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## Microsparging MTBE-contaminated plumes

Microsparging uses air stripping and oxidative decomposition to treat water contaminated with halogenated and non-halogenated VOCs. This process, which can be catalytically accelerated, injects air and ozone into groundwater through spargers. The resulting microbubbles have a very high surface-area-to-volume ratio. "As these microbubbles rise within a column of water, they extract or strip VOCs from groundwater by partitioning from aqueous to the gas phase. Upon entering the microbubbles, the VOCs are rapidly oxidized by Criegee oxidation. The ozone contained within the bubbles reacts to decompose, for instance, a chlorinated ethane molecule in an extremely rapid gas/liquid phase reaction whose end products are carbon dioxide, very dilute hydrochloric acid and water. By increasing the ozone content within the bubbles, the rate of oxidation is increased" (Kerfoot, 2000). MTBE will decompose into alcohols, acetate and formate. Residual oxygen accelerates bioremediation of these breakdown products, converting them to carbon dioxide and water (Kerfoot and McGrath, 2001).

### Bubble formation

According to Kerfoot (2000), "Gas entering a small bubble of volume  $4\pi r^3$  increases until reaching an asymptotic value of saturation." Consequent bubble formation will depend on the following factors:

- the time-varying concentration of the substance in the bubble
- the rate of change of vapor concentration within the gaseous phase
- the external concentration of the VOC
- the absorption constant, set by Henry's partitioning coefficient

### Laboratory testing

Kerfoot studied the reaction of microbubble ozone on MTBE in dilute aqueous solutions. Bench-scale testing involved the following:

- ozone bubbled under pressure into 500 ml Erlenmeyer flasks
- ozone created by a corona-type ozone generator fed by oxygen
- a fluidized soil mixture, simulated by adding 10 grams of fine glass beads (.3 mm) to the flask
- pH maintained in the range of 5.0 to 6.0
- varied ozone concentration, pressure and iron silicate content

After ozone addition, flasks were covered, stored for 60 minutes at room temperature, then analyzed in the gas phase by Gastec tubes, and in the liquid phase by the indigo trisulfonate method. Researchers used a Hewlett Packard gas chromatograph and mass spectrometer to identify MTBE and ozonation products. The researchers also used a laboratory gas chromatograph for quick inspection of MTBE removal during testing.

Kerfoot (2000) found that

- MTBE was rapidly degraded by microbubble ozone injection
- microbubble injection appeared effective in reducing MTBE concentrations to beyond 90 percent of original level
- removal rate was sensitive to ozone concentration, pressure and iron silicate content

### Field testing

Kerfoot tested microsparging at two sites:

- a gasoline spill at an automotive service station
- at the leading edge of a solely MTBE plume upgradient of a water supply well near Lake Tahoe, California

### Automotive service station

At the automotive service station, workers installed a C-Sparge™ unit that

- came equipped with an oxygen generator
- permitted adjustment of ozone concentrations between 100 and 300 ppmv
- had a radius of influence estimated by dissolved oxygen content concentration and redox potential

The site's characteristics are as follows:

- static groundwater at depths of 3 to 6 ft (1 to 1.8 m) below grade
- soil permeability ranging from  $10^{-3}$  to  $10^{-7}$  cm/sec
- groundwater pH of 7.0 to 8.5
- low groundwater flow with hydraulic gradient of 0.0036 ft/ft
- specific conductance range of 850 to 1,100  $\mu$ S
- total BTEX ranging from 5,000 to 24,000 ppb in groundwater
- vertical extent of contamination extending to a maximum depth of 15 ft
- absence of light non-aqueous-phase liquid (NAPL)
- less than 1 ppm dissolved oxygen content in nearby groundwater monitoring wells

The system installed

- had a combined air/ozone flow of 2.2 cfm to the recirculation well
- was set to run for 18 minutes for the low Spargepoint<sup>®</sup>, 11 minutes for the in-well Spargepoint<sup>®</sup>, and 5 minutes for the submersible pump
- operated on 14 cycles of 34 minutes during each 24-hour period

During the 20-day test period, researchers monitored total BTEX and MTBE from a well located 18 feet from the main circulation well. Total BTEX plus MTBE concentrations dropped from 19,220 ppb to 1,004 ppb, a reduction of 94.8 percent. MTBE concentrations dropped from 520 ppb to 6 ppb during the test period, or in excess of 99 percent removal.

### Leading edge of MTBE plume, Lake Tahoe, California

At the leading edge of an MTBE plume near Lake Tahoe, remediators used an oxidative curtain or "bubble fence" to protect a water supply well. Site characteristics are as follows:

- silty, clayey sand
- static groundwater level, 5 ft below grade, although levels have been known to drop 10 ft
- groundwater gradient, 1.125 ft/ft
- observed forward plume advance, 3 to 10 ft/month
- bottom of contaminated zone, 20 ft below grade
- upgradient breadth of plume estimated to be about 50 ft

Workers installed an oxidative curtain at right angles to the plume. The bubble fence consisted of

- 4 dual-level Spargepoints<sup>®</sup> fed by a Model 3600 C-Sparge<sup>™</sup> unit
- Spargepoints<sup>®</sup> installed about 13 ft on center in a line about 40 ft long, 100 ft upgradient of the water supply well
- Spargepoints<sup>®</sup> installed at 21 and 24 ft below grade, and another at 36 and 39 ft below grade

In addition, workers installed monitoring wells upgradient and downgradient of the oxidative curtain. Workers found a level of 6 ppb upgradient from well, but downgradient MTBE levels dropped to non-detect.

### Conclusions

Kerfoot (2000) concluded that

- MTBE can be rapidly removed using fine bubbles that strip contaminants from groundwater
- ozone can effectively decompose contaminants
- in field studies, MTBE and toluene had field attenuation coefficients of five days or less, resulting in reduction to one-half concentration
- benzene, ethylbenzene and xylenes showed half-lives of seven to nine days
- adsorbed soil fractions of MTBE are less than those of benzene fractions
- MTBE removal occurs primarily from the aqueous fraction

### References

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Kerfoot, W.B. and A. McGrath, "Microbubble Oxidation Smokes MTBE and BTEX," *Contaminated Soil Sediment and Water*, Spring 2001; <http://www.aehsmag.com>.

