BENEFICIAL USES OF NUCLEAR WASTES

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ABSTRACT

Deactivation, decommissioning, and final decontamination of radioactive process lines or facilities at DOE sites yield a variety of waste materials and waste forms. A certain fraction of these waste materials contain fairly concentrated residues of Pu and Am. Pu and Am have been produced by neutron capture of U-238 leading to radioactive isotopes of Pu and Am that are relatively short lived with respect to U-238 and have a great quantity of stored energy that is given off as decay energy via alpha-particle emissions. The decay energy available in concentrates of Pu and Am can be converted for beneficial purposes rather than disposed in geologic disposals where the decay heat is lost to the geologic matrix as was done with the Pu and Am produced in nuclear reactors, which was considered a nuisance and disposed of at great expense rather than being gainfully utilized. Some beneficial uses of nuclear materials currently destined for disposal are outlined in this presentation.

1) The use of $^{241}$Am or $^{238}$Pu for radiolytic conversion of CO$_2$ to CO, which can subsequently be catalytically converted to methanol or fuel.
2) The radiolytic decomposition of water or plastic materials to generate hydrogen.
3) The fabrication of miniature neutron sources by nanoscience techniques for in-situ implantation in tumors.
4) The fabrication of wide-area sources for detection of explosives.
5) The production of tritium with a passive cylindrical neutron source with essentially no secondary waste.
6) Accelerated aging studies of whole components by irradiation with α-particles, x-rays, gamma-rays, and neutrons, either individually or in any combination of radiations.
7) Tracking SNM by unique high energy gamma-rays.
8) Detecting land mines with wide-area neutron sources.
9) The chemistry in highly ionized fields; enhancement of chemical reactions.
10) The destruction of unwanted organic gas and gaseous warfare agents.

The successful development and application of the technical areas would open an entire new technological field that would be beneficial to humankind and be of great benefit to the environment and the taxpayer.

BACKGROUND

The number of articles published in scientific journals on the greenhouse effect has increased dramatically in the past ten years. This proliferation of articles in scientific journals is now being followed by an increase of articles in newspapers, news magazines and even popularly distributed magazines. Early articles on the greenhouse effect causing living conditions that would be unbearable range from a few thousand years to 500,000 years. After several recent natural disasters in the past two-three years, such as storms and floods in
Australia, Korea, and California; the disappearance of glaciers and ice flows throughout the globe; and the droughts and forest fires in Spain (1997), Mexico (1998), Indonesia (1998), and the Rocky Mountain region of the United States, including a devastating forest fire that consumed over four hundred dwellings in Los Alamos, New Mexico (May 2000); many new scientists are beginning to appear as proponents of taking new serious actions to reduce the effects of continued and increased pumping of CO\(_2\) gas into the atmosphere. Advances in atmospheric modeling has led scientists to move from a position of furthering the greenhouse debate and continued study of the problem to what can be done to mitigate the increase of CO\(_2\) concentration in the atmosphere. The United States of America has been reluctant or unwilling to be part of a global effort to reduce greenhouse emissions to the atmosphere as evidenced by the 95-0 senate vote to oppose the Kyoto treaty agreement. Countries that once pumped small quantities of CO\(_2\) into the atmosphere are now pouring larger and larger quantities of CO\(_2\) into the atmosphere as coal-burning and automobile power increases. China may exceed the United States in CO\(_2\) emissions by the year 2020. Although there may still be an ongoing greenhouse effect debate, the increase in concentration of CO\(_2\) in the earth’s atmosphere is fairly well agreed upon by most scientists. The increase in CO\(_2\) is demonstrated by the following concentrations.

<table>
<thead>
<tr>
<th>Time</th>
<th>CO(_2) Concentrations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pre-Industrial*</td>
<td>270 ppm</td>
</tr>
<tr>
<td>1958</td>
<td>315 ppm</td>
</tr>
<tr>
<td>2000</td>
<td>360 ppm</td>
</tr>
</tbody>
</table>

* Measured by air bubbles in Greenland ice sheet.

