleistocene age. Its mineralogical composition consists of ~35% smectite, 20% illite, 15% quartz, 10% kaolinite, 10% calcite and minor amounts of feldspar and dolomite (Oscarson 1994). The organic content is 1.2 wt.% organic carbon.

Since clay deposits were formed as sediments in bodies of water, the organic content of their interstitial waters was probably similar to that of modern sediments. The organic content of interstitial waters of aerobic sediments varies between 4 to 20 mg/L as dissolved organic carbon (DOC), while that of anaerobic sediments ranges between 10 and 390 mg/L DOC (Thurman 1986). In a fjord from northwestern Scotland, which received organic matter from a terrestrial source, the DOC of sediment pore water consisted mostly of fulvic-acid-like material (Krom and Sholkovitz 1977). Due to microbial action, in anoxic interstitial waters, DOC accumulates as nonvolatile organic acids, such as fulvic acid, and as volatile organic matter, such as methane (Ben-Yaakov 1973). Therefore, a significant portion of organic carbon in sedimentary clay deposits is likely to consist of humic substances, such as fulvic and humic acids, which are known to complex radionuclides.

As a result of radioactive decay in the used fuel shortly after vault closure, the clay in the buffer will be subjected to radiation fields as high as 52 Gy/h, and temperatures up to 75°C to 95°C, depending upon the arrangement of containers within the vault (Stroes-Gascoyne and West 1994, Baumgartner 1996). The heat and radiation may affect the amount and nature of organic compounds that could be leached from these clays by water. Heating of clay suspensions in groundwater up to 70°C has been shown to increase the amount of leached organic material (Fukunaga et al. 1993). However, the effect may be different for the buffer clay because heating will occur under dry conditions, followed by resaturation with groundwater once the temperature has decreased to natural levels.

Irradiation of sludges was found to increase the concentration of leached organic carbon (Groneman 1976). The total organic carbon (TOC) in landfill leachates was reduced with increased irradiation time (Yamazaki et al. 1982). The irradiation of humic and fulvic acids in solid and solution form by gamma-rays and electrons (8 to 930 kGy) produced changes mainly in particle size, without major changes in structure (Senesi et al. 1977). Aromatic hydrocarbons appear to be more stable than aliphatic hydrocarbons in the presence of ionizing radiation, possibly due to an alkyl side chain cleavage (Dahl et al. 1988). These observations suggest that ionizing radiation from the waste form may produce changes in organic matter which could affect its leachability and structure. Changes in structure would influence its bioavailability and complexation properties.

In the study of WRA organics, attention has been focused on the effect of organics on actinide mobility in groundwater because actinides have isotopes with long half-lives that may pose a potential risk to man. Furthermore, since actinides are believed to be immobile in the geosphere due to their low solubility and strong sorption to mineral surfaces, it is important to address concerns (Mariner and Jackson 1993) that natural organics would complex actinides and make them more mobile than predicted by performance assessment models. The potential effect of organics on radionuclide transport in groundwater systems has been discussed by Carlsen (1989), Choppin and Allard (1985), Norden et al. (1993) and Mariner and Jackson (1993). Organic compounds would have a significant impact on contaminant transport only if they were able to complex with radionuclides or affect their oxidation state, and if they were present in sufficient quantities to compete with inorganic anions.

Naturally occurring organic matter is a complex mixture of humic substances, hydrophilic acids, carbohydrates, carboxylic acids, amino acids, hydrocarbons and other simple organic compounds (Thurman 1986). Actinides can be complexed by organic material containing functional groups such as carboxylate, carbonyl, ether, alcohol, phenol and amine. Fatty acids, amino acids and phenolic compounds are less important for complexing actinides because their complexation constants for actinides are not high and they would require organic concentrations of at least 10 mg/L DOC (Choppin and Allard 1985). Although pigments, such as chlorophyll, have very strong binding constants for metal ions in natural waters and well-aged sediments, the concentrations of these compounds are found to be too low (<10 µg/L) to be important for complexation (Thurman 1986).

The types of organic compounds which have strong enough binding constants and may be present in sufficient quantities in natural waters and sediments to significantly complex cations are hydrophobic acids (including fulvic acid and humic acid), uronic acid and hydrophilic acids (Thurman 1986, Choppin and Allard 1985, Carlsen 1989). The subject of radionuclide complexation with fulvic and humic acids, in particular, has received a great deal of attention in the literature (Kribek and Podlaha 1980, Yamamoto and Sakanoue 1982, Choppin 1988, Moulin et al. 1988, Moulin et al. 1991, Kim et al. 1990).

In addition to producing complexation, humic materials may reduce certain metal ions, possibly due to the presence of hydroquinone structures (Choppin and Allard 1985). For example, soil fulvic acids can reduce Pu(VI) to Pu(IV) (Bondietti et al. 1976), and humics in sea water can reduce Pu(VI), and to a lesser extent Pu(V), to the less soluble Pu(IV) (Choppin et al. 1986).

The organic compounds in clay may stimulate microbial growth, particularly if these organics can be leached by groundwater and transported to local regions of higher porosity. Microbial action could result in gas production, microbe-induced corrosion of containers, or changes in radionuclide migration properties resulting from redox reactions, pH shifts or the release of complexing ligands. The ability of organics leached from clay to induce microbial growth is addressed in another report by Stroes-Gascoyne et al., in preparation.