UTTU thanks William Kerfoot, President of KV Associates, Inc. ([bill@kva-equipment.com](mailto:bill@kva-equipment.com)), for his help on this article.



## Natural attenuation of toluene in a shallow aquifer

Lee and others (2001) examined a shallow, unconfined aquifer contaminated with hydrocarbons. Their study

- assessed reactive product concentration, geochemical parameters and seasonal changes in the vertical distribution of toluene
- evaluated the mechanism and capacity for natural attenuation of toluene in the vadose and groundwater zones

### Study site description and characterization

The study site, 45 km southwest of Seoul, Korea, has heavily petroleum-contaminated soil and groundwater as a result of leaking storage tanks. The site had been contaminated for at least 10 years, with the plume extending more than 500 m downgradient.

Field workers collected soil samples using a soil probe to obtain lithological and contaminant characterization data. At some locations where soil samples were taken, workers installed monitoring wells. "Soil core samples were collected from a depth of 1 m below the surface down to a depth of 6 m at 50 cm intervals. Three clusters of monitoring wells were installed at 20 m, 30 m and 100 m, downgradient from the contamination source... In each of the clusters, monitoring wells with a screen length of 30 cm were installed to intersect the following sampling depths: 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5, 5 and 6 m. In addition, soil gas monitoring ports linked to a sampling stopcock were installed in monitoring wells, enabling each piezometer to sample either groundwater or soil gas depending on screen depth and groundwater level. A fully screened monitoring well was installed upgradient of the contamination source area to measure the background geochemistry of uncontaminated groundwater" (Lee and others, 2001).

Researchers measured the following:

- grain size analyses, performed according to the standard ASTM method
- saturated hydraulic conductivity and air permeability, as a function of average pore radius (total porosity) using the Hazen and Massmann method
- organic carbon content (0.18 percent), using the potassium dichromate method

They also analyzed the following:

- toluene, using U.S. EPA standard methods and a gas chromatograph (HP 5890) equipped with a flame ionization detector
- O<sub>2</sub>, CO<sub>2</sub>, CH<sub>4</sub> levels of respiration gas
- water levels
- dissolved oxygen concentrations and redox potentials, measured with portable equipment
- ferrous iron (Fe<sup>2+</sup>) concentrations using a portable test kit
- nitrate (NO<sub>3</sub><sup>-</sup>) and sulfate (SO<sub>4</sub><sup>2-</sup>) concentrations, analyzed in the laboratory by ion chromatography

The subsurface ranged from poorly sorted silty sands to sandy gravels. Researchers noted a highly conductive zone at 2.4 to 3.6 m. According to Lee and others (2001), "Most of the free-phase contaminants would flow through this highly conductive zone. This relatively uniform and more conductive zone makes this site amenable to in-situ aeration remediation technologies because of the high air permeability."

### Aquifer level changes and groundwater velocity

Researchers found that changes in aquifer levels exceeded 2 m and correlated strongly with rainfall, which significantly affected hydrocarbon fate. "As the level of water table fluctuates in response to groundwater recharge/discharge events, pools of NAPL would be carried up and down with the water table. Some of the NAPL is trapped in the pore spaces below and above the water table, and a smear zone is thus formed in the region of groundwater table fluctuations, where some of the NAPL remains trapped in the form of residual NAPL. This was why contaminant was detected in the unsaturated soils outside the source area.

"In soil or aquifer materials contaminated with NAPL, contaminants from the NAPL will partition onto solid phase materials. Desorption of contaminants is often a rate-limiting step and contaminants will thus continue to leach into the groundwater, forming a secondary source of the contamination. This phenomenon is partially responsible for the tailing effect commonly observed in remediation systems" (Lee and others, 2001).

Because the average organic carbon content was estimated to be at 0.18 percent, the toluene retardation factor (due to adsorption of organic matter) was estimated to be 2.3. Assuming an average hydraulic gradient of 0.006 and effective porosity of 0.3, researchers estimated a groundwater velocity of 0.81 m/day and a toluene velocity of 0.35 m/day.

## Contaminant findings

Maximum soil contamination was 2,653 mg/kg BTEX compounds detected 1 m above and 1 m below the mean groundwater level, a zone within the range of groundwater fluctuation. Soil gas concentrations of toluene ranged from 300 to 1,500 ppm by volume, or about 5 percent of the theoretical concentration calculated from liquid-gas-phase partitioning equilibrium based on Henry's law. Researchers attributed these low concentrations to natural attenuation of toluene in unsaturated soil. The highest levels were detected about 1 m below ground surface. Toluene concentration in soil gas in winter was higher than in the summer.

In groundwater, the highest zone of contamination was from 1 to 1.5 m below the water table. "In summer, when most recharge occurs and the water table typically rises 1-2 m, a smear zone is formed. Subsequently, even if the groundwater concentration becomes diluted to 165-310 mg/L, the contaminant plume depth would increase to approximately 2 m. During the dry season (winter), a high concentration range of 270-480 mg/L was detected; notably, free-phase hydrocarbon in samples at a site was visible. In general, contaminant concentrations trend upward as depth of contamination increases. However, the toluene concentration in the area was relatively low due to significant dilution caused by a rapid rise in the water table during recharge events and relatively slow desorption and transport of toluene into the newly recharged water" (Lee and others, 2001).

## Soil gas concentrations

Soil gas concentrations in the uncontaminated reference area were as follows:

- O<sub>2</sub>, 20.9 percent
- CO<sub>2</sub>, below 0.1 percent
- CH<sub>4</sub>, not detected

Researchers attributed slightly lower oxygen concentrations to natural organic matter decomposition. The slightly elevated CO<sub>2</sub> suggested that hydrocarbons were biodegrading. With depth, CO<sub>2</sub> increased and O<sub>2</sub> was depleted, again due to increased natural biodegradation. At 1.5 m below the surface, a CH<sub>4</sub> concentration of 5 percent was detected; CH<sub>4</sub> concentrations increased to 15 percent in summer, where researchers concluded that an anaerobic state prevailed. When this happened O<sub>2</sub> levels were almost zero.

