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Integrating Microbiology into the Drigg Post-Closure Radiological Safety Assessment

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ABSTRACT

BNFL owns and operates the UK's principal solid Low Level Radioactive Waste disposal site at Drigg in Cumbria, north west England. Drigg has been receiving waste since 1959 with approximately 900,000 m^3 of waste disposed of to date. Waste accepted for disposal at Drigg comes in a variety of forms including rubble, spoil, redundant equipment, scrap and process waste, and typically contains significant metallic and cellulosic components. The organic content of the waste means that microbial activity plays a significant role in the development of the repository environment. Consequently, microbial processes are integrated into many aspects of the Drigg Post-Closure Radiological Safety Assessment (PCRSA). This begins with the identification and screening of relevant features, events and processes, through supporting research, engineering designs and finally integration into radiological safety assessment modelling. This paper outlines how and where microbiology is integrated into the Drigg PCRSA and indicates areas of active research.

INTRODUCTION

BNFL owns and operates the UK's principal solid Low Level Radioactive Waste (LLW) disposal site at Drigg in Cumbria, north west England. Drigg is a near-surface disposal facility that has operated since 1959 with approximately 900,000 m^3 of waste disposed of to date. Historically, waste was loose tipped into shallow trenches with a natural, low permeability layer forming the base. More recently, disposal has been via a containerized, compacted and grouted waste form to a concrete lined vault. Current and future vaults will extend the operational lifetime of the Drigg site into the middle of this century.

Waste accepted for disposal at Drigg must not exceed 4 gigabequerels per tonne alpha and 12 gigabequerels per tonne other activity. These are upper limits and the bulk of the waste accepted falls substantially below this activity content. The waste comes in a variety of forms including rubble, spoil, redundant equipment, scrap and process waste, and typically contains significant metallic and cellulosic components. The presence of this degradable organic material in the Drigg waste stream means that microbial activity is a natural consequence of disposal operations at the site and that microbial activity plays a significant role in the development of the repository environment [1]. Consequently, microbial processes are integrated into the Drigg Post-Closure Radiological Safety Assessment (PCRSA) [2].

UNDERLYING ASSUMPTIONS

There are three basic assumptions that underlie the integration of microbiology into the Drigg PCRSA:

- microorganisms are ubiquitous in the near-surface environment;
- if a microbial habitat is available then a microbial community will exploit it; and
- microbiology can be addressed at the processes rather than the species level.

The first two assumptions are supported by an ever increasing body of evidence indicating that the geosphere is a microbially active environment playing host to established microbial communities. There is also evidence specific to Drigg indicating microbial populations within the site [1,3] and in the associated drift geology [4]. The third assumption recognizes that although there are many thousands of identified microbial species and potentially millions yet to be identified [5] all these populations can be classified by the processes they catalyze.

ANALYSIS OF FEATURES, EVENTS AND PROCESSES

An analysis of Features, Events and Processes (FEPs) is an integral part of any repository performance assessment. In the Drigg PCRSA interaction matrices (Figure 1) [6] have been employed to determine all relevant FEPs [2]. The leading diagonal elements of these interaction matrices were selected via a sequential disaggregation of the Drigg disposal system [2]. At the lowest level of this disaggregation water, gas and micro-biota occupied leading diagonal elements since they were considered to be ubiquitous components of the Drigg disposal system. This approach was cascaded through the entire Drigg process system with microbiological interactions being investigated in the near-field, geosphere and biosphere. A large number of microbiology related interactions were identified and these are summarized in Table I. All these FEPs are considered in the Drigg PCRSA to some extent, either through integration into the PCRSA calculations, via side studies or logical arguments based on current understanding.

