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An Evaluation of Options Relative to the Fixation and Disposal of ^{14}C -Contaminated CO_2 as CaCO_3

Allen G. Croff

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CHEMICAL TECHNOLOGY DIVISION

AN EVALUATION OF OPTIONS
RELATIVE TO THE FIXATION AND DISPOSAL OF
 ^{14}C -CONTAMINATED CO_2 AS CaCO_3

Allen G. Croff

APRIL 1976

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Contents		<u>Page</u>
ABSTRACT.		1
1. SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS		1
1.1 Summary.		1
1.2 Conclusions.		3
1.3 Recommendations.		4
2. INTRODUCTION.		5
2.1 Description of the Problem		5
2.2 Background		6
2.3 Scope and Assumptions.		7
3. PROCESSES FOR THE PRODUCTION OF CaCO_3 FROM CO_2		9
3.1 Direct Reaction of CO_2 with an Aqueous Slaked Lime Slurry		9
3.1.1 Description of the direct fixation process.		9
3.1.2 Direct fixation process flow rates.		12
3.1.3 Discussion of the direct fixation process		12
3.2 Double Alkali CO_2 Fixation Process		14
3.2.1 Description of the double alkali fixation process		14
3.2.2 Double alkali process flow rates.		14
3.2.3 Discussion of the double alkali fixation process.		16
3.3 Comparative Evaluation of the Direct and Double Alkali CO_2 Fixation Processes		17
3.3.1 Availability of design data		17
3.3.2 Complexity.		18
3.3.3 Solids.		18
3.3.4 Corrosion		18
3.3.5 Economics		18
3.3.6 Process recommendation.		19
4. CONCRETION AND PACKAGING OF THE CaCO_3 PRODUCT		19
4.1 Concretion		19
4.2 Packaging.		20

	<u>Page</u>
5. TRANSPORTATION OF THE CaCO ₃ PRODUCT.	21
5.1 Characterization of the CaCO ₃ Product Being Transported . .	21
5.2 Categorization of the CaCO ₃ Product	23
5.2.1 Department of Transportation	23
5.2.2 IAEA regulations	25
5.2.3 Summary of transportation regulations.	27
5.3 Radiation Doses During Transportation and Handling.	27
5.4 Transportation Costs.	28
6. DISPOSAL OF THE CaCO ₃ PRODUCT.	31
6.1 Introduction.	31
6.2 Shallow-Land Burial Options	32
6.2.1 Burial of unpackaged, unconcreted CaCO ₃ product. . .	34
6.2.2 Burial of packaged, unconcreted CaCO ₃ product. . . .	34
6.2.3 Burial of unpackaged, concreted CaCO ₃ product. . . .	35
6.2.4 Burial of packaged, concreted CaCO ₃ product.	36
6.3 Partial Block Burning and Concretion.	36
6.4 Disposal of CaCO ₃ by Hydraulic Fracturing	37
6.5 Deep-Sea Disposal of the CaCO ₃ Product.	39
6.6 Geologic Repository Emplacement of the CaCO ₃ Product. . . .	40
6.7 Summary of the Status of the Options Available for Disposal of the CaCO ₃ Product.	40
7. LOCATION OF THE CO ₂ FIXATION SYSTEM WITH RESPECT TO THE KALC PROCESS.	42
7.1 Elimination of KALC	42
7.2 Size Reduction of the KALC Process.	44
8. ECONOMICS.	45
8.1 Cost of Fixing CO ₂ as CaCO ₃	45
8.1.1 Capital cost	45
8.1.2 Total CO ₂ fixation cost.	45
8.2 Total Cost of Producing and Disposing of ¹⁴ C-Contaminated CaCO ₃	45
9. REFERENCES	54