Also in summer, large increases in CO<sub>2</sub> (to approximately 25 percent) might result from sufficient moisture in the wet season and a decrease in oxygen diffusion into the soil, because pore spaces are water-filled. Low CO<sub>2</sub> in winter suggests that populations of indigenous microorganisms are sufficient, but O<sub>2</sub> concentration limits their activity.

## Soil gas and toluene concentrations

"The decrease in toluene concentration above the water table coincided with a decrease in CO<sub>2</sub> concentrations and an increase in O<sub>2</sub> concentration. The reason for the inverse correlation between O<sub>2</sub> and toluene was that O<sub>2</sub> is depleted by aerobic biodegradation in regions of high toluene concentration. When the concentration of toluene was high, the concentration of O<sub>2</sub> was close to zero, and concentrations of CH<sub>4</sub> and CO<sub>2</sub> increased. This implies toluene decomposition: during the process O<sub>2</sub> was depleted, while CO<sub>2</sub> and CH<sub>4</sub> were generated. This was especially true in low toluene concentration areas. In summer, as the concentration of toluene increased, the concentrations of CO<sub>2</sub> and CH<sub>4</sub> also increased. With higher toluene concentrations, CO<sub>2</sub> and CH<sub>4</sub> concentrations were stable. This indicates a more efficient decomposition of toluene under conditions of low toluene loading and favorable conditions of temperature and moisture in summer rather than in winter" (Lee and others, 2001).

## Conclusions

Lee and others (2001) concluded the following:

- at this site, seasonal water fluctuations exceeded 2 m, a very large magnitude in relation to the average saturation thickness of 3 m of the shallow aquifer; as the water table fluctuated following groundwater recharge/discharge events, a smear zone formed
- biodegradation rates measured during an in-situ respiration test were as high as 5.2 mg/kg/day in summer, and 2.9 mg/kg/day in winter
- toluene degradation occurred through the following microbe-mediated processes (percent contribution to total assimilative capacity is given in parentheses):
  - aerobic respiration by dissolved O<sub>2</sub> (8.9 percent)
  - nitrate reduction (3.2 percent)
  - iron reduction (9.3 percent)
  - sulfate reduction (33.4 percent)
  - methanogenesis (45.2 percent)
- 90 percent of toluene biodegradation in this aquifer occurred by nonaerobic processes

- dissolved toluene at this site ranged from 150 to 480 mg/L, based on theoretical field calculations and data; groundwater had insufficient capacity to intrinsically biodegrade dissolved toluene within a time frame of years
- toluene attenuation rate here was 0.0048 to 0.018/day, giving toluene an estimated first-order biodegradation of 0.0022-0.0094/day; attenuation rates and biodegradation rates were a little higher in summer than in winter, and as depth below the water table increased
- the contribution of biodegradation to overall attenuation was estimated to range from 45.8 to 52.2 percent
- future studies should investigate the effects of changing moisture contents, heterogeneous distribution and diffusive flux of oxygen

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Lee, C.H., Lee, J.Y., Cheon, J.Y. and K.K. Lee, "Attenuation of Petroleum Hydrocarbons in Smear Zones: A Case Study," *Journal of Environmental Engineering*, July 2001; <http://ojs.aip.org/eeo>

See also "Factors Affecting the Distribution of Hydrocarbon Contaminants and Hydrogeochemical Parameters in a Shallow Sand Aquifer," Lee, J.Y., Cheon, J.Y., Lee, K.K., Lee, S.Y. and M.H. Lee, *Journal of Contaminant Hydrology*, Vol. 50; <http://www.elsevier.com/locate/jconhyd>.



## MTBE and TBA anaerobic degradation

The details of anaerobic MTBE (methyl tert-butyl ether) and TBA (tert-butyl alcohol) degradation remain largely a mystery; however, evidence indicates that the potential for anaerobic degradation of MTBE exists. This evidence deserves consideration since most petroleum/MTBE release sites are anaerobic or quickly become anaerobic as oxygen is depleted. Adding additional oxygen to these zones—one method of remediation—can be technically difficult and expensive (Finneran and Lovley, 2001).

### Anaerobic plumes and terminal electron-accepting process

The electron acceptors most prevalent in anaerobic aquifers are

- nitrate
- Fe(III)
- sulfate

BTEX/MTBE oxidation is thermodynamically feasible with the electron acceptors listed above, yet distinct zones develop in a plume. The distribution of zones derives from competition among the microorganisms for the electron donors.

"Nitrate yields the most energy of the anaerobic electron acceptors, and the denitrifying organisms can metabolize the electron donors at concentrations that are too low to sustain the Fe(III) reducers. When nitrate is depleted, the next most favorable acceptor is Fe(III). The Fe(III)-reducing microbes metabolize electron donors at concentrations too low to sustain the sulfate-reducers. In turn, sulfate reducers can out-compete the methanogenic microorganisms" (Finneran and others, 2001). Methanogenesis dominates closest to the contaminant source; because it is the least thermodynamically favorable, it dominates only after nitrate, Fe(III) and sulfate are depleted.

These TEAP (terminal electron-accepting process) zones can be identified by measuring dissolved hydrogen (a key electron donor) in existing monitoring wells.

Remediators typically add oxygen to stimulate microbes, which degrade contaminants. Adding oxygen is problematic for the following reasons:

- oxygen is not very soluble, and adding it to groundwater is inefficient in source zones
- oxygen that enters the source area can be depleted by chemically reacting with reduced compounds, such as Fe(II)
- newly formed Fe(III) can plug oxygen injection or monitoring wells

In contrast, anaerobic electron acceptors are more soluble and less difficult to add to the subsurface.

### Significance of Fe(III)

According to Finneran and Lovley (2001), "Fe(III) is frequently the most abundant potential electron acceptor for the anaerobic oxidation of organic contaminants in polluted subsurface environments."