The solutions offered to solve or mitigate the effect of global warming are many, including:

1. Conduct more studies of greenhouse effect on global environment
2. Devise more workable global plan to limit the quantity of CO\(_2\) released to the atmosphere
3. Develop new technologically driven alternatives to CO\(_2\) producing energy sources
4. Curtail cutting down forested areas around the globe
5. Conduct CO\(_2\) sequestration of CO\(_2\) in large industrial plants
6. Mitigate the greenhouse effect by modification of life forms and plant growth on the earth and ocean
7. Develop technologies that can economically or near economically convert CO\(_2\) to an intermediate or final useable product

Of the seven general means listed for minimizing the greenhouse effect on the earth, only number 2 has the capability to have an immediate impact on the global environment. However, political consideration and well-formed lifestyles will not effectuate this option unless the planet has a sudden catastrophic change that adversely affects populations.

This paper provides the preliminary results of a technological approach that may provide a methodology for conversion of CO\(_2\) to CO and O\(_2\). This methodology will not provide an ultimate solution to the unwanted increase in global CO\(_2\) concentration but may provide the base from which other scientific technologies may flow that can have a mitigating influence in slowing down the rate of CO\(_2\) concentrations emitted to the atmosphere.
INTRODUCTION

The conversion of CO$_2$ to a useful product by chemical or catalytic methods has long been a challenge to chemists from the early history of chemistry. Unfortunately, the thermodynamics of this conversion like the conversion of water to H$_2$ and O$_2$ is economically unfavorable. However, the continued large scale use of fossil fuels for energy is having a long-term deleterious effect on the globe and new sources of energy that do not increase the rate of CO$_2$ concentrations in the global atmosphere are needed. This great global need has led researchers to apply new or modified technologies to seek new methodologies for more effective utilization of existing energy systems or to implement new alternative energy systems that do not increase CO$_2$ emissions to the atmosphere.

This technological approach is to use the alpha-decay energy from certain actinides that are now considered to be nuclear wastes for the purpose of the conversion of CO$_2$ to CO, which can then be catalytically converted to a useful fuel.

SCOPE OF EXPERIMENTAL WORK

The goal of this experimental work is to advantageously utilize the abundant source of energy (MeV) from the nuclear decay process, in the form of particle emissions, from high specific activity actinides or long-lived fission or activation products to convert carbon dioxide (CO$_2$) or water (H$_2$O) to a usable fuel source using a passively operated system with an efficiency that makes the conversion economically feasible. This technical article reports a new methodology for the radiolytic conversion of highly stable compounds such as the greenhouse gas, (CO$_2$), to a more active chemical intermediate such as carbon monoxide (CO) that can be catalytically converted to a useful fuel form such as methane (CH$_4$), methanol (CH$_3$OH), or other energy bearing compounds.

The successful demonstration of this invention is dependent on optimization of at least 5 major parameters:

1) the dimensional design of the irradiation chamber;
2) the alpha-emission rate in the chamber;
3) the conversion enhancement by an energy transference gas;
4) understanding and enhancing of ion-pair formation per $\alpha$-particle; and
5) precluding recombination of CO with other ions

The separation of CO from CO$_2$, O$_2$, and an energy transference gas with subsequent re-injection into the irradiation chamber was considered to be beyond the scope of this experimental work. The conversion of CO to a usable fuel form was also considered to be beyond the scope of this study. However, the use of the highly ionized transference gas phase in the irradiation chamber to accelerate or catalyze the formation of new compounds or compounds that now require high temperature and pressure would be considered to be within the scope this study.
IRRADIATION CHAMBER

Perhaps, the key to this study is the design of an irradiation chamber that provides a high flux of nuclear decay particles from high specific activity nuclides such as $^{238}$Pu, $^{239}$Pu, $^{241}$Am, $^{90}$Sr, $^{137}$Cs that are plated on the inside surface of the irradiation chamber walls. The irradiation chamber is designed to allow optimum utilization of the mean free path length and thus specific ionization potential of a specific nuclear decay particle. The irradiation chamber must be designed to fully utilize the highly energetic nuclear decay particles with energy in the million electron volt (MeV) range to effectively ionize compounds along the path length of the particle.

To demonstrate the effectiveness and efficiency of this concept, an irradiation chamber was designed and fabricated where $^{241}$Am was deposited on the internal walls of the chamber as shown in Fig. 1. Irradiation Chamber.

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**Fig. 1. Irradiation Chamber**

- **Length** = 17 cm
- **Diameter** = 6 cm
- **Ni Thickness** = ~ 0.5 cm
- **Lateral Surface Area (Internal)** = 320.4 cm$^2$
- **Volume** = 480 cm$^3$
- **$^{241}$Am Deposited** = 97 mg
- **$^{241}$Am Activity** = $1.23 \times 10^{10}$ d/s/chamber
The irradiation chamber is 6 cm in diameter and 17 cm in length to allow the alpha-decay particle of 5.48 MeV energy to travel its full path length in air. The path length of an alpha particle from $^{241}\text{Am}$ decay is calculated to be ~ 4-5 cm in $\text{CO}_2$. See Appendix C for the calculation and Appendix B for the nuclear characteristics of $^{241}\text{Am}$.

