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DEGRADATION OF CHELATING AND COMPLEXING AGENTS IN AN IRRADIATED, SIMULATED MIXED WASTE

A. P. TOSTE

Department of Chemistry, Southwest Missouri State University, Springfield, Missouri 65804 (USA)

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A simulant of an actual mixed waste was prepared and used to study organic degradation in nuclear wastes. Citric acid, ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA) and N-(2-hydroxyethyl)ethylenediaminetriacetic acid (HEDTA) were added to an inorganic matrix, which contained no radionuclides. This simulated waste was then γ-irradiated in a ^{60}Co-source up to 100 hours. The organics degraded "exponentially" at first, before tapering off; 80% had disappeared by 100 hours. Numerous degradation products (27 in all), mainly complexor and chelator fragments, ranging from small to complex species, were identified by gas chromatography/mass spectrometry. The waste's total organic content decreased with increasing radiation, indicating that radiolytic degradation is dispersive.

Introduction

Mixed wastes, by definition, contain both radionuclides and hazardous chemicals. Research in our laboratory indicates that such wastes may also contain complex mixtures of organics, including both hazardous and non-hazardous compounds.[1,2] The origins of such wastes can be quite varied. Some have been generated during defense-related reprocessing of spent nuclear fuel, while others are generated in commercial nuclear operations.[3-5] One waste in particular, a complex concentrate waste derived from reprocessing spent fuel at the Department of Energy's Hanford site 20-plus years ago, contained numerous nuclear-related organics.[2] Compounds identified include chelating agents like EDTA, NTA and HEDTA (see Table 1 for complete nomenclature) and complexing agents like citric acid, which have been used extensively in the nuclear industry as decontamination agents, etc.[3-5] Considerable research in several laboratories indicates that such compounds may complicate the management of nuclear wastes by destabilizing waste forms, e. g., cementitious grouts, or by enhancing the environmental mobility of certain radionuclides leached from wastes.[6-9]

Perhaps most significant, our analyses of actual wastes also reveal that, in addition to chelating and complexing agents, mixed wastes may also contain numerous structurally-related compounds, occasionally at relatively high concentrations. The complex concentrate waste, for example, contained an abundance of so-called chelator and complexor fragments.[2] Such compounds are presumably derived from the chemical, radiolytic, thermal and/or biological degradation of the parent organics. They have been detected in heated and

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chelator and complexor fragments in actual wastes indicates that the organic content of nuclear wastes is dynamic, not static. This situation may complicate waste management efforts. Like their parent compounds, the diagenesis products may destabilize waste forms or enhance the environmental mobility of radionuclides; in some cases, perhaps even more so than the parent organics themselves.

The identification of these organic degradation products in actual wastes constitutes a major advance in understanding the chemistry of nuclear wastes. However, nothing specific can be deduced about the mechanisms or rates of organic degradation by simply analyzing actual nuclear wastes. The best one can do is to characterize the process *de facto* in the actual waste, which is often a poorly defined and intractably complex system anyway. To be most meaningful, degradation studies should be conducted with well-defined systems in a controlled, laboratory environment.

In this report we outline an approach for carefully studying organic degradation in nuclear wastes by using simulated wastes and report the results of our first degradation study. We opted to examine the effects of radiolysis on organics in mixed wastes by irradiating a simulated mixed waste. We prepared a simulated complex concentrate waste by adding four parent organics, EDTA, NTA, HEDTA, and citric acid, to an inorganic matrix (without any radioactivity), which was formulated based on past analyses of an actual complex concentrate waste.[2] Aliquots of the simulated waste were then γ-irradiated for 0-100 hrs. Each irradiated waste sample was subsequently analyzed to monitor the disappearance of the parent organics and, most important, the appearance of degradation products.