Studies have shown that benzene degrades anaerobically with the reduction of Fe(III) facilitated by Geobacteraceae bacteria. To accelerate Fe(III) reduction, researchers can add humic acid substances (HS), which result from the breakdown of complex organic matter. "Fe(III)-reducing bacteria can directly reduce HS, and Fe(III), which is oxidized, and can abiotically accept electrons from reduced HS. The HS become re-oxidized and are again free to accept electrons in microbial metabolism. As such the HS act as an electron shuttle between the microorganism and the Fe(III). This is significant because the prevalent form of iron in subsurface environments is Fe(III) oxides. It is

generally accepted that bacterial cells must physically contact the insoluble Fe(III) oxides to reduce them; however, because they are not freely soluble, the Fe(III) oxides are occluded from rapid reduction. Soluble HS stimulate Fe(III) reduction because of this electron shuttling phenomenon, and increased Fe(III) reduction hastens the degradation of organic electron donors" (Finneran and others, 2001).

Researchers had also tested synthetic electron shuttles, such as anthraquinone-2,6-disulfonate (AQDS). Laboratory incubations have shown that AQDS simulated Fe(III) reduction in a fashion similar to that of HS. "Because of its catalytic nature, very little AQDS (or HS) is needed to accelerate Fe(III) reduction. It is constantly recycled—alternating between variant oxidized and reduced states" (Finneran and others, 2001).

The first demonstration of Fe(III) reduction and possible anaerobic degradation of MTBE occurred in laboratory incubations using BTEX- and MTBE-contaminated sediments and Fe(III) from a contaminated plume. [<sup>14</sup>C]-MTBE was added, and after seven months, three percent of the [<sup>14</sup>C]-MTBE was mineralized to <sup>14</sup>CO<sub>2</sub>. Researchers attributed this loss to Fe(III) reduction as oxygen and nitrate were depleted; Mn(IV) and sulfate did not exist in the samples. Other studies have shown that addition of HS, as an electron shuttle, accelerated MTBE degradation. Added chelators did not stimulate MTBE degradation. Each time, Fe(III) was added, however, MTBE degraded rapidly and repeatedly. HS addition was not required as it was recycled.

Another demonstration using freshwater sediments from the Potomac River showed that [<sup>14</sup>C]-MTBE added to sediments produced <sup>14</sup>CO<sub>2</sub> and <sup>14</sup>CH<sub>4</sub> in ratios of 3:1, suggesting the existence of several different microbial populations responsible for MTBE degradation. <sup>14</sup>CO<sub>2</sub> alone was produced when only Fe(III) or Fe(III) plus electron shuttling compounds were added.

### Sulfate reduction

Limited studies of MTBE degradation by sulfate reduction suggest the potential for anaerobic MTBE oxidation. Sulfate would be an ideal electron acceptor because sulfate

- has a greater electron accepting capacity than oxygen
- is freely soluble
- does not react with any reduced compounds in the source zone

Furthermore, sulfate has been found to be effective at stimulating benzene degradation.

### Nitrate reduction and denitrification

Thus far, direct evidence confirming anaerobic MTBE degradation under nitrate reduction and denitrifying conditions does not exist.

### Anaerobic TBA (tert-butyl alcohol)

The results of several studies suggest that under anaerobic conditions, MTBE is degraded to TBA, but no further degradation occurs. Some researchers (Yeh and Novak in Finneran and others, 2001) found TBA degraded in soil under denitrifying conditions, but these conditions may not have been strictly anaerobic. "Studies with aquatic sediment (Potomac River) found that [<sup>14</sup>C]-TBA was readily mineralized to both <sup>14</sup>CO<sub>2</sub> and <sup>14</sup>CH<sub>4</sub> without any lag, with as much as 30 percent of the [<sup>14</sup>C]-TBA recovered as <sup>14</sup>CO<sub>2</sub> and 9 percent as <sup>14</sup>CH<sub>4</sub>. When 900 μmol/kg of unlabelled TBA was added to sediments, it was degraded in the sediment in fewer than 50 days. The rate of TBA consumption was much greater than the rate of MTBE degradation, suggesting that TBA is unlikely to accumulate under anaerobic conditions" (Finneran and others, 2001).

### Future for anaerobic degradation of MTBE and TBA

Anaerobic strategies offer several benefits. They are

- relatively inexpensive
- easy to implement; the electron acceptors can be easily delivered to the subsurface via injection wells
- monitored infrequently

### References

Finneran, K.T. and D.R. Lovley, "Anaerobic Degradation of Methyl tert-Butyl Ether (MTBE) and tert-Butyl Alcohol (TBA)," *Environmental Science & Technology*, Vol. 35, No. 9, 2001; <http://www.pubs.acs.org/>.

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UTTU thanks Derek Lovley, [dlovley@microbio.umass.edu](mailto:dlovley@microbio.umass.edu), for his help on this article.



## Corrosion and Corrosion Control

By N. Dennis Burke and David H. Kroon

Corrosion is a natural phenomenon that follows the laws of science. An understanding of the basic principles aids in the development of preventive measures for controlling corrosion.

### The process of corrosion

The manufacture of metal products is a prime example of how humans have changed the balance of nature. Most metals are not found in nature as pure materials; rather, they are combined with other elements such as oxygen, forming compounds called ores. To obtain metals from ores, the compounds are subjected to extreme heat to break the chemical attractions between the elements. Once the pure metal is isolated, it undergoes rolling, bending and other shaping operations to obtain the desired finished product. All of the refining and production steps for the transformation of an ore to a final product have one thing in common: they add ENERGY to the metal. Energy is added to force the metal to break free of the ore, and more energy is added to shape the end product. Thus, an imbalance of energy exists. Nature does not ignore this imbalance and fights back. As soon as the addition of energy ceases, the metals begin to reform their ores.

A common example of this involves the manufacture of steel from iron oxides. The ore is smelted and refined to make steel ingots, which are folded into sheets of billets for the making of various items such as piping, tanks and pilings. Common experience shows that products made from iron ore do not remain in the condition intended. The steel begins to revert to iron oxides (rust) immediately after it is formed.

To help understand why the reforming of natural ores occurs, consider the process of rolling a round stone up a hill. The higher up the side of the hill the stone is pushed, the greater the energy expended to get to that height. Consider the hill as an "energy hill" with the heights representing different energy levels. Once the addition of energy to the stone ceases, the stone will roll down the energy hill and lose the energy that had been expended to place it in the elevated position.