	Tables	<u>Page</u>
2.1	^{14}C activities, masses, and concentrations in HTGR fuel as a function of nitrogen concentration	10
5.1	Radioactivity of CaCO_3 product	22
5.2	IAEA definition of low specific activity material.	26
5.3	Dose rate at the surface of a 55-gal drum containing CaCO_3 product with ^{14}C and ^{85}Kr	29
5.4	Transportation costs from Oak Ridge to various destinations.	30
6.1	Comparison of the radiotoxicity of ^{14}C with that of other nuclides	41
8.1	Estimated capital cost of fixing CO_2 as CaCO_3	46
8.2	Estimated total cost of fixing CO_2 as CaCO_3 using the direct fixation process	47
8.3	Overall cost of producing and disposing of ^{14}C -contaminated CaCO_3	50
8.4	Bases for Table 8.3.	51
8.5	Summary of case definitions for Table 8.3.	52

	Figures	<u>Page</u>
2.1	Schematic of HTGR fuel reprocessing plant considered.	8
3.1	Schematic of the direct CO ₂ fixation process.	11
3.2	Schematic of the double alkali CO ₂ fixation process	15

AN EVALUATION OF OPTIONS RELATIVE TO THE FIXATION
AND DISPOSAL OF ^{14}C -CONTAMINATED CO_2 AS CaCO_3

Allen G. Croff

ABSTRACT

A paper study was conducted to determine the best method for fixing the ^{14}C -contaminated CO_2 resulting from an HTGR fuel block burner as CaCO_3 , and to determine the best methods for disposing of the CaCO_3 thus produced. The fixation method selected was the direct reaction of a $\text{Ca}(\text{OH})_2$ slurry with the CO_2 . The least expensive disposal options which are likely to be acceptable appear to be the shallow-land burial of either drummed CaCO_3 solid (total cost = \$18.47/kg heavy metal) or drummed CaCO_3 concreted with cement (total cost = \$43.33/kg heavy metal). Neither placing the CO_2 fixation process before the Kr removal process nor separating the bulk of the graphite fuel block from the fuel particles is attractive on both technical and economic grounds. However, reduction of the HTGR fuel nitrogen content appears to be a more attractive method of reducing the ^{14}C release rate.

1. SUMMARY, CONCLUSIONS, AND RECOMMENDATIONS

1.1 Summary

A scoping study has been conducted to determine the best method for the fixation of ^{14}C -contaminated CO_2 as CaCO_3 and to evaluate the various options available for disposing of the CaCO_3 thus produced. The fixation and disposal options were evaluated on the bases of technical merit, economics, and regulatory acceptability.

The two CO_2 fixation processes considered were:

1. A direct process, wherein the CO_2 is reacted directly with a slaked lime $[\text{Ca}(\text{OH})_2]$ slurry to form a CaCO_3 slurry.
2. A double alkali process, wherein the CO_2 is reacted with a NaOH solution to form Na_2CO_3 ; the Na_2CO_3 is subsequently

reacted with a slaked lime slurry to produce the CaCO_3 product and to regenerate the NaOH solution.

The evaluation of these two processes was based on the following criteria:

- (1) design data availability,
- (2) process complexity,
- (3) solids handling,
- (4) corrosiveness of chemicals involved in the process,
- (5) economics.

The CaCO_3 disposal options considered were as follows:

1. Shallow-land burial of
 - (a) unpackaged, unconcreted CaCO_3 ,
 - (b) packaged, unconcreted CaCO_3 ,
 - (c) unpackaged, concreted CaCO_3 ,
 - (d) packaged, concreted CaCO_3 .
2. Hydraulic fracturing (mixing a CaCO_3 slurry with cement and injecting it into deep geological strata).
3. Deep-sea disposal (concretion and dumping in the deep sea).
4. Partial block burning.
5. Emplacement in a geologic repository for material contaminated with alpha emitters.

The CaCO_3 packaging considered in this study was standard 55-gal drums. The concretion method considered was to combine the CaCO_3 with appropriate amounts of water and cement to form a monolithic solid material. Partial block burning involves either drilling or pushing the graphite matrix rods containing the heavy metal (U and Th) out of the graphite fuel block and then subjecting only the graphite matrix-fuel portion to the normal HTGR reprocessing sequence. The remaining graphite block is disposed of intact, along with the reduced amount of CaCO_3 produced from the burning of the graphite matrix. The waste disposal options were also evaluated on the bases of anticipated regulatory acceptability, economics, and technical merit.