Experimental

Experimental Design

Synthetic Waste Preparation. The inorganic matrix of the simulated waste was prepared according to the following composition, based on past inorganic analyses of an actual complex concentrate waste[2]: HNO_3, 2.60 M; $Al(NO_3)_3 \cdot 9H_2O$, 210 mM; $Fe(NO_3)_3 \cdot 9H_2O$, 38 mM; $Ca(NO_3)_3 \cdot 4H_20$, 25 mM: $Cr(NO_3)_3 \cdot 9H_20$, 4.5 mM; KNO_3, 46 mM; $La(NO_3)_3 \cdot 6H_2O$, 0.28 mM; $Nd(NO_3)_3 \cdot 5H_2O$, 0.98 mM; $Mg(NO_3)_2 \cdot 6H_2O$, 11 mM; $Mn(NO_3)_2$, 7.2 mM; $Ni(NO_3)_2 \cdot 6H_2O$, 8 mM; $Zn(NO_3)_2 \cdot 6H_2O$, 7.4 mM; Na_2HPO_4, 22 mM; NaCl, 41 mM; $Na_2B_4O_7$, 21 mM; $Cu(NO_3)2 \cdot 3H_2O$, 0.42 mM; $Pb(NO_3)_2$, 1.8 mM; $(NH_4)_6Mo_7O_{24} \cdot 4H_2O$, 0.45 mM; $Cd(NO_3)_2 \cdot 4H_2O$, 0.55 mM; NaOH, 6.75 M; $NaNO_3$, 2.04 M; Na_2CO_3, 1.22 M; $NaNO_2$, 800 mM; and Na_2SO_4, 7.2 mM.

Concentrated nitric acid was added to half the final volume of deionized water followed by sequential addition of the above inorganics. The salts were added, in the order specified above, to an acidic solution to enhance their solubility. The solution was neutralized with sodium hydroxide, which contributed sodium nitrate *in situ*, using the nitrate present in the

metal salts. Sodium carbonate, the balance of the sodium nitrate and sodium sulfate were added to a basic solution, since these species are volatile in an acidic environment. The final product was very alkaline (pH 13.5), biphasic (~31% solids) and amber in color.

Waste Irradiations. Four organics were added to the inorganic waste matrix as follows: EDTA, 11.8 mM; NTA, 11.9 mM; HEDTA, 18.4 mM and citric acid, 42.3 mM. The simulated waste was then exposed to γ-radiation in a ^{60}Co-source for 4, 8 and 100 hours to study the effect(s) of radiation on organic degradation. The ^{60}Co-source generated a dose rate of 75,000 ± 10% R/hr and a temperature of 90-95 °C. Each of the irradiated samples, as well as a zero radiation sample, were immediately prepared for organic analysis.

Preparation of Analytical Samples

Replicate 0.5 mL aliquots of each irradiated sample were carefully transferred to 5 mL reaction vials and concentrated to dryness under N_2 at 50 °C. Given the high solids content of the sample, samples had to be vortexed and quickly pipetted to insure uniform sampling. Each sample residue was then methylated using a procedure developed previously,[1] yielding an extract of methylated hydrophilic organics for gas chromatography (GC) and combined GC/mass spectrometry (GC/MS) analysis.

GC Analysis

Capillary GC analyses were performed on a Hewlett-Packard (HP) 5880 gas chromatograph using a procedure described previously.[1] The instrument was also equipped with a flame ionization detector (FID) and a nitrogen-phosphorus detector (NPD).

GC/MS Analysis

GC/MS analyses were performed on an HP 5985 GC/MS instrument in the electron-impact (EI, 70eV) or the chemical ionization (CI) mode with CH_4 as the CI gas using a procedure described previously.[1]

Quantitation

Hydrophilic organics identified in the waste's methylated hydrophilic extracts were quantitated using external standards as described previously. A three-point calibration curve of each GC standard (IDA, citric acid, HEIDA, NTA, EDTA and HEDTA; see Table 1 for full nomenclature) was stored in the gas chromatograph's microprocessor; 2-4 μL of each standard was injected in triplicate. Response factors, expressed as nanograms of analyte per GC area counts, were calculated for each standard under analytical conditions identical to those of the sample analyses. The response factor for citric acid was used to quantitate organic acids, including complexor fragments like the carboxylic acids, presumably derived from the degradation of citric acid. Response factors for the chelating agents EDTA, NTA and HEDTA, as well as the commercially available chelator fragments IDA and HEIDA, were assigned to the chelator fragments unavailable commercially, based on GC behavior and structural similarities.

Materials

Simulated Waste Components. The source-term organics (citric acid, EDTA, HEDTA and NTA) were purchased from Aldrich Chemical Company (Milwaukee, Wisconsin). The inorganics used to prepare the waste matrix were purchased from a variety of chemical suppliers.

Standards. The standards used in the GC analyses (IDA, citric acid, HEIDA, NTA, EDTA and HEDTA) were purchased from Aldrich Chemical Company (Milwaukee, Wisconsin).