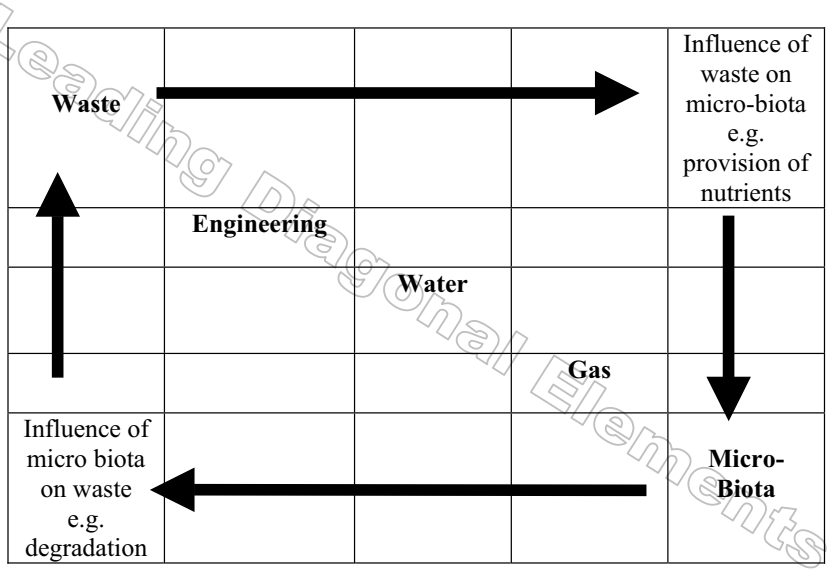

Figure 1. Example Interaction Matrix

Table I. Microbiology related FEP relevant to the Drigg disposal system

	FEP	Explanation
Features	Factors influencing microbial growth and metabolism	Moisture content, pH, pe, surfaces for growth, provision of nutrients and substrates, presence of inhibitors and toxins, temperature, radiation
	Factors affecting microbial distribution	Transport and inoculation, retention through sorption.
	Barriers to transport and release	Barriers which affect ingress or release from the site e.g. waste form, cut-off wall, cap, clay layers, etc.
EVENTS	Effects on growth and metabolism	Change to pH, depletion of substrate, source of toxins resulting in inhibition.
	Effects on water flow	Biological clogging, infilling of cracks and pore spaces by cells and/or polysaccharides.
	Temperature effects	Heat generated by chemical or microbial reactions or radioactivity.
	Influence on particulate and colloidal material	Uptake and transport of radionuclides by microbial cells, stabilization of inorganic colloids by cells or microbial products.
	Formation of films on surfaces	Formation of sorption sites, blinding of existing sorption sites, over coating of sorbed or precipitated radionuclides.
Processes	Influence of microbiota on solution chemistry through production, consumption or accumulation.	Changes to: pH/pe; concentration of aqueous species, complexing agents, dissolved gases, humic materials, simple organics (VFAs), plasticisers, polysaccharides; uptake and accumulation of dissolved species; sorption of dissolved species; isotopic fractionation.
	Generation and utilization of gases	Ammonia, carbon dioxide, hydrogen, hydrogen sulphide, methane, nitrogen, VOCs, other trace gases (N_2O, CO), methylated gases; C-14 and H-3 substituted gases.
	Degradation and deterioration of materials	Degradation of: putresibles and cellulosic wastes; microbially-influenced corrosion of metal, cement, concrete and grout; degradation of aggregates, gravel, clays, minerals; degradation of plastics, paints and resins.
	Changes to properties of wastes and materials	Waste volume reduction, formation of void space, filling of pores and cracks
	Deposition of new solids	Biomineralization, biofilms, formation of organic and humic materials.
	Water flow providing routes for microbial transport	Inoculation into site, migration around site, contact with wastes, release from site.
	Straining or screening of microbes	Sorption onto wastes or other materials, physical straining of cells.
	Evolution and mutation	Changes to species, populations, morphology, etc.