The feasibility of placing the CO₂ fixation system before the Kr removal system to reduce or eliminate the gas volume which the Kr removal system must handle was also investigated.

1.2 Conclusions

The principal conclusions of this study are as follows:

1. A major reduction in the ¹⁴C dose rate can probably be more easily and less expensively achieved by reducing the nitrogen content of the HTGR fuel element rather than fixing the ¹⁴C-contaminated CO₂ as CaCO₃.
2. The disposition of Kr in the CO₂ fixation process is of paramount importance. The relatively short half-life of ⁸⁵Kr (10.73 years) and its relatively high-energy decay particles [β^- , 0.687 MeV (max); γ , 0.514 MeV] as compared to ¹⁴C result in the ⁸⁵Kr being the controlling radiation hazard in all phases of the ¹⁴C-contaminated CO₂ fixation and disposal processes. Krypton's lack of chemical reactivity under normal conditions makes prediction of its behavior virtually impossible without detailed experimental work.
3. The direct CO₂ fixation process appears to be superior to the double alkali process because of decreased complexity, reduced corrosiveness of the chemical reagents involved, and lower cost. The two processes are equivalent with respect to solids handling. Design data availability favors the double alkali process.
4. Shallow-land burial of the CaCO₃ appears to be the best disposal option available. The burial of unpackaged, unconcreted CaCO₃ [Option 1(a), Sect. 1.1] will probably not be acceptable. The future acceptability of burying packaged, unconcreted CaCO₃ [Option 1(b)] or unpackaged, concreted CaCO₃ [Option 1(c)] is not clear at the present time. The burial of packaged, concreted CaCO₃ [Option 1(d)] will probably be acceptable in the future. Thus, the possibly acceptable shallow-land burial options, in decreasing order of economic preference, are:

- (a) Burial of packaged, unconcreted CaCO_3 (\$18.47/kg heavy metal)
 - (b) Burial of unpackaged, concreted CaCO_3 (\$29.11/kg heavy metal)
 - (c) Burial of packaged, concreted CaCO_3 (\$43.33/kg heavy metal when concreted at the burial site; \$55.54/kg heavy metal when concreted at the reprocessing plant).
5. The disposal of CaCO_3 via hydraulic fracturing does not appear to be attractive based on a combination of economic (\$36.78/kg heavy metal), regulatory, and technical grounds.
 6. Deep-sea dumping of the CaCO_3 does not appear to be attractive on both economic (\$56.43/kg heavy metal) and regulatory acceptance grounds.
 7. Partial block burning is unattractive because the maximum volume reduction is only about 50% and the fission products sorbed on the unreprocessed blocks elevate the waste from the low-level category to at least the intermediate level.
 8. The geologic-repository burial of the CaCO_3 is economically unattractive (\$281.39/kg heavy metal) and technically unjustifiable in view of the relatively low toxicity of ^{14}C .
 9. Process complexity and unfavorable economics indicate that it would not be advantageous to place the CO_2 fixation process before the Kr removal process in the off-gas cleanup system.

1.3 Recommendations

If ^{14}C -contaminated CO_2 is to be fixed and disposed of as CaCO_3 , the following developmental requirements must be met:

1. The behavior of Kr in the CO_2 fixation process must be determined. Even when the fixation process follows the Kr removal process, the activity of the Kr is controlling in all cases examined. Thus, the amount of Kr present will have a major impact on shielding requirements, the method used to transport the CaCO_3 , the CaCO_3 disposal methods, and the economics of the overall process.
2. An immediate effort is required to better define the allowable methods for transportation and disposal of the CaCO_3 . Of particular

concern is whether the CaCO_3 must be concreted to a monolithic form before disposal.