Chromatographic Columns. The SE-54 fused silica capillary columns used in the GC and GC/MS analyses were purchased from Supelco, Inc. (Bellefonte, PA).

Reagents, Solvents, and Glassware. The BF_3/methanol (14% w/v) used in the methylation reaction was purchased from PIERCE (Rockford, Illinois). All of the solvents used in the analytical procedure, described previously, were redistilled-in-glass solvents purchased from Fisher Scientific (Fair Lawn, New Jersey). Deionized water, pre-purified for laboratory use, was further purified on a SYBRON/Barnstead NANOpure® system (Barnstead) containing two ion exchange resins and one charcoal filter. All glassware was cleaned in a RBS 35® detergent/deionized water solution (20 ml RBS 35 concentrate/ L water, v/v) followed by NANOpure® water rinses.

Results and Discussion

Degradation of the Source-term Organics

The concentrations of all four original organics decreased with increasing radiation exposure, albeit differentially (Figures la, 1b and Table 1). After 100 hours of radiolysis, virtually all of the HEDTA had degraded (99.8%), followed by EDTA (77.5% degraded), NTA (66.7% degraded) and citric acid (51.1% degraded) (Figure 1b). Depletion also occurred in the zero radiation sample, particularly for HEDTA and NTA, despite the fact that this sample saw no radiation and was quickly processed for analysis to minimize chemical degradation. As Figure 1a illustrates, 14.4% of the collective source-term organics disappeared even without radiation. Analysis of control samples indicated that no significant increase in this zero-radiation loss occurred up to 100 hrs. Most of the degradation is clearly radiologically mediated, but there appears to be a chemically induced component as well.

The loss of parent compounds in the absence of radiation is probably not an analytical artifact. Previous evaluations of the analytical methodology indicate that the procedure is quantitative.[1] Moreover, external standards were used throughout the procedure to assure analytical uniformity. A better explanation is that some of the parent organics appear to be

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very sensitive to the harsh chemical environment of the simulated waste, suggesting that chemically mediated degradation alone may be an important factor in the organic diagenesis of mixed wastes. Studies are currently being designed to test this hypothesis.

The rate of organic degradation in the simulated waste is very vigorous. As Figure 1a

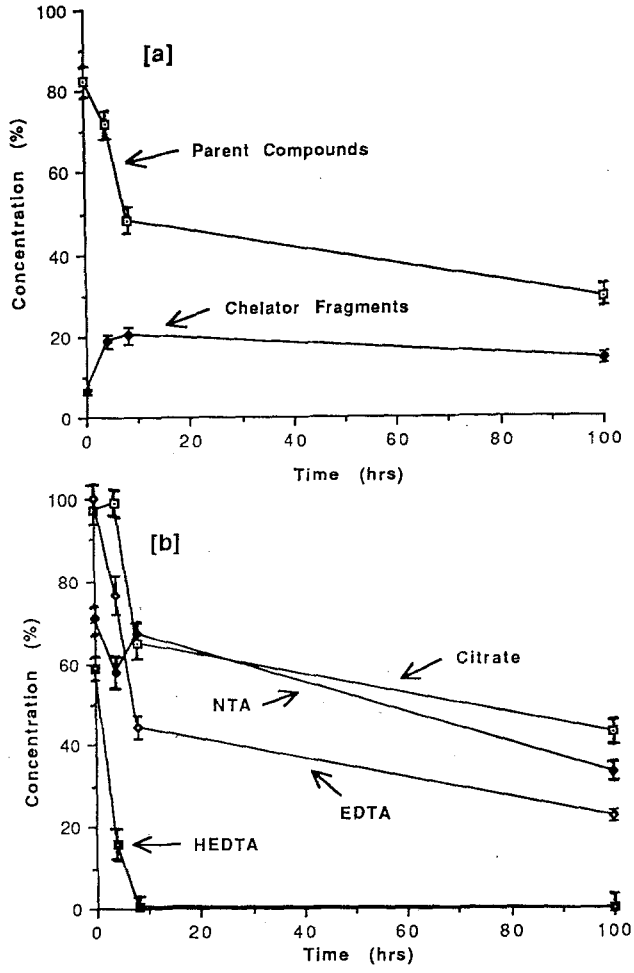

Figure 1. Degradation of a mixture of organics (EDTA, NTA, HEDTA and citric acid) in a simulated mixed waste subjected to γ-irradiation (^{60}Co- source) as a function of irradiation time (0, 4, 8, 100 hr): 1a) Total degradation plots depict the collective disappearance of the parent organics (Citrate, NTA, EDTA and HEDTA) and the collective appearance of degradation products, mainly chelator and complexor fragments; 1b) Individual degradation plots depict the disappearance of each parent organic in the same simulated mixed waste, expressed as the per cent mM ratio of EDTA, NTA, HEDTA or citric acid. Concentration (%) values for both figures were computed as the per cent ratio of total (1a) or individual (1b) organics (mM) remaining or total degradation products (mM) (1a) detected vs. total parent organics (mM) originally added to the simulated waste.

