

Fate of Gases generated from Nuclear Wastes

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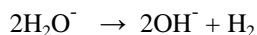
1. Introduction

Low- and intermediate- level radioactive wastes (L/ILW) generated from nuclear power plants, industries, hospitals, and research institutions contain a variety of radionuclides and a large fraction of biodegradable organic materials such as contaminated paper, rubber gloves, shoes, cotton, spent resins, cartridge filters, etc. These wastes are packaged in steel containers and disposed of in the subsurface facility. The backfill materials such as cement, bentonite or crushed rock are used as engineered barriers against groundwater infiltration and radionuclide transport. Gas generation from radioactive wastes is attributed to radiolysis, corrosion of metals, and degradation of organic materials.

Corrosion of steel drums and biodegradation of organic materials in L/ILW can generate gas which causes pressure build up and has the potential to compromise the integrity of waste containers and release the radionuclides and other contaminants into the environment. Performance assessment therefore requires a detailed understanding of the source and fate of gas generation and transport within the disposal system. Here we review the sources and fate of various type of gases generated from nuclear wastes and repositories.

2. Radiolysis

Radiolysis of water in the waste and in the repository will consume water and primarily produce hydrogen.

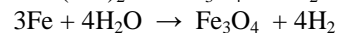


A variety of other gases can also be produced by the alpha radiolysis of cellulose, rubber, and plastic in the waste but it is considered to be minimal from the standpoint of long-term gas production in the repository [1]. However, radiolysis was identified as a significant source of hydrogen generation from bituminised wastes.

3. Corrosion

The corrosion of metals in the repository may produce significant amount of gas. The amount of gas production is dependent on two major factors: (i) the amount of water present in the waste at the time of emplacement and/or the amount of water that enters the repository after closure and (ii) the quantity of

corrodible metals present in the waste. There are two corrosion mechanisms that can occur in the disposal facility: oxic corrosion and anoxic corrosion. Oxic corrosion of the corrodible metals in the waste inventory will consume the oxygen. After the depletion of oxygen initially present in the repository, anoxic corrosion of these materials will dominate. This will produce hydrogen and consume water at rates that will depend on the amount of water present [2]. It is generally accepted that the largest gas generation source will be the anaerobic corrosion of the iron contained in the steel canister.



Microbiologically influenced corrosion (MIC) are associated with localized corrosion underneath a biofilm. Microorganisms affect the physical chemistry of the water-metal interface, and in turn accelerate corrosion rates on many metal surfaces. Biocorrosion can occur due to the metal binding by bacteria or through the creation of anodic and cathodic micro environments within the biofilms. Although the metal waste form (MWF) alloy is resistant to corrosion and selective leaching in geological environment, the microbes may affect the stability of MWF alloy.

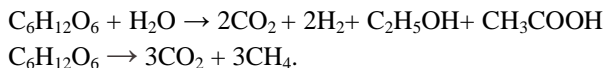
4. Microbial degradation

Microorganisms have been detected in L/ILW sites, deep geological formations, and waste repository sites designated for disposal of high level waste. Indigenous microorganisms and those present in L/ILW can metabolize organic compounds from L/ILW as an energy source aerobically and under anaerobic conditions use nitrate and sulfate as an electron acceptors. Such bacteria can produce H_2 , N_2 , H_2S , CO_2 , and CH_4 gas as a result of anaerobic respiration. Microbial activity is limited by the availability of electron donors and acceptors, moisture, and nutrients (specifically nitrogen); and affected by temperature, pH, Eh, radiation and toxic compounds.

The principal source of microbiological gas production in L/ILW is expected to be the degradation of cellulosic materials such as paper, wood, and tissues. Microbial degradation can take place under both aerobic and anaerobic conditions. The general reaction of aerobic cellulose degradation is as follows:



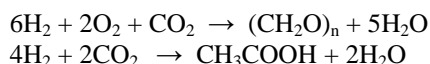
After the depletion of oxygen in the repository, anaerobic microbiological degradation will begin as shown in the following reactions.



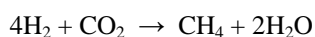
The end products of an anaerobic degradation of organic materials are CO_2 , H_2 , CH_4 , H_2S , N_2O , N_2 , and volatile low molecular weight organic compounds. In addition biomethylation of selenium and iodine to volatile dimethylselenide and dimethyldiselenide and methyl iodide can be generated from the waste.

5. Fate of gases generated from the wastes

Hydrogen is a common microbial metabolic product, which is utilized as an energy source by a variety of bacteria [3]. Oxidation of hydrogen to the reduction of compounds such as oxygen, nitrate, Fe(III), sulfate or carbon dioxide is catalyzed by bacteria. Hydrogen oxidizing bacteria are facultative autotrophs, able to grow with hydrogen as electron donor and with carbon dioxide as the sole carbon source under aerobic or anaerobic environments.



Methanogenic bacteria oxidize hydrogen with the reduction of carbon dioxide to produce methane.



Methanogenic bacteria when incubated with H_2 and CO_2 produced elevated levels of methane gas (Fig. 1).

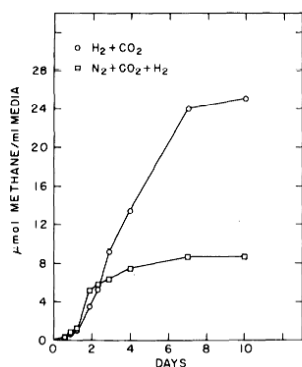
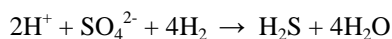


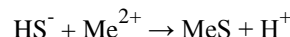
Figure 1. Production of methane by mixed methanogenic culture from synthetic media incubated under 80% H_2 + 20% CO_2 and 85% N_2 + 10% CO_2 + 5% H_2 gas phase.

Sulfate reducing bacteria and other hydrogen-oxidizing bacteria were able to

utilize cathodic hydrogen from mild steel as the only source of energy for growth with sulfate or nitrate as terminal electron acceptor.



Hydrogen sulfide gas generated from sulfate reduction will react with iron and other metals in the waste forming metal sulfides.



Iron and other metal reducing bacteria are also use hydrogen as electron donor.



Several groups of nitrogen fixing bacteria utilize hydrogen and carbon dioxide and reduce atmospheric N_2 to NH_3 .

These studies show that gases generated from the waste repositories are transformed to various organic and inorganic end compounds.

6. Conclusions

Studies on modeling of the fate and transport of repository gases primarily deal with hydrogen and CO_2 . Although hydrogen and carbon dioxide are the major gases of concern, microbial transformations of these gases in the subterranean environments could be significant. Metabolism of hydrogen along with the carbon dioxide results in the formation of methane, low molecular weight organic compounds and cell biomass and thus could affect the total inventory in a repository environment. Modeling studies should take into consideration of both the gas generation and consumption processes over the long-term.

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