The metal hemi-shell could be composed of a select number of metals, but nickel was chosen because of its inertness to acid deposition of $^{241}\text{Am}$ with subsequent drying by heating. A preferable way to deposit or electroplate a uniform thin layer of $^{241}\text{Am}$ would be to electro-deposit or sputter the metal into the hemi-shell inner surface. As much as 5 gm of material could be uniformly distributed along the inner surface of the nickel hemi-shells. The nickel hemi-shells were ribbed to give a rough surface with an Electrode Discharge Machine (EDM). The thickness of the nickel plate was ~0.5 cm to totally attenuate the 60 keV gamma-ray that is emitted 36% of every 100 decay events or alpha particles.

**CHAMBER ALPHA-EMISSION RATE**

The total alpha-particle emission rate of the 97 mg of $^{241}\text{Am}$ deposited on the inner surface of the heavy nickel hemi-shells gave a calculated $4\pi \alpha$-emission rate of $1.23 \times 10^{10}$ $\alpha$'s/sec/97 mg (as calculated in Appendix A). However, ~ 50% of the alpha-particles would be emitted into the gas phase and the remainder would be emitted into the walls of the nickel hemi-shells. So for a $2\pi$ geometry, the $\alpha$-emission rate into the gas phase would be about $6.2 \times 10^9$ $\alpha$’s/sec. The 97 mg of $^{241}\text{Am}$ placed on the inner-surface of the nickel hemi-shell was deposited by placing micro-droplets of an Am chloride solution on the inside surface of hemi-shell and placing the wall of the hemi-shell on a hot plate to dry the salt. The salt crystals that form as a result of the drying process would contribute to absorption of some of the alpha-particle energy and that absorption is expected to be ~ 20%. This would give an effective alpha-particle emission rate of $\sim 5 \times 10^9 \alpha$’s/sec/chamber. As stated earlier, a different and more efficient plating technique could replace the dried-salt technique and give an $^{241}\text{Am}$ $\alpha$-emission rate of $\sim 2.5 \times 10^{11}$ $\alpha$’s/sec/chamber.

**ENERGY TRANSFERENCE GAS**

When pure $\text{CO}_2$ was injected into the irradiation chamber, the production of CO varied with the duration of irradiation. The conversion rate seemed to be high at the beginning of the irradiation and then diminished with time. The total CO generated continued to increase but the generation rate decreased with concentration. The average CO generation rate was ~ 100 ppm per hour over a 24 hour period during initial tests.

Tests were conducted with 50% $\text{CO}_2$ and 50% nitrogen because nitrogen was suspected of being a good energy transference gas. The initial conversion rate for $\text{CO}_2$ to CO increased to ~ 2,000-3,000 ppm per hour and then decreased as the gas phase was stagnant over a 24-hour period perhaps because of radiolytic conversion of CO to C and O.

Tests were also conducted with 50% $\text{CO}_2$ and a 50% Ar atmosphere. The gas generation rate was initially 3,000-4,000 ppm per hour but as before the rate decreased with time, eventually decreasing to <1,000 ppm per hour during a 24 hour test.
Tests conducted with a 50% CO\textsubscript{2} and 50% air showed a disappointing conversion rate of <100 ppm after 4 hours. This indicates that oxygen present in the gas phase is ionized and perhaps recombined with the CO, to form CO\textsubscript{2}. The overall generation rate with air over a 24-hour period was very poor.

Tests using pure CO\textsubscript{2}, N\textsubscript{2} + CO\textsubscript{2}, Ar + CO\textsubscript{2} and other CO\textsubscript{2}-gas mixtures, when mixed with pure H\textsubscript{2} gas to give a 1-2 % mixture, immediately showed a very high concentration of H\textsubscript{2}O in the gas space. This radiolytically formed water was quite detrimental to the GC used for making these measurements. However the reaction of H\textsubscript{2} in the presence of O\textsubscript{2} formed by the radiolytic decomposition of CO\textsubscript{2} to form H\textsubscript{2}O, provided the basis for using this methodology to solve the problem of H\textsubscript{2} generation in shipping containers that contain high alpha-particle rates.