The same downhill process applies to steel. The metal "rolls" down the energy hill to return to its natural energy level. The process of metal releasing energy to reform its natural ores is called corrosion. Corrosion is a normal

occurrence common to all metals not in their natural states. As steel corrodes in either soil or water, electrical energy is released. When metals form ore in these environments, an electric current is generated. Corrosion has two primary aspects:

- a physical change in the metal occurs
- a direct electric current is generated

A corroding metal releases electrical energy, but nature requires that this energy be balanced. To achieve balance, some other metal receives the energy. By receiving energy, the second metal can maintain its level on the energy hill and remain stable. For the electric current to flow, a metallic connection between the two metals must exist. The two metals involved in the corrosion process are electrodes. More specifically, the electrode releasing electrical energy to the environment is an anode, and the one receiving the electrical energy is a cathode.

One other basic requirement is that the metal must be placed in an electrolyte such as soil or water. These are solutions that surround the electrodes and which contain the necessary elements to react with the metal to form an ore. The solution also conducts the electric current flow between the anode and cathode, serving as an electrolyte. The anode releases electrical energy and metal ions into the electrolyte. The metal ions combine with the chemicals adjacent to the anode to form ores, and the electric current flows through a conductor to the cathode. At the cathode, the current causes other ions in solution to react and form films on the surface of the cathode. The balance of energy is maintained. The anode releases energy and loses part of its physical form while the cathode gains an equal amount of energy, and a physical change occurs.

For corrosion to occur, four items must be present:

- two metals at different energy levels (i.e., the anode and cathode)
- an electrolyte
- a conductor

This combination is called a galvanic cell. An example of this is a flashlight battery. The corrosion process involving the zinc case, carbon center post, electrolyte and conductor produces the electric energy required to light a bulb or ring a bell. All of the conditions for a galvanic cell are present.

In a galvanic cell, which is also called a corrosion cell, electrical energy is present. The energy difference between the anode and cathode provides the force to make the current flow. When analyzing a corrosion cell, it is necessary to determine which of the metals is the anode and

which is the cathode. If the current flow in the electrolyte or the conductor were monitored, the identification would be simple, but in many situations this is impractical. Another means of distinguishing anodes from cathodes must be used.

Consider again the energy hill: every metal contains a certain amount of energy and has its own position on the energy hill. The metal's position reflects its energy potential. The more potential that a metal has, the higher up the energy hill it is found. By measuring the energy or potential of a metal, its position on the energy hill relative to other metals can be determined.

In electrical terms, the force that pushes the electric current is voltage. Under similar conditions, the greater the voltage, the greater the current flow. Often a reference electrode is used to measure voltage, and the metals are listed according to this reference. In the corrosion control industry, the copper-copper sulfate electrode is used as the common reference for soils and fresh water and the silver-silver chloride electrode is used for saltwater applications.

Once the positions of the metals on the energy hill are known, the anode and cathode of a galvanic cell can be determined. Any metal on the energy hill will release energy to any one below it and conversely receive energy from any metal above it. Scientists have measured the potential of many different metals and have compiled a list, the Galvanic Series of Metals, which places these metals on the energy hill. Starting with the highest, the listing is:

- magnesium
- zinc
- aluminum
- iron
- lead
- copper
- nickel
- silver
- carbon
- platinum
- gold

Consider a piece of zinc and one of copper connected with a wire and buried in the earth. From this listing, the zinc is the anode because it will release electrical energy to copper, and copper is the cathode because it will receive the energy.

Because nature tends toward a state of equilibrium, a chemical reaction at the anode requires that a chemical reaction occur at the cathode. The cathode reaction is dependent upon electrolyte chemistry. The formation of

hydrogen is the simplest of the possible reactions. Hydrogen ions are naturally present in water. As the metal releases energy into the electrolyte, the electrolyte around the cathode absorbs an equal amount of electrical energy. The hydrogen ions in the water absorb the energy and form hydrogen atoms. The phenomenon caused by reactions at the cathode is commonly referred to as polarization.

The basic galvanic corrosion process can be summarized as follows:

- all metal formed from ores will corrode to achieve a balance of energy
- to maintain balance, a second metal must receive the released energy
- the corroding metal is called an anode, and the non-corroding metal, a cathode
- a suitable environment must exist for the energy to transfer; this environment consists of an electrolyte and a metallic connection between the anode and cathode

The electrical connection conductor between the anode and cathode (as indicated above) is a wire connection not submerged in the electrolyte. In actual practice, this may be any direct connection. Commonly, metals are joined by mechanical couplings, brazing, welding and physical contact.

The following are examples of galvanic cells resulting from two different metals buried or submerged together:

- steel tanks interconnected with copper piping or copper grounding systems
- brass valves inserted in steel piping
- rivets in any vessel that are of different alloys than the vessel
- tube sheets and tubes of different alloys in heat exchangers or surface condensers

Often corrosion occurs without an obvious anode and cathode. Why does a steel fuel tank buried alone develop corrosion pits and penetrations? Why is there corrosion of the copper strands of the copper concentric neutral spiraled along the buried power cable? The answers are the same: different energy levels exist. These differences are more subtle than previously discussed and are caused by differences within the metal or by differences within the electrolyte.

The corrosion of fuel tanks or copper neutrals is caused by galvanic cells. Areas of imbalance within the material or environment cause the energy level differences. For example, a steel tank is formed by bending a piece of steel to a specified shape and welding the sections to form

a containment vessel. The bending imparts work or energy into some portion of the plate.

The welding process heats the plate and changes the energy levels in the heated zone. Although made of one material, the tank has many separate levels of energy. These are ideal for generating galvanic cells.

The cable with copper neutrals traverses many types of soil. The chemical content of these soils varies and the energy levels of copper will vary accordingly. The differences in concentration result in energy imbalances and a galvanic cell.

Another example of a galvanic cell is evident on piling systems along the seacoast. Typically, the most severe corrosion rates are found just below the mean low-water mark and at the mud. In these two areas the oxygen concentration differs greatly from the remainder of the electrolyte. The surface wave movement at the mean low-water to high-water area increases the oxygen level. The zone immediately below has an energy imbalance due to the difference in oxygen concentration. At the mud line, an oxygen concentration differential exists and a galvanic cell forms. Difference in oxygen concentration can also occur at the pipe bottom, which is anodic to the pipe top in underground applications. Such oxygen differentials are frequently the cause of corrosion.