This report will describe the results of a scoping investigation of the leachability of organic material from the reference buffer material for the CNFWMP. To address the effects of heating and irradiation, buffer samples were exposed to various degrees of heating and irradiation before

being extracted with deionized water. The leachates were analyzed for DOC, fulvic acid and humic acid. The functional group content of isolated humics was determined to look for changes in complexation capacity as a result of heating and irradiation. The leachability of organics was also investigated as a function of solution composition and the number of extraction steps.

2. MATERIALS AND METHODS

2.1 BUFFER PREPARATION AND TREATMENT

The buffer material used in this study was a composite of two batches prepared in 1991 and 1992 (ISO-920902 and BM-910305) at the Underground Research Laboratory for large-scale engineering studies. Both batches contained 50 wt.% Avonlea sodium bentonite and 50 wt.% silica sand, along with Fracture Zone 2 groundwater to bring the moisture content to 18%. Although the buffer had been stored in plastic bags, the moisture content had dropped to between 12 and 13% by the beginning of the experiment.

The effect of temperature was evaluated by heating samples of buffer in an oven at 60°C and 90°C for periods of 2, 4, and 6 weeks. A number of samples were irradiated at the Whiteshell Laboratories using a Gammacell 220 Research Irradiator to evaluate the effects of radiation. One sample set that had been heated for 6 weeks at 60°C and 90°C was irradiated to 25 kGy to evaluate the combined effects of temperature and radiation. Unheated buffer samples, with moisture contents of 13% and 18%, were also irradiated to 25 kGy to study the effects of radiation on clays with different moisture contents. A third sample was heated at 110°C overnight to reduce the moisture content to 0% (0.2%) and then irradiated at 25 kGy to evaluate the effect of radiation on a very dry sample. Another buffer sample, with a 12% moisture content, was irradiated to 50 kGy. The moisture contents of the samples heated at 60°C and 90°C were 1% and 0.5% respectively at the time of irradiation.

2.2 LEACHING OF ORGANICS FROM BUFFER

Samples of untreated, heated, and irradiated buffer were leached with deionized water, and in one set of experiments to evaluate the effect of leaching solution composition, with 0.4 N NaOH. Clay suspensions were made by suspending 500 g of buffer in 1 500 mL of water. After shaking the suspensions for 24 h, the clay and sand were removed from solution by three successive centrifuging steps consisting of 6 000 rpm for 1 h, 1 500 rpm for 1 h, and 50 000 rpm for 40 min. The solutions were then filtered through a 0.22 µm membrane to ensure sterilization for microbial experiments that were part of another study (Stroes-Gascoyne, in preparation).

2.3 SAMPLE ANALYSIS

Leachates were analyzed for DOC by high-temperature (875°C) platinum-catalyzed oxidation using an Ionics Model 1555 carbon analyzer. The samples were acidified with phosphoric acid and purged with O₂ to remove inorganic carbon. Ultraviolet and visible absorbance spectra of leachate samples and isolated humic substances were determined to quantify concentrations of

dissolved humic substances and to provide a tool for comparing organics from different sources. UV-visible absorbance measurements were performed with a Hewlett Packard HP 8452A diode-array spectrophotometer, that can scan produce a full spectrum scan, from 190 to 820 nm, in 0.1 s. Quartz cuvettes were used to allow measurements down to 190 nm. Absorbance spectra were saved on computer for quantification using the HP UV-Vis ChemStation (DOS Series) quantification software. Absorbance was quantified with respect to organic carbon using standards made from fulvic acid extracted from the Winnipeg River.

Humic substances were extracted from leachates using Amberlite XAD-8 resin (Thurman and Malcolm 1981). Amberlite XAD-8 resin is a crosslinked polymeric methacrylate ester that can remove humic substances from water by hydrophobic interactions (Vilks 1995). Leachate samples were acidified to pH 2 to make the humic substances more hydrophobic, and were pumped through a column packed with purified XAD-8 resin (Thurman and Malcolm 1981). Adsorbed humics and other hydrophobic acids were backflushed from the resin with 0.1 N NaOH, which changed these substances to their ionic form, making them less hydrophobic. A portion of the extracted solution was acidified to pH 7 and used to determine the total concentration of extracted humics by either DOC or absorbance measurement. These concentrations were normalized to original leachate values using the volume of leachate that had passed through the resin, and the volume of humic sample extracted from the resin. The remainder of the humic sample was acidified to pH 1 to precipitate humic acid. The humic acid was separated from the soluble fulvic acid by centrifuging at 6000 rpm for 20 minutes. The organic content of the fulvic acid sample was analysed to determine the fulvic and humic acid content of the leachates.

2.4 FUNCTIONAL GROUP DETERMINATION

The functional group content of humic materials was studied by acid-base titrations using an automated, computer-programmed titration system (Tanager Scientific Systems, Ancaster, Ontario). Acid-base titration data were obtained between pH 3 and pH 11 at maximum intervals of 0.1 pH units. The reaction temperature was between 18°C and 21°C and the ionic strength of the organic solutions was adjusted to 0.1 with NaNO₃ to improve electrode performance and minimize changes in ionic strength due to the addition of acid or base. The titration results were modeled with discrete site analysis to obtain the concentrations of acid-base reactive sites (Brassard et al. 1990). In this discrete affinity spectrum model it was assumed that the overall acid-base properties of a sample can be treated as the sum of a mixture of individual monoprotic acids or sites, with pKa (-log of the acidity constant) values ranging from 3 to 11 and spaced at 0.2 pKa units (Kramer et al. 1989). A linear programming approach (Brassard et al. 1990) was used to determine the concentrations and the pKa values of acids that could reproduce the titration data. The concentrations of these acids were normalized to the DOC in each sample and reported as meq/g organic carbon.