The radiolytic formation of CO seemed to be most efficient early in the irradiation period (first 1-2 hours) with gases such as N\textsubscript{2} and Ar that did not contain O\textsubscript{2}.

**ALPHA-PARTICLE PATHWAY DYNAMICS**

Actinides such as U, Pu, and Am are heavy elements that contain nuclei that are unstable to radioactive decay with emission of a charged particle such as an alpha or beta particle. The half-life of the actinide and the energy of the decay particles is proportional to the instability of the specific actinide. Radioactive decay by emission of an alpha particle is the primary mode of decay for actinides such as \textsuperscript{238}Pu, \textsuperscript{239}Pu, \textsuperscript{241}Am, and \textsuperscript{244}Cm. The alpha particle begins as the nucleus of He atom with 2 protons and 2 neutrons and 2 electrons. However, the high energy of the decay process releases an alpha particle with such high velocity that the alpha particle is stripped of its electrons and is a doubly charged particle. The primary alpha particle emitted from the decay of \textsuperscript{241}Am contains 5.486 MeV of energy and is relativistic near the point of emission. The diameter of the alpha particles are about $2 \times 10^{-13}$ cm whereas the atom to atom bond distance between a carbon to oxygen atom is about $1.2 \times 10^{-8}$ cm. Consequently, the possibility of the alpha particle colliding with another nucleus in a gaseous medium is highly unlikely as shown in Fig. 2. Diagram of Alpha-Particle Path Length Dynamics.
However, the number of CO$_2$ atoms per cubic centimeter of gas is about $2.7 \times 10^{19}$ molecules. Six electrons surround each carbon atom and each oxygen atom has 8 electrons that have a high probability of colliding with the alpha particle that travels 4-5 cm in a CO$_2$ matrix. The collision of an alpha particle with a rest mass of $6.6 \times 10^{-24}$ gm with an electron with a rest mass of $9.1 \times 10^{-28}$ gm, mass ratio of 7,253, results in acceleration of electrons and deceleration of the alpha particles. The energetic secondary electrons are available to disrupt the bonds between carbon and oxygen atoms lending to dissociation of the CO$_2$ molecule or ionization of the atoms. A multiatomic molecule such as CO$_2$ can have atoms with translational (linear) as well as rotational kinetic energy relative to the central carbon atom. This linear alternate symmetric and asymmetric stretching changes from potential to kinetic energy similar to a simple harmonic oscillator as shown in Fig. 3. Modes of Vibration within CO$_2$ Molecules.
The atoms within the CO$_2$ molecule vibrate with a linear and rotational velocity of about $4.4 \times 10^4$ cm/sec. An alpha particle from $^{241}$Am will have an initial velocity of about $2 \times 10^9$ cm/sec. As the alpha particle passes through CO$_2$ gas with a pathlength of 4-5 cm, it will interact with the electrons surrounding the oxygen and carbon atoms and impel some of these secondary electrons at high velocities that can lead to 60-80% of the ionizations produced by the main alpha particle. As the doubly charged alpha particle loses energy by interaction with the electron shells of C and O atoms, the alpha particle will itself pick up one or two electrons near the end of its path that will be characterized by straggling. The effectiveness of the rebound electrons in breaking the carbon-oxygen bond relative to the alpha particle itself is not known. But the fact that we have been able to effect over $7.5 \times 10^6$ conversions per alpha particle, points to secondary electrons being responsible for the conversion.

**ION-PAIR FORMATION PER ALPHA PARTICLE**

An ion-pair due to radiolytic modification of a gaseous compound may be described as the formation of a positively charged ion and an equally charged negative ion (most usually an electron) resulting from an atomic interaction of an incident radiation particle or ray with a target neutral atom or molecule or medium with such molecules. A curve showing the average number of ions per unit of distance along the path length of the initially monoenergetic beam of ionizing
particles, such as an $\alpha$ or $\beta$-particle, is called a Bragg Curve or Bragg Ionization Curve. The average specific ionization of an alpha particle from $^{241}$Am passing through a gas mixture is a function of four parameters:

1) kinetic energy, of alpha particle,
2) velocity, of alpha particle,
3) range of alpha particle in gas matrix, and
4) ionization potential including secondary electron ionization.

The statistical average of ion pairs formed along the path of an alpha particle varies with the four above parameters and can be seen in the Bragg Curve shown in Fig. 4. Bragg Curve, showing ionization vs. distance.

