Could this New Technology Prolong Life on Earth? Radiolytic Conversion of CO₂ to a Usable Fuel

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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 Early articles on the greenhouse effect causing living popularly distributed magazines. 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 CO₂ 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 CO₂ 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 CO₂ into the atmosphere are now pouring larger and larger quantities of CO₂ into the atmosphere as coal-burning and automobile power increases. China may exceed the United States in CO₂ emissions by the year 2020. Although there may still be an ongoing greenhouse effect debate, the increase in concentration of CO₂ in the earth's atmosphere is fairly well agreed upon by most scientists. The increase in CO_2 is demonstrated by the following concentrations.

<u>Time</u>	CO ₂ Concentrations		
Pre-Industrial*	270 ppm		
1958	315 ppm		
2000	360 ppm		

^{*} 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 CO₂ released to the atmosphere
- 3. Develop new technologically driven alternatives to CO₂ producing energy sources
- 4. Curtail cutting down forested areas around the globe
- 5. Conduct CO₂ sequestration of CO₂ 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 CO₂ 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 considerations and well-formed lifestyles will not lend to effectuation of 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 and subsequent conversion of CO to methanol as a usable fuel. 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 CO₂ to CO, which can then be catalytically converted to a useful fuel.

GOAL AND OBJECTIVES

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_2O) 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_3OH) , or other energy bearing compounds.

APPROACH

The successful demonstration of this invention is dependent on the establishment and optimization of at least 6 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 utilizing to advantage α-particle pathway dynamics;
- 5) understanding and enhancing of ion-pair formation per α -particle; and
- 6) precluding recombination of CO with other ions.

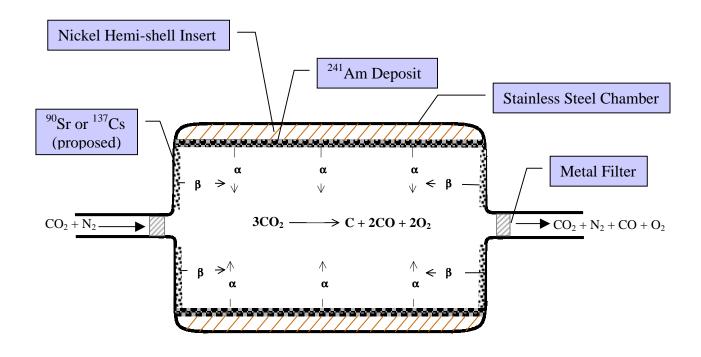
The separation of CO from CO_2 , O_2 , and an energy transference gas with subsequent reinjection into the irradiation chamber was considered to be beyond the scope of this experimental work. The conversion of CO to a usable fuel form was also considered to be beyond the scope of this study. However, 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.

PROJECT DESCRIPTION

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 ²³⁸Pu, ²³⁹Pu, ²⁴¹Am, ⁹⁰Sr, ¹³⁷Cs 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 ²⁴¹Am was deposited on the internal walls of the chamber as shown in Figure 1.



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\begin{array}{rcl} Length & = & 17 \text{ cm} \\ Diameter & = & 6 \text{ cm} \\ Ni Thickness & = & \sim 0.5 \text{ cm} \\ Lateral Surface Area (Internal) & = & 320.4 \text{ cm}^2 \\ Volume & = & 480 \text{ cm}^3 \\ & & & & \\ ^{241}\text{Am Deposited} & = & 97 \text{ mg} \\ & & & & \\ ^{241}\text{Am Activity} & = & 1.23 \times 10^{10} \text{ d/s/chamber} \end{array}
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Figure 1. Irradiation 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 Am decay is calculated to be \sim 4-5 cm in CO₂. See Appendix C for the calculation and Appendix B for the nuclear characteristics of 241 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 ²⁴¹Am with subsequent drying by heating. A preferable way to deposit or electroplate a uniform thin layer of ²⁴¹Am would be to electrodeposit 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 Am deposited on the inner surface of the heavy nickel hemi-shells gave a calculated 4π α -emission rate of 1.23×10^{10} α 's/sec/97 mg (as calculated in Appendix A). However, $\sim 50\%$ of the alpha-particles would be emitted into the gas phase and the remainder would be emitted into the walls of the nickel hemishells. So for a 2π geometry, the α -emission rate into the gas phase would be about 6.2×10^9 α 's/sec. The 97 mg of 241 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 $\sim 20\%$. This would give an effective alpha-particle emission rate of $\sim 5 \times 10^9$ α 's/sec/chamber. As stated earlier, a different and more efficient plating technique could replace the dried-salt technique and give an 241 Am α -emission rate of $\sim 2.5 \times 10^{11}$ α 's/sec/chamber.