In all examples, the metallic connection between the anodes and cathodes is the structure itself. A separate connection is not necessary.

Galvanic cells are the most common causes of corrosion. A second general category of corrosion is caused by electrolytic cells. Electrolytic corrosion results when an external source of DC current is received on one portion of a metallic structure and discharged on another. Although not as prevalent, electrolytic corrosion often leads to rapid failures in concentration areas.

The operation of DC transit systems often causes electrolytic corrosion on neighboring structures. A portion of the DC traction power current leaves the rails of the transit system. This current flows through the earth to a structure where it is received onto the surface. The current flows along the structure to an area where it is discharged to return to the electrical ground of the DC traction power system. In the area of current discharge, concentration corrosion occurs. One ampere of current flowing from steel carries approximately 20 pounds of metal into solution in one year. Often, only a few grams of metal need to be lost in a concentrated area to penetrate the wall of a containment vessel, which can result in catastrophic corrosion failures.

When trolley systems existed, this type of corrosion occurred throughout metropolitan areas. Early recognition of the severe consequences of electrolytic corrosion prompted many people to refer to corrosion as electrolysis. Electrolytic action is, however, only one of the major causes of corrosion. In addition to transit systems, common sources of DC current that cause electrolytic corrosion include welding machines, DC motors, HVDC transmission and cathodic protection systems.

The four requirements for corrosion are also present in electrolytic cells:

- the cathode, the area where the current is received
- the anode, the area where the current is discharged
- the soil, the electrolyte
- the metal structure, the conductor between the anode and cathode

In electrolytic corrosion, the energy imbalance is created by an outside source of current flowing through the electrolyte.

In practice, the corrosion process usually involves more than one of many possible combinations of corrosion cells. Any analysis of corrosion must define each of the four components in the cell. Within those components, the possible variations are then identified. With all of the factors at hand, a systematic corrosion control solution can be outlined.

## Corrosion control

The most common methods of corrosion control involve

- material selection
- coatings
- electrical insulation
- cathodic protection

Each measure has distinct advantages and disadvantages; however, all should be considered when planning a comprehensive corrosion control program.

**Material selection** needs to be addressed during project design. Through the use of non-metallic components, corrosion can be avoided. Most often, however, non-metallic materials have operating limitations that necessitate the use of metals. Under these circumstances, similar metals should be incorporated in the design to avoid the interconnection of materials that are not close to each other on the energy hill. This serves to reduce the driving force between the anode and cathode in the corrosion cell and thereby decreases the rate of corrosion.

The proper selection of materials for the environment is very important. Many materials perform well in one environment but corrode very rapidly in another. Aluminum, for example, is extremely resistant to corrosion in the atmosphere because it forms a protective oxide layer that inhibits further corrosion. In salt water or soil environments, aluminum rapidly corrodes because the oxide layer that forms does not have the same protective properties. Stainless steel performs similarly. When stainless steel is placed in an oxidizing environment, a very thin oxide forms, which passivates the metal surface. If this film is damaged by such things as abrasion or the presence of chloride ions, the stainless steel will corrode at approximately the same rate as mild steel.

The careful selection of materials can reduce but cannot prevent corrosion. Differences in the electrolyte and differences along the surface of the metal will create an energy imbalance that results in corrosion. This cannot be entirely avoided.

**Coatings** are often used for corrosion control. A barrier coating insulates the anode and cathode of the corrosion cell from the environment. By insulating the metallic surface from the electrolyte, coatings prevent the corrosion reaction from occurring.

Many types of coatings exist, but whatever the material used, it should have the following characteristics:

- high dielectric strength to effectively insulate the structure from the electrolyte
- relative inertness with respect to the environment to ensure a long life
- high impact resistance to avoid damage during shipping, handling and installation
- good bonding characteristics to ensure permanent contact to the structure
- low water absorption properties to maintain the dielectric strength

After a coating is selected, great care must be exercised to ensure that the coating is properly applied. A coating failure usually occurs due to improper application and not to a breakdown of the coating material. Surface preparation is critical to coating performance.

Whatever the material selected and no matter how well the coating is applied, coatings are not perfect. Voids or holidays will exist. These may be large enough to be seen or may be tiny, microscopic pinholes that cannot be detected even with a flaw detector (jeep, holiday detector). Attempts to achieve perfect coatings have been made, but such attempts have been unsuccessful and

prohibitively expensive. Coatings will also deteriorate with time. As with all things, coatings suffer from both imperfection and aging.

The proper selection and application of a coating system will reduce the total metal loss but will not prevent corrosion. Unfortunately, coatings concentrate the corrosion activity at any imperfections. This intensifies the corrosion and results in penetrations more rapidly than if the structure were uncoated.

**Electrical insulation** is another method of corrosion control that can be used to break the conductor that connects the anode and cathode in the corrosion cell. Insulating fittings come in many forms, including unions, couplings, flanges and spools. These are most often used to electrically separate dissimilar metals. Common applications include the installation of an insulating fitting at a gas service meter to insulate the steel gas line from a copper water service that is electrically connected at the water heater. Insulating flanges are often used at well heads to break the electrical connection between bare steel well casings and coated steel flow lines. Pipelines are sometimes insulated at station locations to electrically isolate the steel pipeline from the copper grounding system. In all cases, the insulator is used to break the electrical continuity between the anode and the cathode.

Insulation serves only to break apart obvious corrosion cells. It will not prevent local corrosion activity resulting from such conditions as different oxygen concentrations. Insulated fittings must not be used indiscriminately, because, at times, electrical continuity is desired for other reasons including electrical grounding and cathodic protection.

**Cathodic protection** principles can be best understood by examining the corrosion cell. At the anode, metal is consumed, but at the cathode, the energy level is maintained. By making the entire structure a cathode, all surfaces that receive sufficient current can maintain their position on the energy hill. This is cathodic protection.

Cathodic protection requires an outside source of direct current that flows through the electrolyte onto the metal surface. One source for this current may be a DC power supply connected to a relatively inert electrode that is placed in the soil or water. Current is forced to flow from the electrode, through the electrolyte to the metal surface. This is **impressed current cathodic protection**.