![Fig. 4. Bragg Curve, showing ionization vs. distance](image)

The initial kinetic energy of the alpha particle from decay of the $^{241}$Am nucleus approaches relativistic velocities but begins to decrease with distance (path length) because of elastic collisions and electron stripping interactions. The alpha-particle with an initial charge of 2+ and large mass (4 AMU) has a high specific ionization and begins to slow down to a kinetic energy level of ~ 1 MeV. This energy level is comparable to the energy of the orbital velocity of the outer K-shell electrons of the target CO$_2$ atoms. At this velocity (kinetic energy) the alpha particle from decay of $^{241}$Am (5.486 MeV) begins to experience inelastic collisions with CO$_2$ atoms, which give rise to nuclear deceleration rather than electronic slow down. It is during this inelastic collision phase that optimum dissociation of CO$_2$ might occur.
An alpha-particle emitted from the decay of $^{241}$Am (5.486 MeV) is calculated to have a range in CO$_2$ of 4-5 cm and a $\Delta E/\Delta x$ of 1.09 MeV/cm (see Appendix C for the calculation). For a 5.486 MeV alpha particle with a path length of 4-5 cm in CO$_2$, the specific ionization or number of ion pairs formed per cm of travel is calculated as follows:

\[
\frac{(5.486 \text{ MeV}) \times (10^6 \text{ eV/MeV})}{(34 \text{ eV/ion-pair})(4.5 \text{ cm})} = 3.6 \times 10^4 \text{ ion pairs/cm} \quad \text{(Eq. 1)}
\]

\[3\text{CO}_2 \xrightarrow{\alpha\text{-particles}} C + 2\text{CO} + 2\text{O}_2\]

Where:
- 5.486 MeV = Energy of $^{241}$Am $\alpha$-particle
- 34 eV/ion-pair = Average ionization energy for CO$_2$
- 4.5 cm = Approximate path length of 5.486 MeV $\alpha$-particle in CO$_2$

For 4-5 cm of travel the alpha particle will generate:

\[
(3.6 \times 10^4 \text{ ion pairs/cm})(4.5\text{cm}) = 161,353\text{ion pairs} \quad \text{(Eq. 2)}
\]

The relative ratio of elastic to inelastic collisions of the alpha particle that result in dissociation of the CO$_2$ molecules is not known and is a key factor in the optimization of conversion reactions. For the overall process of converting CO$_2$ to CO with an economical methodology, each alpha particle must make several hundred thousand molecular conversions. Assuming the conversion rate of CO$_2$ to CO in an optimum energy transference gas is 1,000 ppm per hour, the number of conversions necessary to achieve that rate is calculated as follows.

At 1,000 ppm/hr, the number of moles @ STP =

\[
\frac{1,000 \mu \text{mol}/\text{hr}}{2.24 \times 10^7 \mu \text{mol/mole}} = 4.46 \times 10^{-5} \text{ mole/L/hr} \quad \text{(Eq. 3)}
\]

\[
(4.46 \times 10^{-5} \text{ mole/L/hr})(6.02 \times 10^{23} \text{ molecules/mole}) = 2.68 \times 10^{19} \text{ molecules/L/hr} \quad \text{(Eq. 4)}
\]

\[
\frac{2.68 \times 10^{19} \text{ molecules/L/hr}}{(4.92 \times 10^9 \text{ molecules/L/\alpha\text{-particle}})(3.60 \times 10^6 \text{ sec/hr})} = 1.5 \times 10^6 \text{ molecules/\alpha\text{-particle}} \quad \text{(Eq. 5)}
\]

At 5,000 ppm/hr the number increases to $7.56 \times 10^6$ molecules/L/\alpha\text{-particle}. The empirically determined number of $7.56 \times 10^6$ molecules/L/\alpha\text{-particle} is about a factor of ten higher than the theoretically calculated number of ion pairs. This factor of 10 may point to an enhancement by the carrier or energy transference gas.