3. The reaction rate of CO_2 with a Ca(OH)_2 slurry must be obtained or measured as a function of time, temperature, and reactant concentration. In the case of the double alkali process, the causticization reaction needs further investigation. The effect of various procedures for slaking lime must be investigated for either the direct or double alkali process.
4. Although continuous vacuum filters and spray dryers are used commercially, an investigation into various dewatering methods and the resulting product characteristics is required.
5. In view of the cost and logistical complexity of producing and disposing of ^{14}C -contaminated CO_2 , the following recommendations are made:
 - (a) A major effort should be made to determine whether or not ^{14}C is, in fact, a significant radiological hazard.
 - (b) If ^{14}C is a significant hazard, a major effort should be made to determine whether a reduction in the nitrogen concentration in the graphite will reduce the ^{14}C hazard to acceptable levels.
 - (c) If the ^{14}C cannot be released, the possibility of isotopic separation should be investigated.

2. INTRODUCTION

2.1 Description of the Problem

The principal objectives of this project were to investigate:

- (1) potential processes for the fixation of the ^{14}C -contaminated CO_2 stream from an HTGR fuel block burner as CaCO_3 ;
- (2) various options for packaging, concretion, transportation, and disposal of the CaCO_3 ;
- (3) the feasibility of placing the CO_2 fixation process before the Kr removal process to eliminate or reduce the size of the Kr removal process, and

- (4) the feasibility of separating the fuel particles from the graphite blocks by burning off the mechanically separated graphite matrix, with subsequent disposal of the unburned graphite blocks.

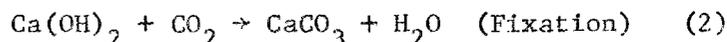
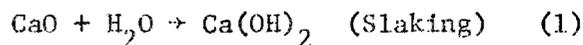
These investigations were to include such matters as technical feasibility, regulatory acceptability, developmental requirements, and economics. The results of these investigations were to be recommended processes for the fixation and disposal of ^{14}C -contaminated CO_2 as CaCO_3 , along with an outline of the major research and development work required to implement these recommended CO_2 fixation and disposal processes.

2.2 Background

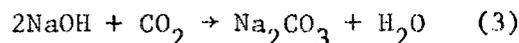
The basic hypothesis of this investigation was that the stream of ^{14}C -contaminated CO_2 resulting from the burning of HTGR graphite fuel blocks cannot be released to the environment. This investigation in no way addresses more basic questions such as whether the production of ^{14}C can be reduced, what fraction of the ^{14}C is released during burning, and whether the ^{14}C can, in fact, be released to the environment. It was also assumed that the CO_2 would be fixed as CaCO_3 , with no effort being undertaken to determine if a more favorable fixation medium exists. Since the CaCO_3 fixation process uses an inexpensive reagent (lime), is thermodynamically favorable ($\Delta H \cong -25$ kcal/g-mole), and results in a stable solid (CaCO_3 decomposes at temperatures $> 1500^\circ\text{F}$), it would appear that the probability of finding a better fixation process without a major research and development effort is rather remote.

The direct reaction of CO_2 with solid CaO to form CaCO_3 was rejected because (1) the rate of reaction is very slow in the absence of H_2O and (2) the CaCO_3 layer formed on the outside of the CaO particle reduces the rate at which the CO_2 diffuses to the unreacted CaO core, further lowering the reaction rate.

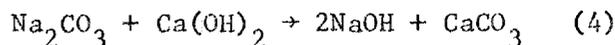
Rejection of the dry CO_2 - CaO process for CO_2 fixation leads to the consideration of two aqueous processes. The first of these (the direct process) involves the direct reaction of the CO_2 with a slurry of slaked lime [$\text{Ca}(\text{OH})_2$] according to the following reactions:



A second aqueous process (the double alkali process) involves the reaction of CO_2 with NaOH to form Na_2CO_3 :



The Na_2CO_3 is then causticized with the slaked lime produced in reaction (1):



Either KOH or NH_4OH can be substituted for the NaOH in this process.