Energy Transference Gas

When pure 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% CO_2 and 50% nitrogen because nitrogen was suspected of being a good energy transference gas. The initial conversion rate for CO_2 to CO increased to $\sim 2,000$ -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% CO₂ 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_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_2 . The overall generation rate with air over a 24-hour period was very poor.

Tests using pure CO_2 , $N_2 + CO_2$, $Ar + CO_2$ and other CO_2 -gas mixtures, when mixed with pure H_2 gas to give a 1-2 % mixture, immediately showed a very high concentration of H_2O in the gas space. This radiolytically formed water was quite detrimental to the GC used for making these measurements. However the reaction of H_2 in the presence of O_2 formed by the radiolytic decomposition of CO_2 to form H_2O , provided the basis for using this methodology to solve the problem of H_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_2 and Ar that did not contain O_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 238 Pu, 239 Pu, 241 Am, and 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 241 Am contains 5.486 MeV of energy and is relativistic near the point of emission. The diameter of the alpha particles are about 2×10^{-13} cm whereas the atom to atom bond distance between a carbon to oxygen atom is about 1.2×10^{-8} cm. Consequently, the possibility of the alpha particle colliding with another nucleus in a gaseous medium is highly unlikely as shown in Figure 2.

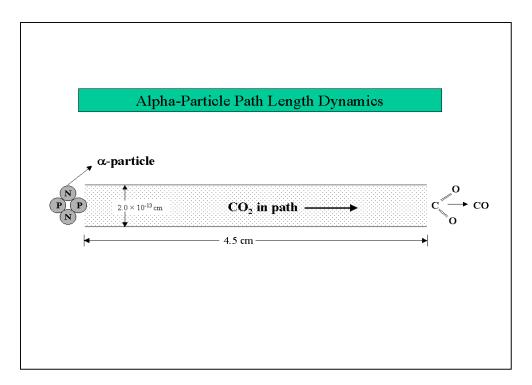


Figure 2. Diagram of Alpha-Particle Path Length Dynamics

However, the number of CO_2 atoms per cubic centimeter of gas is about 2.7×10^{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 6.6×10 -24 gm with an electron with a rest mass of 9.1×10 -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 Figure 3.