**RECOMBINATION OF CO WITH OTHER IONS**

The primary reaction that governs the generation of CO is:

\[
(\text{Eq. 6})
\]
However, many competing reactions can occur to lessen the yield of the primary reaction.

$$2CO + O_2 \xrightarrow{\text{activity}} 2CO_2 \quad \text{(Eq. 7)}$$

$$2CO \xrightarrow{\text{activity}} 2C + O_2 \quad \text{(Eq. 8)}$$

An increasing concentration of $O_2$ appears to enhance reaction 2 and 3. Although the alpha-particle flux will result in dissociation of $CO_2$, an increase in the concentration of CO eventually makes CO vulnerable to dissociation, also. As stated in an earlier section, the presence of $N_2$ or Ar were beneficial in increasing the concentration of CO for the first hour or two in the irradiation chamber, however, the generation rate slowed down after a CO concentration of about 3,000 to 4,000 ppm was achieved. The flow rate through an irradiation chamber and the residence time in the chamber becomes important to maximize the generation of CO and minimize the recombination of CO and $O_2$.

The ratio of $CO_2$ to the energy transference gas that will maximize the generation of CO in a high alpha-activity field is important but the complete effect has not been determined at this time. Addition of a beta emitter such as $^{90}\text{Sr}$ or $^{137}\text{Cs}$ to the end plate of the irradiation chamber would be expected to augment the generation of CO but this has not yet been tested. The beta particles have a much longer path length than alpha particles and would be allowed to travel along the longer axial length of the irradiation chamber. Beta particles from remote handled spent fuels have been shown to be highly effective for recombination of $H_2$ and $O_2$ and are also expected to be effective for dissociation of $CO_2$ to form CO. An irradiation chamber of beta activity solely, has not been tested and may be of interest in determining the overall effectiveness of beta radiation vs. alpha radiation.

CONVERSION OF EXPERIMENTAL SCALE CHAMBER TO ENGINEERING SCALE MODEL

The experimental chamber designed and fabricated for demonstration of the proof-of-concept was the basis for calculations of a larger engineering scale model. The potential of the engineering scale model for producing a final usable product such as methane, methanol, ethane, ethanol or some other usable intermediate or fuel source has been calculated with appropriate assumptions, as follows.
Irradiation Chamber Assumptions:

- Length = 2m = 200 cm
- Diameter = 8 cm
- Lateral Area (2\(\pi r h\)) = 5,026 cm
- Volume (\(\pi r^2 h\)) = 10.05 L
- \(^{241}\text{Am Mass (per cm}^2\)) = 20 mg/cm
- \(^{241}\text{Am Mass (total)} = 100.5 gm
- \(^{241}\text{Am Activity (per cm}^2\)) = 2.54 \times 10^9 d/s/cm
- \(^{241}\text{Am Activity (total)} = 1.27 \times 10^{13} d/s/chamber = 344 \text{Ci/chamber}
- Flow Rate = 10 L/min
- \(\text{CO}_2\) to CO Conversion Rate = 5,000 ppm/min = 5,000 \(\mu\)L/L/min = 50,000 \(\mu\)L/chamber/min @ 2.24 \times 10^7 \(\mu\)L/mol = 2.23 \times 10^{-3} mol/chamber/min = 3.2 mol of CO per day

1 mol CO yields 1 mole CH\(_3\)OH

Therefore = 3.2 mol of CH\(_3\)OH per day/chamber

\[
\frac{(3.2 \text{ mol/day}) (32.04 \text{ gm/mole})}{0.79 \text{ gm/cm}^3} = 130 \text{ cm}^3 \text{ methanol/day/chamber}
\]

If we maintain 10 chambers/module, our rate of production would be 1.3 L/day/module. Mobile Corporation has reported the commercial conversion of methanol to gasoline with a zeolite catalyst.

**ENHANCEMENT OF EFFICIENCIES**

The conversion of \(\text{CO}_2\) to CO can be optimized by additional experimentation to determine the basic mechanisms that could improve the conversion. It is currently unknown what the percentage of conversions are from multiple secondary electron interactions during the early (0-2 cm) life of an \(\alpha\)-particle or from direct interaction of the \(\alpha\)-particle near the end of the path (3-5 cm). It might be beneficial to increase the temperature and therefore the vibrational, translational, and rotational energy of the \(\text{CO}_2\) molecule, which might lower the threshold energy, required to release an oxygen atom. Other physical properties that could be modified to yield a higher rate of conversions is to change the dimensions of the irradiation chamber, increase pressure in the irradiation chamber, add a beta emitting radionuclide, apply a magnetic field, apply UV or laser light to the irradiation chamber, apply ultrasonic vibration to the gas volume, or optimize the concentration of the energy transference gas through a method of continuous variations. The concentration of CO in a \(\text{CO}_2\) matrix will be limited according to the secondary reactions for radiolytic degradation of CO to carbon and oxygen and the recombination of CO with O\(_2\). The highest efficiencies for conversion of \(\text{CO}_2\) to CO would be if every molecule of CO produced would be immediately separated from the radiolytic field because of a difference in physical or chemical properties. For example, as a supercritical fluid,
carbon dioxide can undergo radiolytic decomposition to CO, which would become a gas and be easily separated from the CO$_2$ matrix. This instantaneous separation of CO from CO$_2$ would make the CO unavailable for continued bombardment by alpha particles because of the difference in physical properties between CO$_2$ and CO as shown in Table I. Physical Properties of CO$_2$ and CO