The direct process is used commercially to produce whiting (or precipitated chalk), a very pure form of CaCO_3 having specific physical characteristics which make it useful as a paint pigment. A process analogous to the double alkali process is used to scrub SO_2 from power plant stack gas. The resulting product in the case of a stack gas scrubber is a CaSO_3 - CaSO_4 mixture. The double alkali process could also be used to produce whiting, although the washing required as a result of the NaOH present has made this application less attractive than other whiting manufacturing processes.

2.3 Scope and Assumptions

The HTGR fuel reprocessing plant under consideration was assumed to reprocess 36 fuel elements per day for 300 days per year, approximately the size of a demonstration-type facility. This results in the burning of 3600.0 kg of carbon per day, yielding 13,200.0 kg of CO_2 per day. Two cases were also examined wherein the fuel sticks were assumed to be removed from the graphite block by drilling or by simply pushing them out, resulting in only part of the fuel element being burned.

For the purposes of this study, the reprocessing plant was assumed to consist of the fuel element burners, the pre-KALC off-gas treatment systems, the KALC process for removing Kr , and the CO_2 fixation process. The reference case, shown in Fig. 2.1, has the KALC process preceding the CO_2 fixation

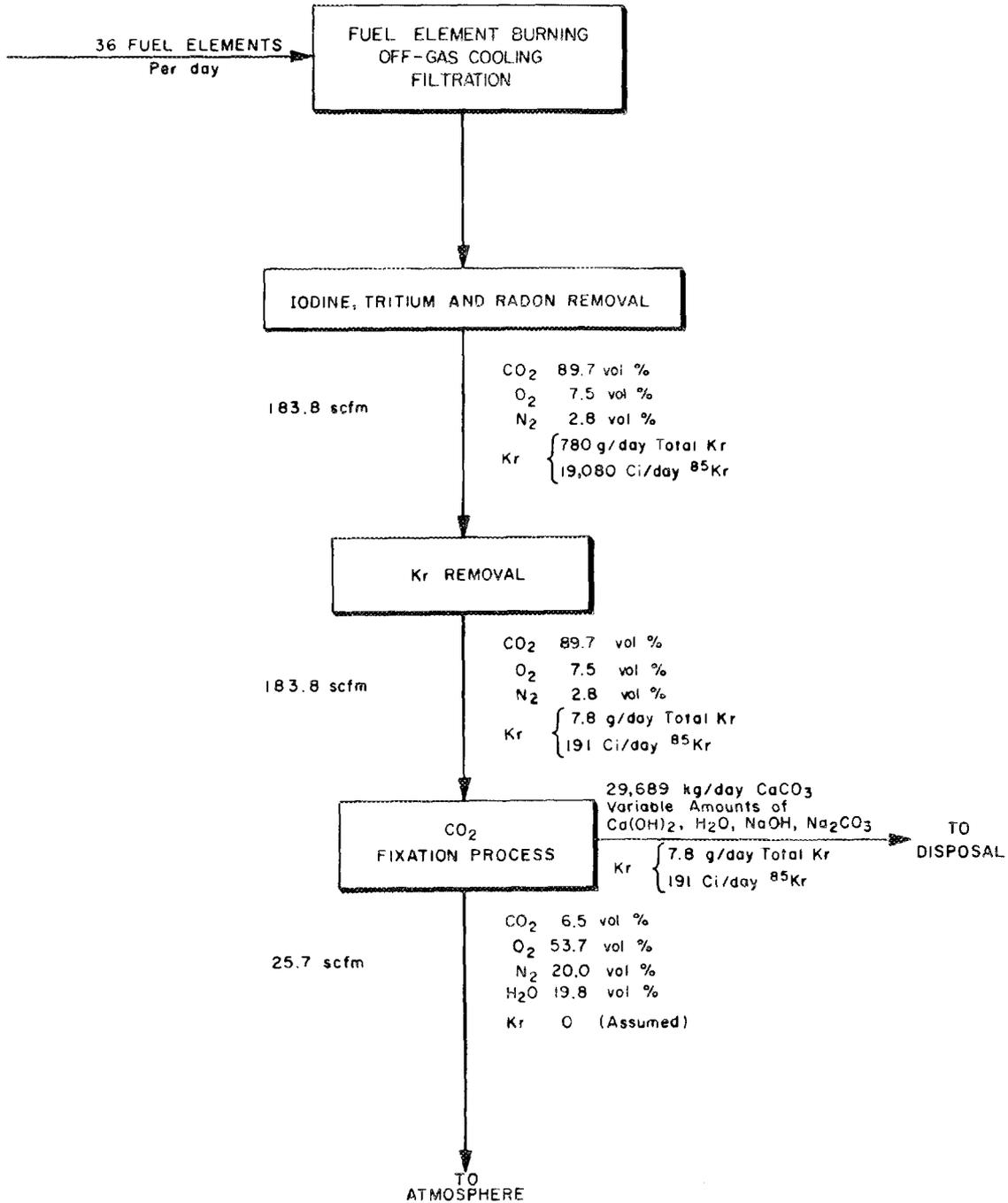


Fig. 2.1. Schematic of HTGR fuel reprocessing plant considered.