WASTE DEGRADATION AND GAS GENERATION

The Drigg site and associated waste contain a wide range of nutrients and substrates capable of sustaining a broad range of microbial processes [3] (Table II). Site investigations and experimental studies both indicate that microbial gas generation is a significant process for the Drigg site [1,3,7], with methane and carbon dioxide being the most common gases. This can be clearly seen from small (10 L) and larger scale (215 L) waste degradation experiments (Figure 2, Table III). These experiments consist of simulated LLW, Drigg rainwater and Drigg soil in sealed vessels. These systems consistently generate methane and carbon dioxide for extended time periods (Figure 2). Methane generation curves from these experiments are being used to determine cellulose degradation rates for modelling studies by back calculating the amount of cellulose represented by the methane detected. These curves suggest that there are two phases of gas generation (Figure 2) giving two cellulose degradation rates (Table IV). There are inherent uncertainties in these calculations since they do not take into account methane originating from hydrogen oxidation or cellulose that has been metabolized into soluble products or microbial biomass. Interactions between microbiology and corrosion are more difficult to establish but it is likely that corrosion hydrogen represents a significant energy source for the Drigg microbial population and that some of the methane detected is generated via the oxidation of hydrogen. Research is ongoing into providing more direct estimates of cellulose degradation rates. Samples taken from these experiments indicate microbial attachment to all types of waste materials (paper, plastics, neoprene) but evidence of a structured biofilm is inconclusive. Molecular biology analysis of leachates and waste materials from these experiments indicate a varied microbial community with cellulolytic *Clostridia*, Sulphate Reducing Bacteria (SRB) and methanogens being detected. The Drigg disposal system supports a variety of SRB. Investigation of the Drigg trenches indicates that a significant proportion of the Iron Reducing Bacteria (IRB) are SRB utilizing an alternative electron acceptor. Experimental studies to investigate uranium reduction by the Drigg IRB have been unsuccessful although it has been reported in the literature [8].

Table II. Summary of supporting evidence for the microbial processes

Microbial Process	Site Observation	Electron Donor	Electron Acceptor	Waste Degradation Studies	Enrichment Cultures	Direct Identification
Aerobic metabolism		Present	Present	✔	✔	
Denitrification		Present	Present		✔	
Fermentation	✔	Present	Present	✔		✔
Iron reduction		Present	Present		✔	✔
Sulphate reduction		Present	Present	✔	✔	✔
Acetogenesis		Present	Present	✔		
Methanogenesis	✔	Present	Present	✔	✔	

Investigation of the Drigg vault environment suggest a similar evolution to that seen in the trenches. Long term, cemented monolith experiments (>5 years) are yet to produce data on gas generation. Smaller scale experiments at low grout to water ratios indicate a typical anaerobic evolution with the generation of volatile fatty acids (VFA) and a mildly acidic environment. These suggest that rates of microbial activity are sufficiently high to control the chemical evolution. Additional experiments at higher grout to water ratios are underway to further expand on these findings. Simple scoping experiments which combined Drigg grout, cellulose and rainwater [9] did not establish the generation of alkaline cellulose degradation products such as

Iso-saccharinic acid. Although no inoculum was added to these experiments, a significant microbial population developed. This is an area of ongoing research and work is currently ongoing to confirm these findings and expand understanding of the Drigg vault environment.

Table III. Gas generation from large sale (215 L) trench simulants.

Time/ Days	Replicate	N_2 (%)	O_2 (%)	CO_2 (%)	CH_4 (%)	Ar (%)	H_2 (%)
31	1	79.66	17.1	2.21	0.006	0.93	0.085
	2	78.61	19.49	0.85	0.004	0.92	0.14
	3	78.84	18.5	1.73	0.004	0.92	0.003
149	1	78.48	1.66	15.63	3.3	0.92	0.007
	2	78.97	0.38	19.4	0.3	0.92	0.035
	3	78.32	10.43	10.28	0.05	0.92	0
430	1	0.83	0.13	27.69	71.32	0.018	0.007
	2	0.3	0.1	29.88	69.7	0.01	0.005
	3	43.04	2.27	24.45	29.76	0.48	0.009
899	1	0.36	0.12	27.27	72.23	0.013	0.005
	2	0.17	0.14	29.56	70.11	0.007	0.001
	3	3.07	0.1	22.35	74.43	0.05	0.004
1172	1	5.14	0.14	23.35	71.29	0.061	0.012
	2	1.63	0.5	25.85	71.98	0.024	0.007
	3	1.24	0.18	19.91	78.64	0.02	0.008