3. RESULTS

3.1 DOC AND HUMIC ANALYSES

The results of DOC and humic analyses of buffer leachates are summarized in Table 1 and Figure 1. The reported DOC concentrations in the leachates are an average of results obtained with the carbon analyzer and absorbance measurements. Based on the 0.31 wt.% concentration of organic carbon in Avonlea bentonite, the mixed buffer should have an organic concentration of 1550 mg/kg, giving the 500 g of extracted buffer a TOC inventory of 775 mg. The DOC concentrations of the leachates represent between 1.3 and 4.2% of the TOC inventory. They are also about 10 to 20 times higher than DOC concentrations observed in deep WRA groundwaters (Vilks and Bachinski 1996).

The concentrations of humic substances are given as mg/L organic carbon for fulvic acid, humic acid (Figure 1) and total humics (Table 1). These concentrations are also about 10 to 20 times higher than humic concentrations observed in deep WRA groundwaters (Vilks et al. 1996). The column "Percent Fulvic" gives the fraction of total humics which is made up of fulvic acid, showing that leachable humics consisted of 35% to 100% fulvic acid. The humic substances accounted for between 19% and 90% of DOC leached by deionized water.

The ratio of absorbances for fulvic acid samples at 460 and 660 nm is reported as an E_4/E_6 ratio. Even though the E_4/E_6 ratios have been correlated to molecular weight and degree of humification, these ratios do not provide a direct indication of history, source or molecular weight (Vilks 1995). However, since they are easily obtained as part of fulvic acid concentration measurements, the E_4/E_6 ratios were reported in order to highlight differences in leachable fulvic acid. Fulvic acids generally have higher E_4/E_6 ratios than humic acids (2-5 for soil humic acids), which have a more reddish color resulting in more absorbance at 660 nm (Thurman 1986). The E_4/E_6 ratios shown in Table 1 fall in the range reported for fulvic acids from streams and rivers (Thurman 1986).

3.2 FUNCTIONAL GROUP ANALYSES

The acidity constants and concentrations of acids and functional groups determined by discrete site analysis of acid-base titration data can be presented as affinity spectra for samples of humics leached from buffer (Figures 2 to 4). Functional groups with pKa ($-\log K_a$) values <6 are carboxylic acids; those with pKa values from 7 to 8.5 may represent β -dicarbonyls, enols, and alcohols usually associated with aliphatic carbon structures; and phenolic acids, associated with aromatic carbon, may be represented by pKa values from 9 to 10.5 (Clair et al. 1991). The carboxylic functional groups would most likely be involved in complexation reactions with organics, and therefore would determine the complexation capacity of humic substances.

The total concentrations of these types of functional groups, expressed as meq/g organic carbon, are given in Table 2. Humics leached from untreated buffer by deionized water had higher

carboxyl concentrations per gram carbon, than the average values reported for humics extracted from WRA groundwaters. In WRA humics the average respective concentrations of carboxylic groups, β -dicarbonyls, and phenolic groups, were 7.7 ± 0.6 meq/g, 1.8 ± 0.4 meq/g, and 6.6 ± 1.2 meq/g (Vilks et al. 1996). Humics that were leached from buffer material using 0.4 N NaOH had carboxyl concentrations that are more typical of humic substances extracted from soils and groundwaters. Heating and radiation affected functional group concentrations, although the trends are difficult to predict.

3.3 EFFECT OF HEATING

Heat treatment of buffer before extraction with deionized water resulted in an increase in the amount of leachable organics. There was a general increase in the DOC with temperature and heating duration (Table 1). The amount of leachable total humics also increased with heating, due largely to an increase in the amount of leached fulvic acid. This also tended to increase the E_4/E_6 ratio of extracted humics. Heating also tended to reduce the concentration of carboxyl functional groups and increase phenolic groups in leached humics (Table 2). The number of different carboxylic sites or pKa values increased with heating times greater than four weeks.

3.4 EFFECT OF RADIATION AND MOISTURE CONTENT

Irradiation of buffer did not have a significant effect on the amount of leachable DOC (Table 1). The irradiated sample with 0% moisture content cannot be considered because it was heated at 110°C overnight to remove moisture before irradiation. The irradiated samples with 13% and 18% moisture contents, which had not been heated at all, produced similar DOC as the unheated sample. The amount of total leachable humics and their contribution to the DOC were reduced by radiation. The leachability of humic acid was more significantly reduced than that of fulvic acid. Samples exposed to both heat and radiation contained virtually no leachable humic acid. This implies that either humic acid has been made more resistant to leaching or else that within the leachable fraction of humic substances radiation has converted humic acid to fulvic acid. The E_4/E_6 ratio of leached humics was increased by radiation, with the most noticeable effect occurring in samples with the higher moisture content. In general the effect of radiation was greatest in the buffer with the highest moisture content. This is expected because moisture is required for radiolysis reactions. In most cases the humics leached from irradiated buffer had a significantly higher concentration of carboxyl functional groups per gram of organic carbon, compared to humics which had not been irradiated. Since carboxyl functional groups are believed to be responsible for binding radionuclides to natural organics, exposure to radiation may increase the complexing capacity of organics in buffer materials.