process. An alternate case was also examined wherein the CO₂ fixation process was placed before the KALC process.

The principal mode of ¹⁴C production in HTGRs is the ¹⁴N(n,p)¹⁴C reaction. Since the degree to which nitrogen will be present in the fuel is not yet well known, nitrogen impurity levels of 0, 26 (expected case), and 100 ppm were considered. The ¹⁴C content of the off-gas stream is given in Table 2.1 for these three impurity levels based on data given in ref. 1. The ¹⁴C production in the 0-ppm ¹⁴N case is principally a result of the ¹³C(n,γ) reaction, which also accounts for 19.3% of the ¹⁴C production in the 26-ppm ¹⁴N case.

One of the principal assumptions made in this study was that all of the Kr which enters the CO₂ fixation process exits combined (physically) with the CaCO₃ product. This assumption results in the "worst case" with respect to CaCO₃ concretion, transportation, and disposal since the radioactivity of the CaCO₃ product is maximized. However, it is to be emphasized that this assumption is totally speculative, since there are no data available concerning the disposition of Kr in this type of a system. Because this assumption regarding Kr is controlling in most parts of the CO₂ fixation and disposal processes, the reader should be aware that the conclusions of this report, particularly with respect to transportation, may be drastically altered if the major portion of the Kr is not, in fact, entrained with the CaCO₃ product.