Table 1
Organics Identified in an Irradiated, Simulated Mixed Waste

Compound[c]	Concentration (mM)[a,b] 0 Hr	4 Hr	8 Hr	100 Hr
Parent Organics				
Citric Acid	41.05	41.82	27.46	18.2
Nitrilotriacetic Acid (NTA)	8.43	6.90	7.98	3.97
N-(2hydroxyethyl)ethylenediaminetriacetic Acid (HEDTA) (d)	10.89	2.90	0.07	0.04
Ethylenediaminetetraacetic Acid (EDTA)	11.90	9.04	5.24	2.65
Chelator and Complexor Fragments				
Iminodicarboxy Acid (ICC)	0.12	4.78	1.77	2.24
Iminodiacetic Acid (IDA)		0.09	trace	trace
N-(Methyl)iminocarboxyacetic Acid (MeICA)		trace	trace	trace
N-(2-Hydroxyethyl)iminodiacetic Acid (HEIDA) (d)		0.09	8.05	4.05
N-(Methyl)iminodiacetic (MeIDA)		trace	trace	trace
N-(Ethyl)ethylenediamine-N',N'-carboxyacetic Acid (EtEDCA) (e)				trace
N-(Methylamine)iminodiacetic Acid (MAIDA)	0.12	5.16	3.56	3.24
N-(Carboxy)ethylenediamine-N,N'-diacetic Acid (CEDDA)			0.26	0.01
N-(Carboxy)ethylenediamine-N',N'-diacetic Acid (CEDD'A) (e)				*0.06*
N-(Ethyl)-N,N'-(dimethylamine)propylenediamine-N'-acetic Acid (EtDMAPD'A)		0.35	0.20	0.10
Ethylenediaminetriacetic Acid (ED3A) (e)	0.02	0.28	0.52	0.46
N-(Methylamine)-N'(methyl)ethylenediamine-N,N'-diacetic Acid (MeMAEDDA)		0.15	0.19	0.04
N-(2-Carboxyethyl)iminodiacetic Acid (CEIDA)		0.03	0.04	0.04
N-(Ethylene)ethylenediaminetriacetic Acid (EeED3A)		trace	trace	trace
N-(2-Hydroxyethyliminoacetic)ethylenediamine-N',N'-diacetic Acid (HEIAEDD'A)			trace	
N-(Ethyl)ethylenediaminetriacetic Acid (EtED3A)		0.08	trace	trace
N-(Methylamine)ethylenediaminetriacetic Acid (MAED3A)	0.09	0.09	1.19	0.80
N-(2-Hydroxyethyliminoacetic)-N-(methylidine)-ethylenediamine-N',N'-diacetic Acid (HEIAMEDDA) (d)		trace	trace	trace
[C,N-Di(2-hydroxyethyl)]ethylenediaminetriacetic Acid (DHEDTA) (d)			trace	
Ethylenediaminecarboxytriacetic Acid (EDC3A)		trace	0.02	0.01
Butylenediaminecarboxytriacetic Acid (BDC3A)		trace	trace	trace
Heptadecanoic Acid	0.27	0.35	0.11	0.06
Octadecanoic Acid			0.01	0.05
Propanedioic Acid	4.07	4.07	0.76	0.46
Decanedioic Acid			0.07	0.09
Undecanedioic Acid	0.42	0.13	0.26	0.15
Dodecanedioic Acid	0.4			
Unknown (MW 343)			trace	trace
Unknown (MW 344)			trace	trace
Unknown (MW 345)		trace	trace	trace
TOC Recovery (%)[f]	**90.2**	**82.5**	**61.0**	**27.0**

a) No entry indicates compound is below detection level. b) Exact contributions of unknown organics to waste's total organic content cannot be determined unequivocally. c) Methylated (BF_3/Methanol), acids identified as methyl esters. d) Identified as lactone by GC-MS and past GC-FTIR analyses.[1,2] e) Identified as lactam by GC-MS and past GC-FTIR analyses.[1,2] f) Percent total organic carbon (TOC) computed as the ratio of the concentration (µg C/L) of organics identified vs. nominal concentrations of source-term organics added to the simulated waste.