3.5 EFFECT OF LEACHING TECHNIQUE

Experiments were carried out to determine whether subsequent extraction steps would continue to leach organics and whether there would be any differences in the properties of these organics. The buffer mixtures were also extracted with 0.4 N NaOH, which is used to extract humic substances from soils.

Deionized water continued to leach DOC from buffer even with three extraction steps (Table 3). The total amounts of organic carbon extracted from untreated and irradiated buffer represented 4.8% and 7.8% of the buffer's carbon reservoir, respectively. There was a slight reduction of DOC with progressive extraction of untreated buffer. However, the amount of DOC from irradiated buffer appeared to increase with progressive extraction steps. Humic information for the untreated buffer is not available as a function of extraction step because humics were isolated from a combined sample from three extraction steps. The amount of humics from the irradiated buffer decreased with progressive extraction steps, although there were no distinct differences in functional group content between steps (Table 2).

The NaOH leached significantly more DOC than deionized water (Table 3). The total amount of leached DOC represented about 38% of the buffer's total carbon inventory, while the total amount of leached humic substances accounted for 22% of the inventory. The DOC concentrations remained high even after three extraction steps. However, in the third extraction step there was a significant decrease in the total concentration of humics. During the first two extractions the humics were dominated by humic acid, while in the last step fulvic acid accounted for most of the humic substances. The functional group content of humics from the first two extractions is typical of humic substances extracted from water and soil. In the last extraction there was a slight increase in carboxyl functional groups and a very significant increase in phenolic groups.

4. DISCUSSION

Buffer material made with Avonlea bentonite contains organic carbon, some of which is desorbed when buffer is contacted with water. Although not investigated in this study, the proposed clay for use in the backfill (Lake Agassiz clay) has about four times more organic carbon, some of which would be desorbed by contact with water. Therefore, the results of this study would also apply to the backfill, although the effects of heating and radiation would be reduced because of the distance between backfill and waste. The amount of leached DOC in the first extraction represented only 1.3% to 4.2% of the total carbon inventory. However, the DOC concentrations in the leachates were significantly higher than DOC concentrations observed in deep groundwaters from granite.

The total amount of humics extracted with 0.4 NaOH accounted for 22% of the carbon inventory, and by definition (Stumm and Morgan 1981) represented the total content of humic acid and fulvic acid. In soil science humic substances are considered to be made up of three components, consisting of leachable humic and fulvic acids, and of humin, which cannot be extracted with either base or acid. Structurally, humin is similar to fulvic and humic acid, but there are differences in molecular weight and functional group content (Schnitzer and Khan 1972). After three leaching steps 62% of the buffer's organic inventory remained attached to the clay. The amount of humin in this remaining organic fraction was not determined because the amount of leachable DOC remained high in the third extraction step. Since humin also contains functional groups that can complex with radionuclides, one should note that regardless of how much fulvic

and humic acid is leached from the buffer, some organic complexing capacity will remain attached to the clay surfaces.

The organic material in the buffer will be affected by heat and ionizing radiation from the used fuel. Heating increases the amount of leachable fulvic acid and other organic components, leading to an increased DOC. The maximum duration of heat exposure was only six weeks, and it is not clear what effect a longer exposure would have. Radiation reduces the amount of leachable humics, with the greatest impact on humic acid. However, the complexing capacity of humics leached from irradiated buffer may still be significant because the concentration of carboxyl functional groups per gram organic carbon in these humics is significantly higher than from buffers that were not irradiated.

The amounts of DOC and humic materials leached from buffer material is significant. The leached organics have been shown to stimulate microbial growth (Stroes-Gascoyne et al. 1996). In addition, the leached humics have carboxyl functional groups which could complex with radionuclides such as the actinides. If one takes the average concentration of total humics from Table 1 (6 mg/L) and the average density of carboxyl functional groups from Table 3 (16.5 meq/g), the average organic complexing capacity of buffer leachates is 1×10^{-4} eq/L. This complexing capacity is similar to organic complexing capacities observed in Canadian Shield surface waters, which are high enough to affect the chemistry of actinides (Vilks et al. 1996).

The effect of organic complexation on radionuclide mobility within the near-field and the surrounding geosphere depends to a large extent on the leachability and mobility of organics in the buffer. This study has shown that organics will be desorbed from the buffer-clay mixture by contact with water. Certainly the pore water within the buffer will have very elevated concentrations of organics. Within the buffer, the chemistry of those radionuclides, which interact with organic functional groups, will be determined by a partitioning of humic substances between solution and mineral surfaces. Once in solution, the resulting mobility of complexed radionuclides will depend upon the migration properties of organic molecules.

Any groundwater reaching the containers and waste fuel will have much higher concentrations of organics than those predicted from organic concentrations within deep groundwaters of the Canadian Shield. Although the migration of larger humic acid molecules toward the waste form might be restricted, there is no evidence that smaller fulvic acid molecules would not be able to migrate from the buffer, particularly if thermal gradients have induced convective water movement. This would affect the solubility of actinides, such as uranium, and of other potential vault constituents, such as copper.