<table>
<thead>
<tr>
<th>Gas</th>
<th>MW</th>
<th>Density @ 0°C</th>
<th>MP</th>
<th>BP</th>
<th>Solubility in H$_2$O @ 0°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO$_2$</td>
<td>44.01</td>
<td>1.977 g/L</td>
<td>-56.6°C</td>
<td>-28.5°C</td>
<td>171.3 gm/100 cm$^3$</td>
</tr>
<tr>
<td>CO</td>
<td>28.01</td>
<td>1.250 g/L</td>
<td>-199.0°C</td>
<td>-191.5°C</td>
<td>3.5 gm/100 cm$^3$</td>
</tr>
</tbody>
</table>

The tremendous difference of physical properties of CO$_2$ and CO could be advantageously used to separate CO from CO$_2$. As a supercritical fluid, CO$_2$ would remain as a fluid where as CO would evolve and be separated as a gas. Chemically, CO is much more reactive than CO$_2$ and could be separated out based on a reaction with certain other gases that would readily separate from CO$_2$. Molecular sieves with specific pore size matrices have been successfully used for separation of CO$_2$ from CO. A separation process that will separate each molecule of radiolytically generated CO from the bulk CO$_2$ and the irradiation field would augment the feasibility of this process. The most useful separation process would separate CO from CO$_2$ rather than CO$_2$ from CO.

**CONCLUSIONS**

The concept of using the decay energy of high specific activity actinides or fission/activation products for radiolytic dissociation of stable compounds such as CO$_2$ or H$_2$O has been shown to be feasible. The yield of this methodology must be improved by a factor of 10 to 50 for this technology to become economically feasible. This may be accomplished by improvements in the geometry and mitigation of radiolytic dissociation and recombination of CO at higher concentrations. The success of this technology resides with the willingness of the Department of Energy (DOE) in utilizing the large volumes of wastes containing high specific actinide activity ($^{238}$Pu, $^{239}$Pu, $^{241}$Am) and fission products such as $^{90}$Sr and $^{137}$Cs. These high specific activity nuclides are presently scheduled to be buried as wastes. Before disposal of high specific activity wastes, the US DOE should pursue the beneficial uses of these wastes. There are many other uses of actinide waste that would be beneficial for augmenting energy resources. This is but one specific proposal of many that could be pursued by the US-DOE, for the beneficial use of nuclear waste.
Appendix A
Calculation of Alpha-Particle Emission Rate

\(^{241}\)Am Nuclear Properties:

\[ t^{1/2} = 432.7 \text{ annums} \]

\[ \lambda = \frac{0.693}{(432.7 \ a) (365.25 \text{ d/a}) (1,440 \text{ min/a}) (60 \text{ sec/min})} \]

\[ = 5.075 \times 10^{-11} \text{ sec}^{-1} \]

\[ \frac{dN}{dt} = N\lambda \]

\[ \frac{dN}{dt} = \frac{(1 \ gm) (6.02 \times 10^{23} \text{ atoms/gm-atom}) (5.075 \times 10^{-11} \text{ sec}^{-1})}{241 \text{ gm/gm-atom}} \]

\[ = 1.268 \times 10^{11} \alpha' \text{ s/sec/gm} \]

\(^{241}\)Am plated on walls of chamber is \( \sim 97 \) mg.

\[ = 1.23 \times 10^{10} \alpha' \text{s/sec/97 mg} \]

Assume 2\{ alpha-emission into gas phase

\[ = 6.15 \times 10^{9} \text{ d/sec into gas phase} \]

Assume 20% of \( \alpha \) are absorbed by salt deposit, then

\[ = 4.92 \times 10^{9} \alpha' \text{s/sec into gas phase} \]

Assume 5,000 mg is plated on inner walls of Ni hemi-shell.