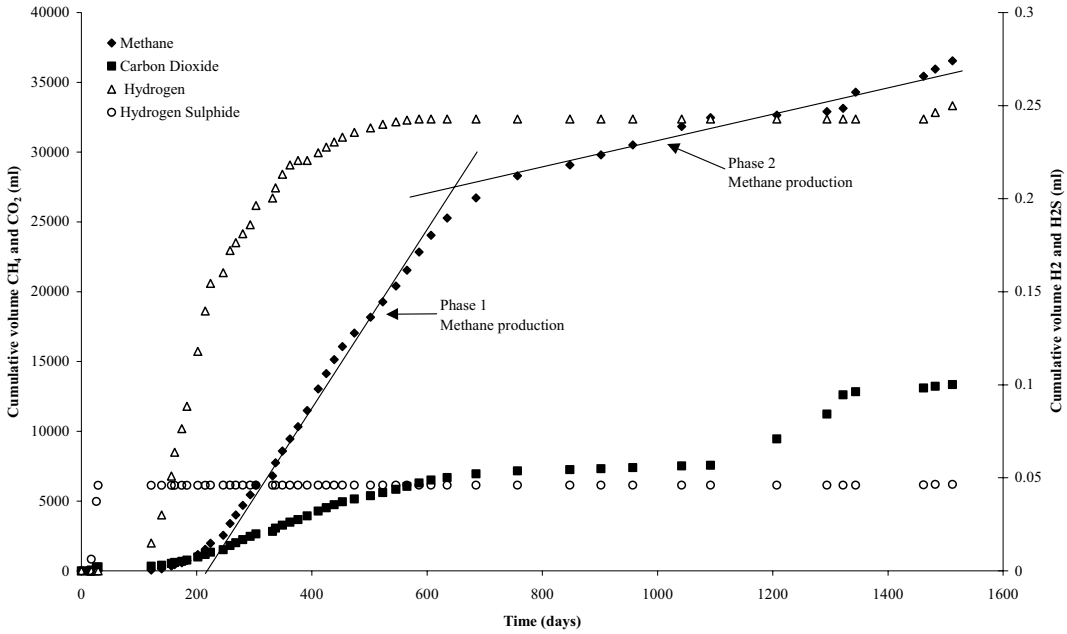

Figure 2. Gas generation from one of a set of small scale (10 L) trench simulants.

Table IV. Cellulose hydrolysis rates derived from the small scale trench simulants

Replicate	Initial rate (s^{-1})	Secondary rate (s^{-1})
1	4.122×10^{-9}	7.927×10^{-10}
2	3.139×10^{-9}	4.756×10^{-10}
3	4.122×10^{-9}	5.391×10^{-10}

POST-CLOSURE ENGINEERING

The proposed post-closure engineering design for the Drigg site is composed of a multi-layer cap, cut-off walls and an engineered drainage system. The design of the cap is influenced by microbial waste degradation both due to gas generation and waste settlement. Consideration of gas generation has resulted in the cap design incorporating a gas release pathway. This granular gas release blanket is designed principally to allow gas to escape from the site in a controlled manner. The accommodation of gas release into the cap design has implications on the assessment of risks associated with radon release since the gas vents provide a route for radon release. Waste degradation along with corrosion and the collapse of voids within the waste contributes to the amount of settlement likely to occur within the site. Estimates of settlement post 2100AD when the final cap will be installed suggest that the cap will have to withstand settlement of approximately 600mm for the trenches and between 200 and 300mm within the vaults. These calculations take into account waste degradation prior to final capping and the overburden provided by the cap itself. These estimates of the likely degree of settlement are factored into cap designs to ensure that the cap can continue to perform as required after settlement has occurred.