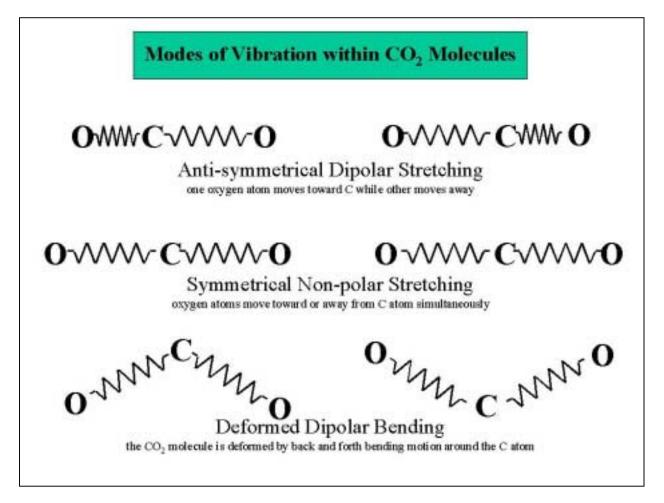


Figure 3. Modes of Vibration within CO₂ Molecules

The atoms within the CO_2 molecule vibrate with a linear and rotational velocity of about 4.4×10^4 cm/sec. An alpha particle from 241 Am will have an initial velocity of about 2×10^9 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 7.5×10^6 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 α or β -particle, is called a Bragg Curve or Bragg Ionization Curve. The average specific ionization of an alpha particle from ²⁴¹Am 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 Figure 4.

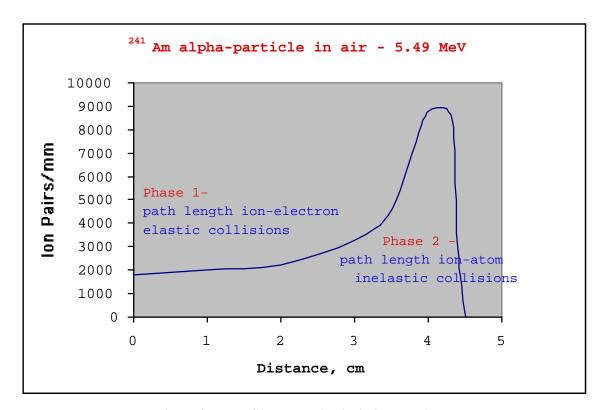


Figure 4. Bragg Curve, showing ioniation v.s. distance

The initial kinetic energy of the alpha particle from decay of the ²⁴¹Am 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 CO₂ of 4-5 cm and a Δ E/ Δ 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 CO₂, the specific ionization or number of ion pairs formed per cm of travel is calculated as follows:

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

Where:

5.486 MeV = Energy of ²⁴¹Am α-particle 34 eV/ion-pair = Average ionization energy for CO₂ (NBS Handbook 85, ICRU Report 106, 1962, Washington, DC) 4.5 cm = Approximate path length of 5.486 MeV α-particle in CO₂

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

$$(3.6 \times 10^4 \text{ ion pairs / cm})(4.5 \text{cm}) = 161,353 \text{ ion pairs}$$
 (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 =

$$\frac{1,000 \ \mu\ell/L/hr}{2.24 \times 10^{7} \ \mu\ell/mole} = 4.46 \times 10^{-5} \ mole/L/hr$$
 (Eq. 3)

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

$$\frac{2.68 \times 10^{19} \text{ molecules/L/hr}}{(4.92 \times 10^9 \text{ a/s/sec}) (3,600 \text{ sec/hr})} = 1.5 \times 10^6 \text{ molecules/L/}\alpha - particle}$$
(Eq. 5)

At 5,000 ppm/hr the number increases to 7.56×10^6 molecules/L/ α -particle.

The empirically determined number of 7.56×10^6 molecules/L/ α -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:

$$3CO_2 \xrightarrow{\alpha-particles} C + 2CO + 2O_2$$
 (Eq. 6)

However, many competing reactions can occur to lessen the yield of the primary reaction.

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

$$2CO \xrightarrow{\alpha - activity} 2C + O_2 \tag{Eq. 8}$$

An increasing concentration of O_2 appears to enhance reaction 2 and 3. Although the alphaparticle 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 CO are 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 CO.