Organics from the buffer and backfill will also migrate into the geosphere. The impact on DOC concentrations in groundwater in the nearby geosphere will depend on the flux of organics from the buffer and backfill and on the transport of organic molecules away from the near field. The release of organic molecules to the geosphere will depend to a large extent on the physical state of the buffer and backfill. The development of cracks or preferential flow paths would promote a more rapid release of both organic and inorganic constituents. In the case of an intact buffer and backfill the release of organics would be much less efficient than in the leaching tests described in this report, and would be limited by organic mobility within compacted clay. Unless there is a

significant number of open fractures through the buffer and backfill permitting convective transport, any radionuclides that manage to reach the geosphere will have to pass through an organic-rich pore water. Those radionuclides which interact with organics are most likely to be complexed with organic matter. The mobility of these radionuclides would then be determined by the migration properties of the organic molecules with which they are complexed and by the reversibility of radionuclide's attachment to the organic molecule.

5. CONCLUSIONS

This study has shown that the clay materials used to make buffer and backfill mixtures contain organic substances which are leachable by groundwater. Heat and ionizing radiation from the used fuel will affect the amount and nature of organics leached from these clay materials. In addition to enhancing microbial growth, these organics include humic substances which can complex with certain radionuclides, such as the actinides. Once the temperature of the used fuel has decreased enough to allow the buffer and backfill to cool, groundwater will rehydrate the buffer and eventually the waste containers and waste forms. This water will contain significant amounts of organics, from the buffer and backfill, that will complex with uranium and other actinides, affecting their solubility and mobility. The mobility of organic molecules in the backfill and buffer is an important parameter that will determine which type of complexing ligands are likely to reach the container and waste form, or be released to the geosphere. Mobility would also control which type, if any, of organically complexed radionuclides is likely to pass through the buffer and backfill to the geosphere. Therefore, in addition to quantifying the release of organics from compacted buffer and backfill, the mobility of complexing organics in the buffer and backfill needs to be characterized.

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TABLE 1

SUMMARY OF DOC AND HUMIC ANALYSES OF BUFFER LEACHATES

SAMPLE	IRRAD	HEAT TREATMENT (deg. C)	HEATING DURATION (weeks)	DOC OF LEACHATE (mg/L)	FULLVIC ACID (mg/L)	HUMIC ACID (mg/L)	TOTAL HUMICS (mg/L)	PERCENT FULVIC	HUMICS % OF DOC	FULVIC E4/E6 RATIO
untreated		-	-	8.9	1.9	2.2	4.1	46	46	5.5
60-2		60	2	9.8	3.3	2.7	6.0	55	61	10.2
60-4		60	4	12.2	5.2	3.3	8.5	61	77	9.7
60-6		60	6	12.6	3.4	1.8	5.2	66	42	
60-6 25KGY	25 kGY	60	6	15.4	6.0	0.0	6.0	100	43	11.6
90-2		90	2	15.8	7.9	4.3	12.2	65	77	12.0
90-4		90	4	19.6	8.7	7.2	15.9	55	90	14.5
90-6		90	6	17.9	4.4	3.0	7.4	59	39	
90-6 25KGY	25 kGY	90	6	17.2	3.2	0.0	3.3	100	19	14.9
0% moisture	25 kGY	110	0.10	14.5	3.2	0.2	3.4	94	24	13.4
13 % moisture	25 kGY	-	-	9.2	2.3	1.8	4.1	55	35	15.0
18% moisture	25 kGY	-	-	8.5	1.9	0.5	2.4	79	27	16.4
12 % moisture	50 kGY	-	-	7.4	1.6	0.6	2.2	73	29	9.9

TABLE 2

FUNCTIONAL GROUP CONTENT OF HUMICS FROM LEACHATES

Sample	Extraction	Carboxyl Groups (meq/g DOC)	Dicarbonyls, Alcohols and Enols (meq/g DOC)	Phenolic Groups (meq/g DOC)
60°C, 2 weeks		13.0 ± 9.0	1.3 ± 1.1	39.0 ± 17.0
60°C, 4 weeks		9.5 ± 2.3	4.5 ± 3.7	13.0 ± 6.0
60°C, 6 weeks		16.0 ± 3.4	0.1 ± 0.1	23.8 ± 3.4
60°C, 6 weeks, 25 kGy		17.2 ± 2.3	1.2 ± 0.2	6.5 ± 1.9
90°C, 2 weeks		5.3 ± 2.3	1.3 ± 1.1	28.0 ± 14.0
90°C, 4 weeks		24.0 ± 4.0	0.8 ± 0.7	45.0 ± 26.0
90°C, 6 weeks		13.2 ± 2.6	0.4 ± 0.3	10.9 ± 1.1
90°C, 6 weeks, 25 kGy		17.2 ± 0.8	1.1 ± 0.8	4.6 ± 2.6
25 kGy, 0% moisture, 110°C, 0.1 week		30.5 ± 1.3	2.4 ± 1.4	3.0 ± 2.1
25 kGy, 13% moisture		9.1 ± 1.3	1.5 ± 1.1	10.7 ± 2.3
25 kGy, 18% moisture		31.9 ± 4.7	6.4 ± 1.5	12.4 ± 0.8
50 kGy, 12% moisture		45.2 ± 1.5	8.0 ± 3.0	7.2 ± 5.1
DIW leachate, 25 kGy, 13% moisture	1st	9.1 ± 1.3	1.5 ± 1.1	10.7 ± 2.3
DIW leachate, 25 kGy, 13% moisture	2nd	13.6 ± 6.4	1.3 ± 0.0	6.8 ± 0.0
DIW leachate, 25 kGy, 13% moisture	3rd	11.6 ± 2.8	0.7 ± 0.5	9.0 ± 6.4
NaOH leachates, untreated	1st & 2nd	6.1 ± 0.3	4.1 ± 2.1	5.4 ± 3.1
NaOH leachates, untreated	3rd	4.0 ± 2.0	7.3 ± 5.2	37.9 ± 2.6
DIW leachates, untreated	1st, 2nd, 3rd	16.1 ± 1.8	1.3 ± 1.0	8.3 ± 0.3