The proposed engineering design includes an engineered drainage system to conduct any resulting leachate out of the site via an acceptable pathway. This approach has been adopted since it cannot be assured that a resistive layer or cut-off barrier will remain 100% effective; in the event of some water ingress into the site, the resulting leachate has to be conducted away from the site to prevent potential “bathtubbing”. Column experiments are currently being run to investigate the ability of simulated Drigg leachates to clog both artificial and natural drainage features. Results to date indicate that some reduction in pore volume occurs but not sufficient to cause failure. Microbial activity in the columns results in the generation of methane and carbon dioxide with the removal of VFA and sulphate. Tracer tests to investigate the impact of biofilms and minerals precipitated within the columns on the retardation of radionuclides are currently underway.

RADIOLOGICAL SAFETY ASSESSMENT

The Drigg PCRSA integrates microbial activity into the conceptual and mathematical models it employs. This is particularly the case for the aqueous source term model and the gaseous exposure pathways. The source term for the Drigg PCRSA is calculated using the DRINK (DRIgg Near-field Kinetic) model which utilizes the BNFL Generalized Repository Model (GRM) [3,10,11]. GRM considers kinetically controlled steel corrosion and microbial

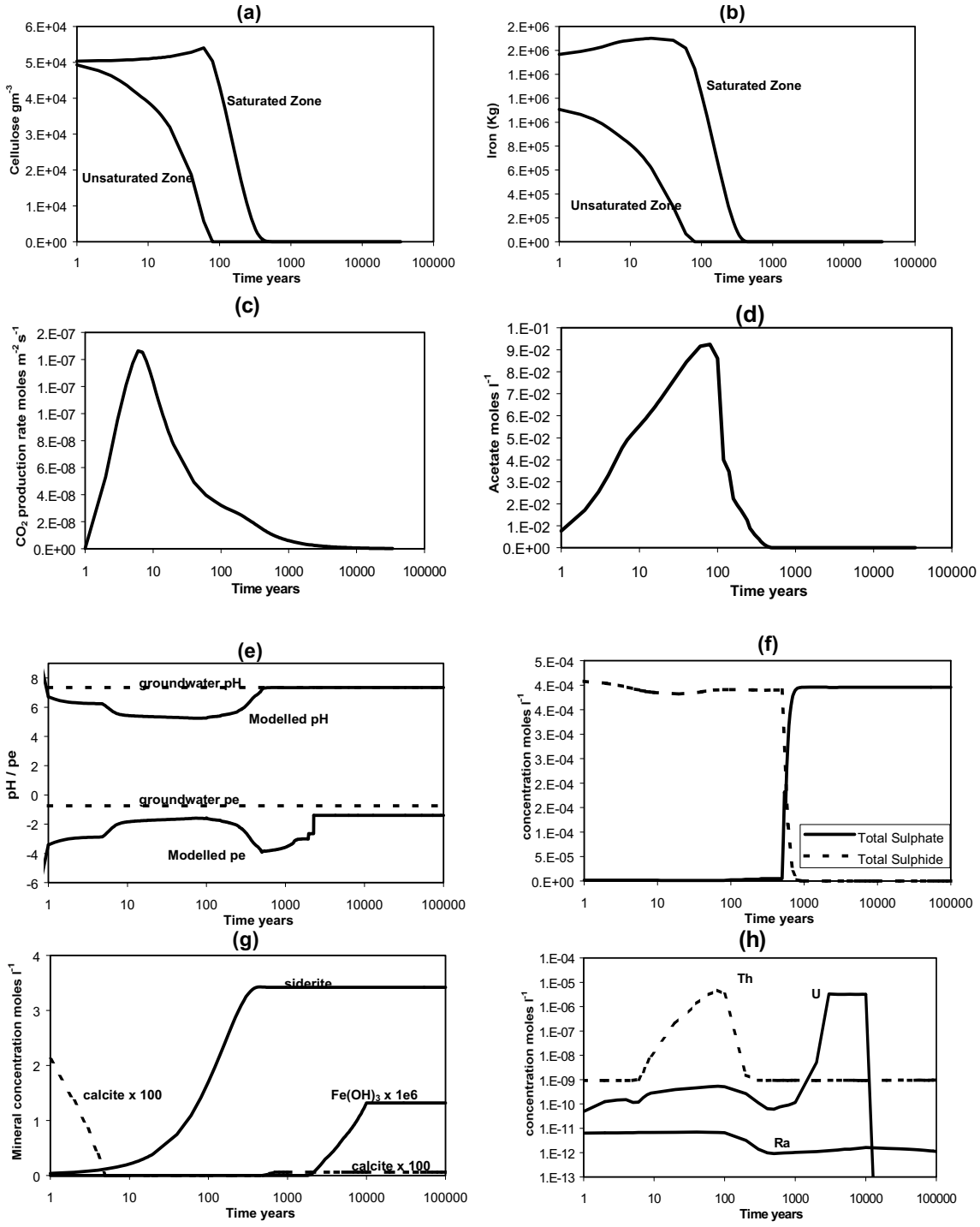

Figure 3. **(a)-(g)** Concentration profiles of waste materials and degradation products simulated by the DRINK model for a representative model cell from the Drigg trenches, **(h)** concentration profiles of solubility controlled radionuclides.

induced cellulose degradation reactions [3]. The products of these processes are used to determine an evolving redox condition, taking account of kinetically controlled microbially mediated redox reactions. Redox potential (pe) is calculated by using standard mass action equations [12] considering the most oxidizing couple. The resulting pe is used as a constraint for equilibrium speciation and mineral equilibrium calculations by a routine based on PHREEQE [13], which determines the pH and master