3. PROCESSES FOR THE PRODUCTION OF CaCO₃ FROM CO₂

3.1 Direct Reaction of CO₂ with an Aqueous Slaked Lime Slurry

3.1.1 Description of the direct fixation process

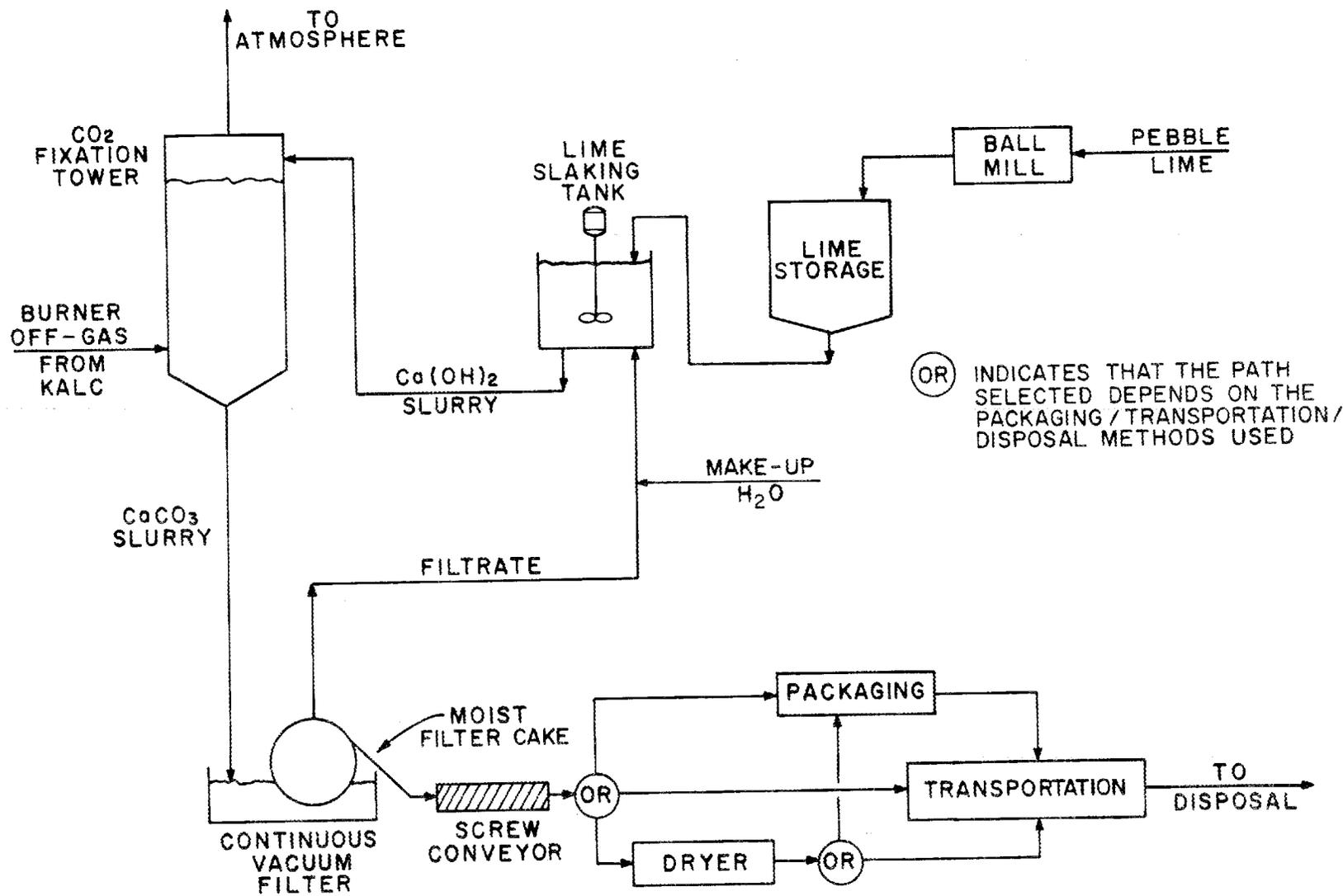
A schematic of the direct CO₂ fixation process to produce CaCO₃ is shown in Fig. 3.1. Pebble lime (CaO) is pulverized in a ball mill and then moved to storage. The lime is slaked (hydrated) as needed in the lime slaking tank, according to reaction (1) [Sect. 2.2]. The slaked lime thus produced is relatively insoluble, with about 0.185 g of Ca(OH)₂ dissolving in 100 g of water at 0°C. The solubility decreases to 0.077

Table 2.1. ^{14}C activities, masses, and concentrations in HTGR fuel as a function of nitrogen concentration^a

^{14}N (ppm)	Levels of ^{14}C	
	Per fuel element	Per day ^b
0	0.030 Ci	1.08 Ci
	0.007 g	0.25 g
	69.2 ppb	69.2 ppb
26	0.16 Ci	5.76 Ci
	0.036 g	1.30 g
	359 ppb	359 ppb
100	0.53 Ci	19.1 Ci
	0.12 g	4.32 g
	1212 ppb	1212 ppb

^a ^{14}C production via $^{13}\text{C}(n,\gamma)$ is included in these values.

^b Assuming 36 fuel blocks per day.