Another current source may be obtained by connecting a metal that is higher on the energy hill to a metal that requires protection. The higher-energy material becomes the anode in an intentionally designed corrosion cell and sacrifices itself to protect the structure. This is **galvanic anode cathodic protection**.

Both impressed current and galvanic anode cathodic protection require that current flow from the cathodic protection anode, through the electrolyte, onto the cathode surface. Through the addition of energy, the cathode, or protected structure, maintains the energy level of the refined metal. A familiar application of cathodic protection is household hot water heaters. Small magnesium rods are installed inside the tank to provide cathodic protection to the submerged metallic components.

Because cathodic protection current must flow onto the metal surface, cathodic protection can be applied only when the structure is buried, submerged, or embedded in an electrolyte. Unfortunately, cathodic protection is not applicable for controlling atmospheric corrosion. It cannot be used in the form described to protect automobiles from corroding. Galvanizing, which is essentially the deposition of zinc on a steel surface, is a form of cathodic protection. The zinc is the anode and the steel is the cathode. In this case, the cathodic protection anode is directly applied to the surface of the cathode.

## Reference

Burke, N.D. and D.H. Kroon, "Corrosion and Corrosion Control," Technical Paper Library, CP-1, Corpro Companies, Inc.; <http://www.corpro.com>.

UTTU thanks James T. Lary, [jlary@Corpro.com](mailto:jlary@Corpro.com), Corpro Companies, Inc., for his help on this article.



## Research notes

### BTX Biodegradation in Activated Sludge Under Multiple Redox Conditions

Guihua, M. and N.G. Love, *Journal of Environmental Engineering*, June 2001; <http://ojsps.aip.org/eeo>

Researchers evaluated BTX biodegradation in an activated sludge system that used the following redox regimes: denitrifying, anoxic, microaerobic (low-dissolved oxygen) and aerobic. The definition of microaerobic varies from a constant dissolved oxygen level of < 1 mg/L to 8.9 mg/L. Researchers accomplished their evaluation by

- tracking BTX and electron acceptor concentrations and enzyme activities specific to different pathways for BTX metabolism
- defining the oxidation-reduction potential under which BTX biodegradation occurred
- evaluating features of microaerobic metabolism

Guihua and Love (2001) concluded:

- toluene and *m*-xylene were biodegraded under denitrifying conditions
- benzene, *p*- and *o*-xylene were recalcitrant under denitrifying conditions; however, they were biodegraded by aerobic dioxygenase pathways under aerobic (DO > 4 mg/L) and microaerobic (DO < 0.2 mg/L) conditions
- biodegradation rates for benzene, *p*- and *o*-xylene were twice as fast under nitrate or nitrite-supplemented microaerobic conditions as compared to NO<sub>x</sub>-deficient microaerobic conditions
- biodegradation rates for benzene, *p*- and *o*-xylene, facilitated by NO<sub>x</sub>-supplemented microaerobic and aerobic conditions, were similar
- microaerobic metabolism of benzene, *p*- and *o*-xylene was facilitated by certain enzymes that were capable of being induced under NO<sub>x</sub>-supplemented microaerobic conditions to levels comparable to those under aerobic conditions
- oxidation reduction potential is a sensitive indicator of the electron acceptor condition present in the sequencing batch reactor systems and shows promise for use in full-scale systems that incorporate denitrifying and/or microaerobic treatment strategies

### Evaluation of Standard Methods for the Analysis of Methyl Tert-Butyl Ether and Related Oxygenates in Gasoline-Contaminated Groundwater

Halden, R.U., Happel, A.M. and S.R. Schoen, *Environmental Science & Technology*, Vol. 35, No. 7, 2001; <http://www.pubs.acs.org/>.

Researchers studied evaluated standard methods (Methods 8021B, 8260B and D4815) for identifying MTBE and other oxygenates with the intention of

- assessing and comparing the performance of analysis tools that were straightforward, inexpensive and available
- identifying and characterizing limitations of existing analytical protocols
- providing information critical for interpreting analytical results archived in LUST databases
- providing guidance on method selection to LUST site regulators and managers

This study was based on "a statistical analysis of 424 groundwater split samples collected in 1996 and analyzed by a large number of commercial laboratories using both GC-PID and GC-MS methods."

Halden and others (2001) concluded that care is essential when analyzing MTBE data. Site managers, for instance, must be aware that although Method 8021B is commonly used, it gives low data quality at elevated TPH concentrations. Furthermore, researchers assert that "EPA Method 8020A/21B (photoionization detection) was unfit for monitoring tert-butyl alcohol (TBA) and frequently yielded false-positives (12-50 percent of samples) and inaccurate results when ether oxygenates were monitored in aqueous samples containing high concentration of TPH (> 1,000 µg/L)." This test can, however, be considered if the matrix is well-characterized and contains no or only minimal levels of interfering TPH compounds. The authors suggested that a "robust, accurate, precise and widely applicable monitoring tool" for TBA and ether oxygenates would be EPA Method 8260B and ASTM Method D4815. The authors also advised that care be taken when measuring MTBE concentrations with high TPH concentrations obtained by using EPA Methods 8020/21. These MTBE concentrations could be inaccurate or false-positive.

### Hydrogeological Parameter Estimation Using Geophysical Data: a Review of Selected Techniques

Hubbard, S.S. and Y. Rubin, *Journal of Contaminant Hydrology*, Vol. 45, 2000; <http://www.elsevier.com/locate/jconhyd>.

This article describes some of the geophysical techniques used to characterize field sites. According to Hubbard and Rubin (2000), "Much of the problem and initial cost of subsurface remediation comes from field site characterization. Three-dimensional information about the heterogeneous subsurface is needed to identify the key controls on the flow and contaminant transport processes." The processes described include radar and seismic methods that acquire data from a densely sampled gridwork in combination with conventional direct hydrological measurements. Parameters estimated include saturation, permeability and hydraulic parameter spatial correlation.

### Articles of additional interest include :

"Abiotic Oxidation of Petroleum Bitumens Under Natural Conditions," Charrie-Duhaut, A., Lemoine, S., Adam, P., Connan, J., and P. Albrecht, *Organic Chemistry*, Vol. 31, 2000; <http://www.elsevier.nl/locate/orggeochem>.