species concentrations, including those radionuclides which are solubility controlled. The microbiological processes included in GRM [3] are those outlined in Table II. Example output from a recent DRINK run [14] can be found in Figure 3 which shows changes in waste materials, degradation products and solubility controlled radionuclides.

Impacts from the gaseous exposure pathway are calculated using the DEGAS (Drigg Evaluation of GAS) code [2]. DEGAS determines the dose and risks associated with the evolution and migration of radioactive gases from the Drigg site. It incorporates a simple microbial gas generation model to account for the potential generation of ^{14}C labeled methane and carbon dioxide, and tritiated methane. It assumes direct metabolism of glucose resulting from cellulose hydrolysis, to methane and carbon dioxide under anaerobic conditions and carbon dioxide under aerobic conditions. The proportion of ^{14}C and tritium in the organic portion of the waste is user defined. DEGAS also models the potential generation of methylated forms of ^{79}Se, ^{126}Sn and ^{129}I. It should be stressed that the DEGAS model, as with many assessment models, is highly conservative. There is no evidence of the generation of methylated gases at Drigg and site data indicates that emissions of ^{14}C and tritiated methane and ^{14}C carbon dioxide represent negligible risk.

CLOSING COMMENTS

The presence of microbially degradable material, principally cellulose, in the Drigg waste stream means that microbial activity and associated gas generation are a natural consequence of disposal operations at the Drigg site. Consequently an understanding of microbiology is key to understanding the behavior and evolution of the Drigg site. Through site observation and experimental studies it has been possible to integrate microbiology into the Drigg PCRSA. Microbiology is a key component of the FEPs analysis, engineering designs and source term modelling. Other microbial processes such as the role of biofilms in the retardation of radionuclide are areas of ongoing research which will be integrated into the Drigg PCRSA once these investigations are mature.

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