TABLE 3

EFFECT OF LEACHING METHOD ON EXTRACTABLE ORGANICS

SAMPLE	LEACHING SOLUTION	LEACH #	IRRAD.	DOC OF LEACHATE (mg/L)	FULVIC ACID (mg/L)	HUMIC ACID (mg/L)	TOTAL HUMICS (mg/L)	PERCENT FULVIC	HUMICS % OF DOC	FULVIC E4/E6 RATIO
untreated	DIW	1		8.6	2.4	1.2	3.7	67	42	9.5
		2		8.4	2.4	1.2	3.7	67	44	9.5
		3		7.7	2.4	1.2	3.7	67	47	9.5
untreated	NaOH	1		63.9	10.8	42.8	53.6	20	84	8.3
		2		70.0	10.0	45.8	55.8	18	80	8.4
		3		30.4	4.2	1.8	6.0	70	20	6.3
13 % moisture	DIW	1	25KGy	9.2	2.3	1.8	4.1	55	45	15.0
		2	25KGy	11.8	1.8	2.4	3.4	42	29	18.5
		3	25KGy	18.96	0.98	0.63	1.61	60.869565	8.491561	15.5

ORGANIC COMPOSITION OF BUFFER LEACHATES

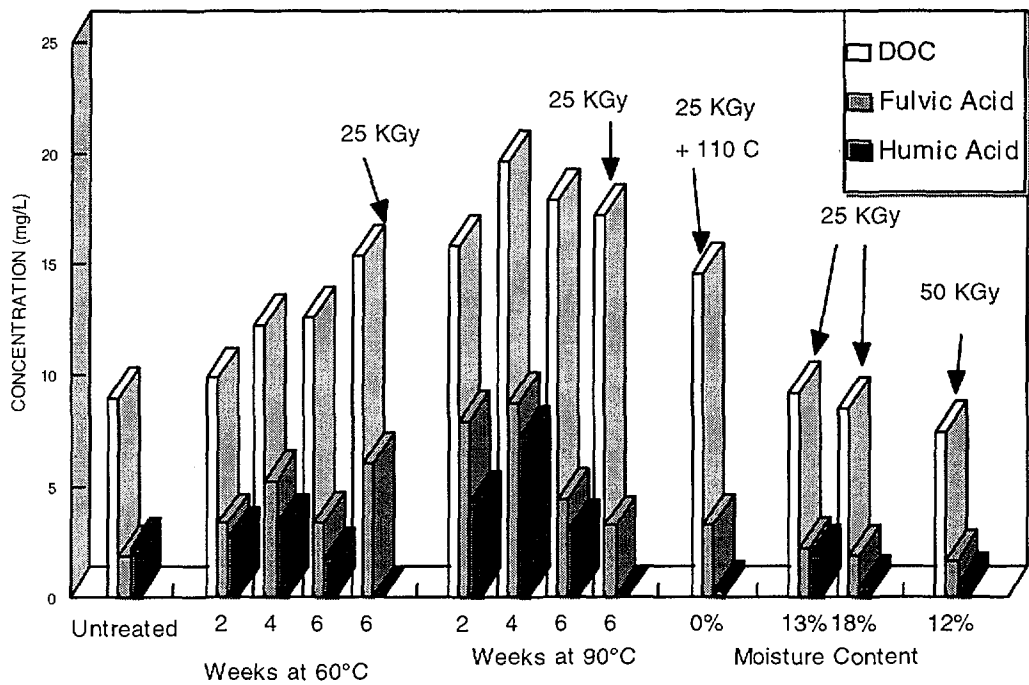


FIGURE 1: Summary of DOC, Fulvic Acid and Humic Acid Concentrations in Leachates From Buffer Exposed to Various Amounts of Heating and Irradiation