The ratio of CO₂ 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 ⁹⁰Sr or ¹³⁷Cs 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 H₂ and O₂ and are also expected to be effective for dissociation of CO₂ 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:

 $\begin{array}{rcl} \text{Diameter} & = & 8 \text{ cm} \\ \text{Lateral Area } (2\pi \text{rh}) & = & 5,026 \text{ cm}^2 \\ \text{Volume } (\pi \text{r}^2 \text{h}) & = & 10.05 \text{ L} \\ & ^{241}\text{Am Mass (per cm}^2) & = & 20 \text{ mg/cm}^2 \\ & ^{241}\text{Am Mass (total)} & = & 100.5 \text{ gm} \\ & ^{241}\text{Am Activity (per cm}^2) & = & 2.54 \times 10^9 \text{ d/s/cm}^2 \\ & ^{241}\text{Am Activity (total)} & = & 1.27 \times 10^{13} \text{ d/s/chamber} \\ & = & 344 \text{ Ci/chamber} \end{array}$

Flow Rate = 10 L/min

 CO_2 to CO Conversion Rate = 5,000 ppm/min

Length

= 5,000 μl/L/min = 50,000 μl/chamber/min

2m = 200 cm

@ $2.24 \times 10^7 \,\mu$ l/mol = $2.23 \times 10^{-3} \,\text{mol/chamber/min}$

= 3.2 mol of CO per day

1 mol CO yields 1 mole CH₃OH

Therefore = $3.2 \text{ mol of CH}_3\text{OH per day/chamber}$

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

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.

FUTURE ACTIVITIES - ENHANCEMENT OF EFFICIENCIES

The conversion of 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 α -particle or from direct interaction of the α -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 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 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 CO. The highest efficiencies for conversion of 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 CO₂ matrix. This instantaneous separation of CO from CO₂ would make the CO unavailable for continued bombardment by alpha particles because of the difference in physical properties between CO₂ and CO as shown in Table I. Physical Properties of CO2 and CO.

Table I. Physical Properties of CO₂ and CO.

		Density @			Solubility in H ₂ O
Gas	MW	0 °C	MP	BP	@ 0 °C
CO_2	44.01	1.977 g/L	-56.6 ^O C	-28.5 °C	171.3 gm/100 cm ³
CO	28.01	1.250 g/L	-199.0 ^o C	-191.5 ^O C	$3.5 \text{ gm}/100 \text{ cm}^3$

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_2 . 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_2 .

APPLICATION AND BENEFITS

This new technology could be applied to technological processes that result in effluents with relatively high concentrations of CO₂ such as coal and natural gas fired power plants. Improved efficiency of the conversion process could lead to conversion of less concentrated gas streams that emit CO₂.

The overall benefit of this new technology is that CO_2 , an unwanted and harmful greenhouse gas, can be economically converted to a useful product with an essentially passive radionuclide system that can continue for extended lengths of time (\sim 50-100 yr) without having to be regenerated. This new technology would complement sequestion rather than compete with that technology but would have the advantage of converting CO_2 to a useful and much needed fuel form rather than emitting it to the atmosphere or solidifying for disposal.

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_2O 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 (²³⁸Pu, ²³⁹Pu, ²⁴¹Am) and fission products such as ⁹⁰Sr and ¹³⁷Cs. 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 ACalculation of Alpha-Particle Emission Rate

²⁴¹Am Nuclear Properties:

$$t \frac{1}{2} = 432.7 \ annums$$

$$\lambda = \frac{0.693}{(432.7 \ a) \ (365.25 \frac{d}{a}) \ (1,440 \frac{min}{d}) \ (60 \frac{sec}{min})}$$

$$= 5.075 \times 10^{-11} \ sec^{-1}$$

$$\frac{dn}{dt} = N\lambda$$

$$\frac{dn}{dt} = \frac{(1 \ gm) \ (6.02 \times 10^{23} \frac{atoms}{gm-atom}) \ (5.075 \times 10^{-11} sec^{-1})}{241 \frac{gm}{gm-atom}}$$

$$= 1.268 \times 10^{11} \ \alpha' \ s / sec / gm$$

$$^{241} Am$$
 plated on walls of chamber is ~ 97 mg.
$$= 1.23 \times 10^{10} \ \alpha' s/sec/97 \ mg$$

Assume 2π alpha–emission into gas phase = 6.15×10^9 d/sec into gas phase

Assume 20% of α are absorbed by salt deposit, then = 4.92×10^9 α 's/sec into gas phase