illustrates, the collective degradation of the source-term organics is very rapid, with the kind of exponential behavior one might expect in radiolysis reactions. Initially, the source-term organics disappear very rapidly, up to ~8 hrs, after which the process slows considerably. Both the rate and extent of degradation varies, depending on the specific organic. EDTA, citric acid and HEDTA degraded exponentially, but HEDTA degraded much more rapidly and extensively than the other compounds (Figure 1b). Its marked depletion in the zero radiation sample suggests that HEDTA is particularly sensitive to chemical degradation, followed by very vigorous radiolysis. Citric acid's degradation begins slowly but picks up quickly, acquiring exponential-like behavior. In contrast, NTA's degradation is relatively linear, suggesting that it is less susceptible to degradation. The same general trend in relative stabilities has been observed in a solution study.[13] Perhaps, the many cations present in the simulated waste protect NTA from radiation damage, whereas the EDTA and particularly the HEDTA are much more susceptible.

A number of factors likely contributed to the differential degradation of the various organics, particularly the zero-radiation disappearance of NTA and HEDTA; probably a combination of pH, metal ions, temperature (a by-product of nuclear decay) and radiation, of course. The harsh chemical environment and the high radiation are certainly the obvious energy sources. However, the operating temperature of the ^{60}Co-source, 90-95 °C, must also be considered, although solution studies indicate that thermal degradation of such compounds is most pronounced above 200°C.[4,12]

The literature contains numerous examples of solution studies on the degradation of chelating and complexing agents. For example, there is ample evidence that cations bind differentially to chelating and complexing agents, altering their structural and chemical properties, including susceptibility to degradation.[3, 14] Studies have been carried out on the thermal or radiolytic decomposition of chelating and complexing agents in aqueous solutions.[11-13] However, very little data exist in the literature on the combined thermal/radiolytic degradation of chelating and complexing agents over extended periods of time, simulating the ages of actual wastes, decontamination operations, or waste management procedures such as volume reduction .[5] Moreover, most of the information available on the thermal and radiolytic stability of chelating and complexing agents deals with synthetic, single-component solutions. The complex composition, e. g., 18 different cations, of the simulated waste used in this diagenesis study precludes any facile comparisons.

Formation of Degradation Products

Numerous chelator and complexor fragments, 27 in all, were identified by GC/MS analysis (Table 1). They all bear structural similarities to one or more of the parent organics. The diagenesis products range from a small fragment like iminodicarboxy acid (MW 105) to large species like HEIAMEDDA (MW 320; see Table 1 for complete nomenclature). The larger diagenesis products are presumably derived from the recombination of reactive, smaller fragments formed during radiolysis. Such behavior is not surprising given the

energetic nature of radiolysis reactions. Considering the experimental design of this initial study, i. e., a mixture of four organics in the waste matrix, and the complexity of the pool of degradation products identified, it is not possible to trace the origin of each chelator or complexor fragment. Experiments with individual organics need to be performed, and are currently being planned. However, judging from the structural complexity of the products identified, it is clear that the degradation of chelating and complexing agents is varied and complex.

A number of carboxylic acids and unknown compounds were also identified in several irradiated samples. Based on the GC/NPD analyses and mass spectral fragmentation patterns, these compounds do not contain nitrogen atoms, and are, therefore, presumably derived mainly from citric acid degradation, probably with some involvement from the acetate ligands of the chelating agents. The dicarboxylic acids, in particular, are probably derived from citrate degradation. Again, individual degradation studies will have to be performed to substantiate these speculations.

Comparison of the organic content of the irradiated simulants and the actual waste is very revealing. Most of the chelator and complexor fragments identified in the actual waste[2] were also identified in the simulated waste; fairly good agreement given the disparate and complex origins of the two "wastes." For example, chelator fragments like ED3A, IDA and HEIDA are major components in both wastes. Carboxylic acids were also identified in both wastes.