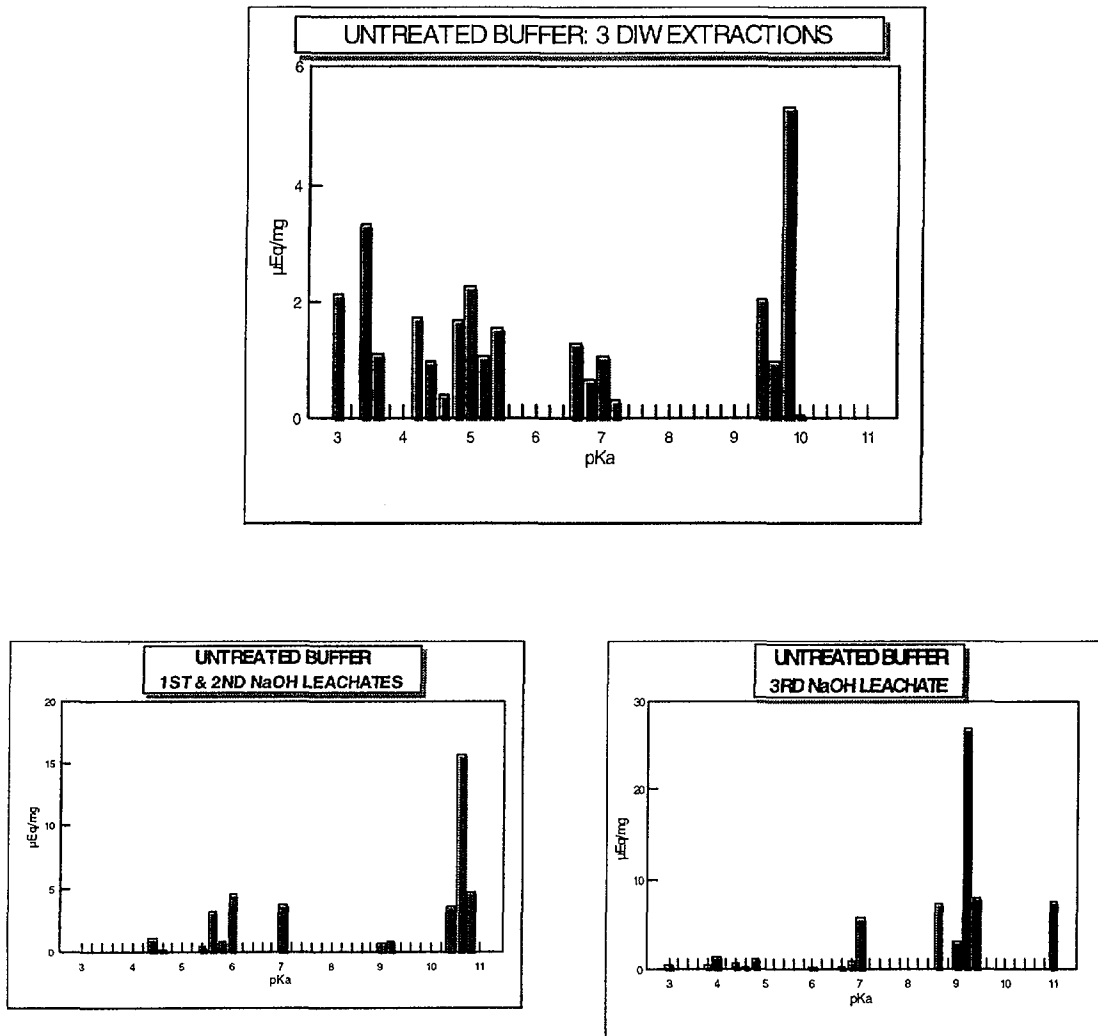


FIGURE 2: Affinity Spectra Determined by Discrete Site Analysis of Acid-Base Titration Data of Humics Leached From Buffer That Had Not Been Treated With Heat or Radiation. Leaching solutions included deionized water (DIW) and 0.4 N NaOH.