Then activity = \( 2.54 \times 10^{11} \alpha' \text{s/sec}/5 \text{ gm} \) @ 2\{
Appendix B
Nuclear Characteristics of $^{241}$Am

Half-Life $= 432.7$ Y
$\lambda = 5.075 \times 10^{-11}$ sec$^{-1}$
Specific Activity $= 1.268 \times 10^{11}$ d/s/g
$\alpha_1 = 5.486$ MeV (86.0%)
$\alpha_2 = 5.443$ MeV (12.7%)
$\alpha_3 = 5.389$ MeV (1.3%)

Gamma-rays with yields
- 59.5 keV  100% relative (36% absolute)
- 17.8 keV  51.2% relative
- 13.9 keV  37.5% relative
- 20.8 keV  13.8% relative
- 26.4 keV  7.0% relative
Appendix C
Calculation of $\Delta E/\Delta x$ and Range of Alpha-particle in CO$_2$

$$\Delta E/\Delta x = \left[ \frac{4\pi e^4 Z_1^2 Z_2 N}{(m_0 v^2)} \right] \ln \left[ \frac{2m_0 v^2}{I} \right]$$

Where:
- $\Delta E/\Delta x = \text{change in kinetic energy of charged particle in ergs/cm}$
- $e = \text{charge on electron (4.8 \times 10^{-10} \text{ stat c})}$
- $Z_1 = \text{charge of } \alpha \text{ particle (2)}$
- $Z_2 = \text{atomic number of interacting atoms per cm}^3 \text{ (effective) = 16 for CO}_2$
- $N = \text{number of interacting atoms per cm}^3 = 2.71 \times 10^{19} \text{ molecules/cm}^3 \text{ (STP)}$
- $m_0 = \text{rest mass of electron (9.1083 \times 10^{-28} \text{ g})}$
- $v = \text{velocity of } \alpha \text{-particle (cm/sec) = 1.63 \times 10^9 \text{ cm/sec}}$
- $I = \text{average excitation potential of atom (≈ 80 eV)}$
- $m = \text{mass of an } \alpha \text{-particle = 6.64 \times 10^{-24} \text{ g}}$
- $E = \text{energy of an } \alpha \text{-particle = 5.49 MeV}$

$$\text{KE (}\alpha\text{-particle)} = \frac{1}{2}mv^2$$
$$v^2 = 2 \left( \frac{KE}{m} \right)$$
$$v = (2.65 \times 10^{18} \text{ cm}^2/\text{s}^2)^{1/2}$$
$$= 1.63 \times 10^9 \text{ cm/s}$$

$$\ln \left[ \frac{2m_0 v^2}{I} \right] = \ln \left[ \frac{2(9.1083 \times 10^{-28})(2.65 \times 10^{18})}{80(1.6 \times 10^{12})} \right]$$
$$= \ln (37.71)$$
$$= 3.63$$

$$N = (6.025 \times 10^{23} \text{ molecules/mole})(0.001977 \text{ g/cm}^3)/(40 \text{ g/mole})$$
$$= 2.71 \times 10^{19} \text{ molecules/cm}^3 \text{ for CO}_2$$

$$\Delta E/\Delta x = \left[ \frac{4\pi e^4 Z_1^2 Z_2 N}{(m_0 v^2)} \right] \ln \left[ \frac{2m_0 v^2}{I} \right]$$
$$= \left[ \frac{4\pi(4.8 \times 10^{-10})^4(2)^2(16)(2.71 \times 10^{19})}{(9.1083 \times 10^{-28})(2.65 \times 10^{18})} \right] \ln 37.71$$
$$= \left[ \frac{4\pi(5.3 \times 10^{-38})(4)(16)(2.71 \times 10^{19})}{(2.41 \times 10^{-9})} \right] (3.63)$$
$$= 1.74 \times 10^{-6} \text{ erg/cm}$$
$$= (1.74 \times 10^{-6} \text{ erg/cm})/(1.6 \times 10^{-6} \text{ erg/MeV})$$
$$= 1.09 \text{ MeV/cm; equivalent to (5.49/1.09) = 5.03 cm}$$

An alternative but direct calculation of the range of an alpha-particle can be made according to the following relationship:

Range of $\alpha \approx 0.309 \text{ E}^{1.5}\text{ cm}$
$$\approx 0.309(5.49)^{1.5} \text{ cm}$$
$$\approx 3.97 \text{ cm}$$

Approximate range of $^{241}$Am $\alpha$-particle = 4-5 cm due to straggling