Examination of the literature suggests even more parallels. In those solution degradation studies where the products were actually analyzed, various species were detected, ranging from gases, e. g., H_2, CO, CO_2 and NH_3, to alkylamines and chelator fragments.[4,11] EDTA degrades into IDA and HEIDA, even autocatalytically in the presence of Fe^{3+}.[4] NTA degraded more slowly, and MeIDA was the major product.[4] All of these chelator fragments, and many more, were identified in both the actual and simulated wastes.[2]

The shape of the complexor/chelator appearance curve suggests a complex process. The formation of degradation products is initially positive (increasing slope up to ~8 hr), but then declines (Figure 1a). The concentrations of most chelator/complexor fragments in Table 1, particularly major components like ICC, HEIDA, MAIDA and MAED3A, increase and peak between 4-8 hr, followed by a decline. The exception is propanedioic acid. Its concentration is highest and nearly constant up to 4 hr, followed by a rapid decline to 8 hr, suggesting, perhaps, that it is formed mainly via the early burst of chemical degradation, followed by degradation via radiolysis. A likely explanation for the eventual loss of all the fragments is that the formidable radiation energies eventually begin to cause a net degradation of those degradation products formed initially into other species which lie outside the range of the analytical procedure.

Organic Recoveries and Radiodiagenesis

The mass balance for the organics of each waste irradiation was computed by measuring the total organic carbon (TOC) recovery of each incubation, using a procedure sensitive to one part per million. The TOC content was, in turn, computed by quantitating each organic species as μg C/L before and after radiolysis. The total TOC recovery declined from 90.2% to a low of 27.0% with increasing radiation (Table 1). As Figure 1a illustrates, 70.5% of the source-term organics had disappeared after 100 hrs of radiation. Again, based on past analyses and methods validation studies, not to mention the high TOC recovery of the zero-point incubation, the loss does not appear to be due to inherent deficiencies in the analytical procedure used. The explanation is undoubtedly dispersive degradation. Apparently, the formation of chelator and complexor fragments simply cannot keep up with the degradation of the parent compounds and the fragments themselves. This is undoubtedly true, but there is also an analytical explanation.

The loss of TOC may also be due to procedural limitations. The methylation reaction, combined with GC and GC/MS analysis, has worked very well, even quantitatively, for analyzing the source-term organics, as well as the chelator and complexor fragments.[1,2] However, no single analytical procedure is capable of isolating and characterizing the plethora of organics in wastes. Given the high radiation energy, it is likely that degradation products are formed which lie outside the analytical "scope" of the procedure. Based on solution studies in the literature, compounds like alkylamines, gases like CO_2, and volatile organics are among the likely candidates for the missing organics.[4,11] Expansion and refinement of the analytical scheme, to include additional procedures like amine derivatization and purge-and-trap analysis for gases, are currently being planned.

Concluding Remarks

Based on this study, combined with past analyses of actual wastes, it is clear that chelating and complexing agents in a mixed waste undergo vigorous and dispersive degradation into numerous chelator and complexor fragments. The high radiation field of the actual complex concentrate waste, ~1 Ci/L, is certainly more than enough to produce such organic degradation.[2] However, it is also likely that the waste's thermal energy and harsh chemical environment, combined with the mitigating effects of metal ions, all contribute to organic degradation.

The experimental strategy used in this study for studying organic degradation in nuclear wastes appears to be very promising. The organic contents of the actual and simulated complex concentrate wastes proved to be very similar. The correlations are very compelling, given the disparate origins of the wastes, and tend to validate the concept of using simulated wastes to study organic degradation. Another, more practical, argument can be offered for simulation studies. Use of simulated wastes, without the radiation, permits younger students, who are otherwise "genetically impressionable," to participate in nuclear-related research, without the hazards of radiation exposure. Otherwise, student participation in such

research is very difficult, if not impossible, practically speaking. Given the acknowledged and growing need for developing future scientists and engineers in waste and environmental management, new approaches, like the one proposed in this study, need to be developed and implemented.

The findings of such laboratory studies on simulated wastes should be immediately beneficial to engineers charged with the daunting task of managing actual mixed wastes. For example, consider the differential degradation rates of the souce-term organics, particularly the rapid and virtually complete degradation of the HEDTA. This information could be used to facilitate the selection of specific agents for future nuclear operations, e. g., decontamination campaigns. Given the potentially adverse impact of chelating agents on a waste's management, it might be advantageous to select a chelating agent which degrades most extensively. Care needs to be exercised, however, before jumping to facile conclusions. It is clear that organic degradation is very complex, but very little is currently known about the specifics of the process itself.

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