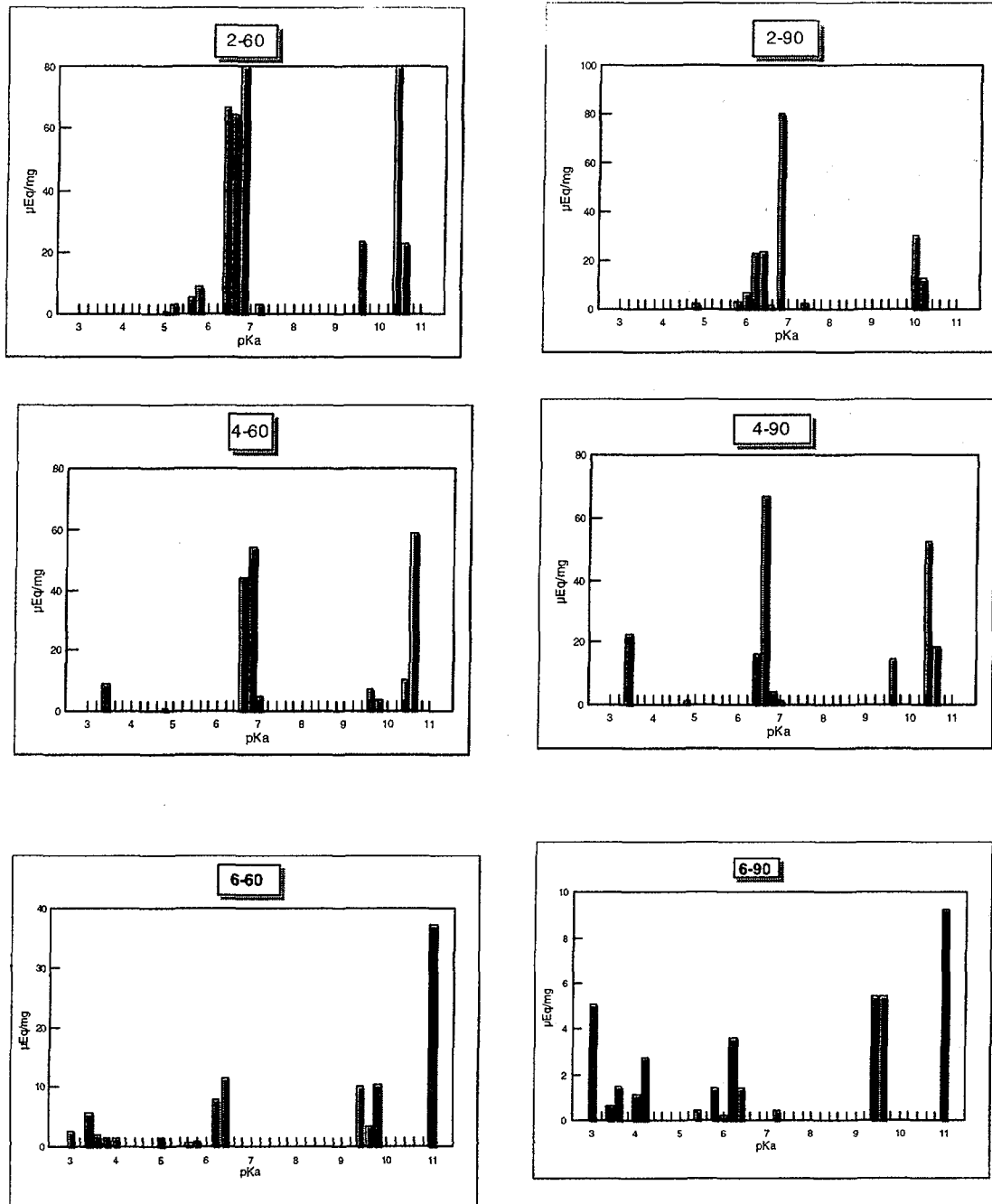


FIGURE 3: Affinity Spectra Determined by Discrete Site Analysis of Acid-Base Titration Data of Humics Leached From Buffer That Had Been Treated With Heat (60°C and 90°C) for Periods of 2, 4, and 6 Weeks. Sample coding is as follows: # of weeks-temperature.

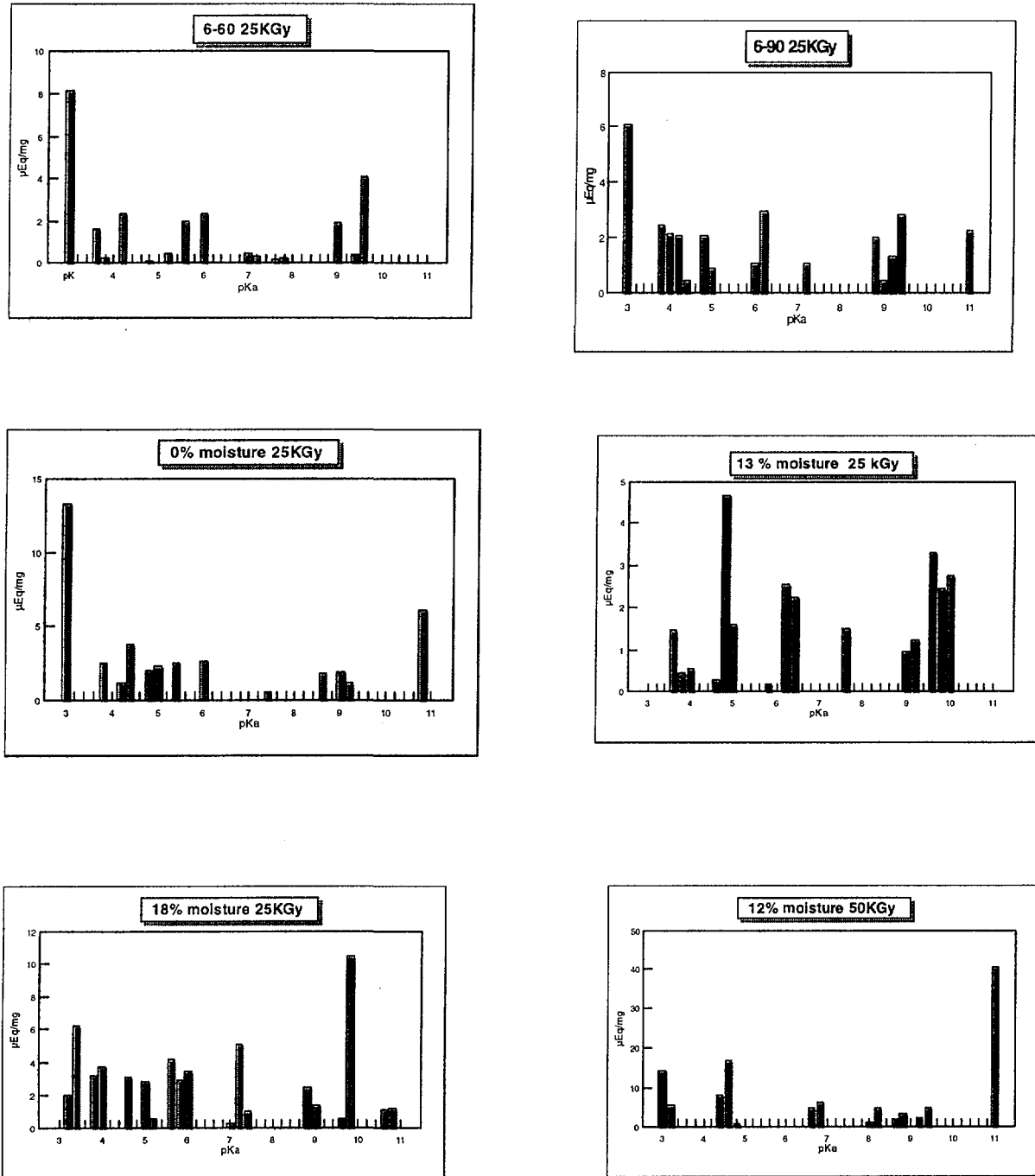


FIGURE 4: Affinity Spectra Determined by Discrete Site Analysis of Acid-Base Titration Data of Humics Leached From Buffer That Had Been Treated With Ionizing Radiation at Various Moisture Levels

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