"Automotive Sources of Carcinogenic Polycyclic Aromatic Hydrocarbons Associated with Particulate Matter in the Chesapeake Bay Region," Dickhut, R.M., Canuel, E.A., Gustafson, K.E., Liu, K., Arzayus, K.M., Walker, S.E., Edgecombe, G., Gaylor, M.O. and E.H. Macdonald, *Environmental Science & Technology*, Vol. 34, No. 21, 2000; <http://www.pubs.acs.org>.

"Carbon Isotope Fractionation During Anaerobic Biodegradation of Toluene: Implications for Intrinsic Bioremediation," Ahad, M.E., Sherwood Lollar, B., Edwards, E.A., Slater, G.F. and B.E. Sleep, *Environmental Science & Technology*, Vol. 34, No. 5, 2000; <http://www.pubs.acs.org>.

"Effects of a Trapped NAPL on the Diffusive Release of Organic Contaminants from Saturated Soils," Schaefer, C.E. and D.S. Kosson, *Environmental Engineering Science*, Vol. 17, No. 3, 2000; <http://www.liebertpub.com/>.

"Engineering Reliability Assessment of Contaminant Removal by Pneumatic Fracturing," Ding, Y., Schuring, J.R. and P.C. Chan, *Practice Periodical of Hazardous, Toxic and Radioactive Waste Management*, Vol. 4, No. 1, January 2000; ASCE, 1801 Alexander Bell Drive, Reston, Virginia 20191-4400.

"Environmental Implications on the Oxygenation of Gasoline with Ethanol in the Metropolitan Area of Mexico City," Schifter, I., Vera, M., Diaz, L., Guzman, E., Ramos, F. and E. Lopez-Salinas, *Environmental Science & Technology*, Vol. 35, No. 10, 2001; <http://www.pubs.acs.org/>.

"Hydrophobic Hollow Fiber Membranes for Treating MTBE-Contaminated Water," Keller, A.O. and B.G. Bierwagen, *Environmental Science & Technology*, Vol. 35, No. 9, 2001; <http://www.pubs.acs.org/>.

"Physical versus Biological Hydrocarbon Removal During Air Sparging and Soil Vapor Extraction," Aelion, C.M. and B.C. Kirtland, *Environmental Science & Technology*, Vol. 34, No. 15, 2000; <http://www.pubs.acs.org/>.

"Oil Spill Remediation Using Magnetic Separation," Chun, C.L., and J.W. Park, *Journal of Environmental Engineering*, May 2001; <http://ojs.aip.org/eeo>.

"Remediation of Petroleum Refinery Waste Pond Contents: A Case Study," Loehr, R.C., Webster, M.T., Johnson, D.L., Kegel, E.M. and J.D. Meyers, *Practice Periodical of Hazardous, Toxic and Radioactive Waste Management*, Vol. 4, No. 1, October 2000; ASCE, 1801 Alexander Bell Drive, Reston, Virginia 20191-4400.

"Stochastic Analysis of Oxygen- and Nitrate-Based Biodegradation of Hydrocarbons in Aquifers," Kaluarachchi, J.J., Cvetkovic, V. and S. Berglund, *Journal of Contaminant Hydrology*, Vol. 41, 2000; <http://www.elsevier.com/locate/jconhyd>.

"The Efficiency of Capping to Control Air Emissions from Exposed Contaminated Sediments and Dredged Material," Ravikrishna, R., Choy, B.C., Valsaraj, K.T., Reible, D.D., Thibodeauz, L.J., Price, C.B. and J.M. Brannon, *Environmental Engineering Science*, Vol. 17, No. 2, 2000; <http://www.liebertpub.com>.



## Information sources

### U.S. EPA publications and information

Publications that can be downloaded from <http://clu-in.org/techpubs.htm> include

- Applying the Concept of Effective Data to Environmental Analyses for Contaminated Sites (EPA 542-R-01-013)
- Clarifying DQO Terminology Usage to Support Modernization of Site Cleanup Practice (EPA 542-R-01-014)
- Initiatives to Develop Web Sites Including Information about Brownfield Properties (EPA 542-R-01-017)
- The Relationship Between SW-846, Performance Based Measurement Systems (PBMS) and Innovative Analytical Technologies (EPA 542-R-01-015)
- Development of Recommendations and Methods to Support Assessment of Soil Venting Performance and Closure (EPA 600-R-01-070), <http://www.epa.gov/ada/pubs/reports.html>

Handbook of Groundwater Protection and Cleanup Policies for RCRA Corrective Action (EPA 530-R-01-015), <http://www.epa.gov/correctiveaction/resource/guidance/gw/gwhandbk/gwhndbk.htm>

Summary of Workshop on Biodegradation of MTBE (EPA 626-R-01-001A), <http://www.epa.gov/ORD/NRMRL/Pubs/625R01001/625R01001.pdf>

### U.S. EPA Web sites

Assessment and Remediation of Contaminated Sediments (ARCS), <http://www.epa.gov/glnpo/arcs/arcsguide.html>

Forum on Managing Contaminated Sediments at Hazardous Waste Sites, <http://www.epa.gov/superfund/new/sedpresent.htm>

Great Lakes National Program Office (GLNPO), <http://www.epa.gov/glnpo/sediments.html>

Watershed Assessment, Tracking and Environmental Results, <http://www.epa.gov/waters>

### Other Web sites

European Sediment Research Network (SedNet), <http://www.mep.tno.nl/SedNet/>

NOVA, <http://www.pbs.org/wgbh/nova>

Sediment Management Work Group, <http://www.smwg.org>

Sediments Remediation Action Team, <http://www.rtdf.org>

### Other publications

Annual Report of the RTDF Phytoremediation Action Team: TPH Subgroup: Cooperative Field Trials, <http://www.rtdf.org/public/phyto/phytodoc.htm>

Methods for Determining Inputs to Environmental Petroleum Hydrocarbon Mobility and Recovery Models (API 4711), <http://api-ep.api.org/filelibrary/4711.pdf>

UTTU obtained this information from Contaminated Soil Sediment & Water (<http://aehsomag.com>), Iowa Groundwater Quality (<http://www.igwa.org/>), and TechDirect (<http://clu-in.org/techpubs.htm>). UTTU thanks the editors and writers for allowing us to reprint this information.