Assume 5,000 mg is plated on inner walls of Ni hemi-shell Then activity = 2.54×10^{11} α 's/sec/5 gm @ 2π

Appendix B Nuclear Characteristics of ²⁴¹Am

Half-Life = 432.7 Y

 $\lambda = 5.075 \times 10^{-11} \text{ sec}^{-1}$

Specific Activity = $1.268 \times 10^{11} \text{ d/s/g}$

 $\alpha_1 = 5.486 \text{ MeV } (86.0\%)$ $\alpha_2 = 5.443 \text{ MeV } (12.7\%)$

 $\alpha_3 = 5.389 \text{ MeV } (1.3\%)$

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 ΔE/Δx and Range of Alpha-particle in CO₂

$$-\Delta E/\Delta x = [(4\pi e^4 Z_1^2 Z_2 N)/(m_o v^2)] \ ln \ [2m_o v^2/I]$$
 Where:
$$-\Delta E/\Delta x = \text{change in kinetic energy of charged particle in ergs/cm}$$

$$e = \text{charge on electron } (4.8 \times 10^{-10} \text{ stat c})$$

$$Z_1 = \text{charge of } \alpha \text{ particle } (2)$$

$$Z_2 = \text{atomic number of interacting atoms per cm}^3 = 2.71 \times 10^{19} \text{ molecules/ cm}^3 \text{ (STP)}$$

$$m_o = \text{rest mass of electron } (9.1083 \times 10^{-28} \text{ g})$$

$$v = \text{velocity of } \alpha \text{-particle } \text{ (cm/sec)} = 1.63 \times 10^9 \text{ cm/sec}$$

$$I = \text{average excitation potential of atom } (\approx 80 \text{ eV})$$

$$m = \text{mass of an } \alpha \text{-particle} = 6.64 \times 10^{-24} \text{ g}$$

$$E = \text{energy of an } \alpha \text{-particle} = 5.49 \text{ MeV}$$

$$\text{KE } (\alpha \text{-particle}) = (1/2) \text{mv}^2$$

$$v^2 = 2 \text{ (KE/m}$$

$$= 2(5.49 \text{MeV})/(1.6 \times 10^{-6} \text{ erg/MeV})/(6.64 \times 10^{-24} \text{ g})$$

$$= 2.65 \times 10^{18} \text{ cm}^2/\text{s}^2$$

$$v = (2.65 \times 10^{18} \text{ cm}^2/\text{s}^2)^{1/2}$$

$$= 1.63 \times 10^9 \text{ cm/s}$$

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$$= 1.63 \times 10^9 \text{ cm/s}$$

$$ln \ [2m_o v^2/I] = ln \ \{2[(9.1083 \times 10^{-28})(2.65 \times 10^{18})]/[80(1.6 \times 10^{-12})]\}$$

$$= ln \ (37.71)$$

$$= 3.63$$

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

$$= 2.71 \times 10^{19} \text{ molecules/cm}^3 \text{ for CO}_2$$

$$-\Delta E/\Delta x = [(4\pi e^4 Z_1^2 Z_2 N)/(m_o v^2)] \ln [2m_o v^2/I]$$

$$= [4\pi (5.3 \times 10^{-38})(4)(16)(2.71 \times 10^{19})/(9.108 \times 10^{-28})(2.65 \times 10^{18})] \ln 37.71$$

$$= [4\pi (5.3 \times 10^{-38})(4)(16)(2.71 \times 10^{19})/(2.41 \times 10^{-9})] \ (3.63)$$

$$= 1.74 \times 10^{-6} \text{ erg/cm}/(1.6 \times 10^{-6} \text{ erg/MeV})$$

$$= 1.09 \text{ MeV/cm; equivalent to } (5.49/1.09) = 5.03 \text{ cm}$$

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 α -particle = 4-5 cm due to straggling