11

Fig. 3.1. Schematic of the direct CO₂ fixation process.

g per 100 g of H_2O at $100^\circ C$. The slaked lime slurry, which is about 12 wt % $Ca(OH)_2$, is then pumped to the fixation tower, where it is contacted by CO_2 being bubbled through a sparger at the bottom of the tower. The tower was assumed to have no plates or packing. The slurry resulting from the CO_2 fixation contains about 15 wt % $CaCO_3$ which is produced according to reaction (2). The $CaCO_3$ is also quite insoluble, with only 0.0015 g dissolving in 100 g H_2O at $25^\circ C$. The aqueous stream contains about 0.6 wt % $Ca(OH)_2$, resulting from the assumed 5 mole % excess of slaked lime over that which is theoretically required. The slurry is then filtered on a continuous filter, with the filtrate being used for slaking. The filter cake is transported by screw conveyor either to the packaging system, the transport system, or a dryer, depending on the shipping and disposal methods used.

3.1.2 Direct fixation process flow rates

This process is assumed to remove 99% of the CO_2 initially present. The flow rates and compositions of the gas stream entering and leaving the fixation tower are summarized in Fig. 2.1. The slurry flow rate through the tower is about 31.5 gpm. Lime is required at a rate of 17,457 kg/day.

The filter cake will contain about 60 wt % solids and 40 wt % water, assuming that a relatively large $CaCO_3$ particle size is attained. The product rate is 30,787 kg of solids [$CaCO_3 + Ca(OH)_2$] per day (855 kg of solids per fuel element) along with 20,524 kg of H_2O per day (570 kg of H_2O per fuel element). The water, of course, may or may not be with the solids, depending on whether or not the filter cake is dried. This water loss, plus that evaporated in the tower, results in 20,791 kg of H_2O being required as make-up each day.

3.1.3 Discussion of the direct fixation process

The direct process uses relatively well-known technology and is used industrially to produce $CaCO_3$.²⁻⁶ However, the industrial process uses large excesses of CO_2 since complete conversion of the $Ca(OH)_2$ to $CaCO_3$ is required. The industrial $CaCO_3$ manufacturing facilities typically

have these large excesses of CO_2 available from lime kilns ($\text{CaCO}_3 \xrightarrow{\text{heat}} \text{CaO} + \text{CO}_2$). Since batch processes are used and since the endpoint of reaction (2) can be determined by a simple pH measurement, very little mass transfer and reaction rate data were required in the past. Hoog,² of the Mississippi Lime Company, stated that they had no mass transfer data that related to the direct carbonation of slaked lime. Lepley,³ of Technical Marketing, has mass transfer results, but considers them proprietary. The data from ref. 3 could most likely be obtained by approaching upper echelon management personnel and offering safeguards to prevent release of the data. The technical literature has also been searched, but data do not appear to be available.

As was stated previously, a large CaCO_3 particle size is desirable because it allows for a higher solids density in the product.² A large CaCO_3 particle size is favored by:^{2,3} (1) a high Ca(OH)_2 concentration, (2) a high reaction temperature, and (3) seeding the fixation tower. These conditions are also desirable because they tend to reduce the aqueous volumes that must be pumped and to increase the rate at which the carbonation reaction proceeds.

The flocculence of the solids in the slurry may mean that agitation by the CO_2 bubbles in the fixation tower will cause the tower to behave like a completely mixed stirred reactor rather than a countercurrent tower with no axial mixing. If this occurs, several completely-mixed stirred reactors with the gas stream flowing countercurrent to the slurry would probably be more effective.

Conversations with Hoog² and Lepley³ indicate that, in industrial practice, the CaCO_3 production processes (both direct and double alkali) tend to lose a significant portion of the product. The principal reasons for this are leaky pump seals, pipes, tanks, etc. Since the CO_2 fixation system under consideration here will involve radioactive materials, careful attention must be given to quality assurance and maintenance.

Each of the operations involved in the direct CO_2 fixation process, shown in Fig. 3.1, have been and are being practiced commercially. Most of the commercial processes are batch CO_2 fixation because of the desire

to produce pure CaCO_3 and the availability of excess CO_2 . However, given data on the CO_2 absorption and reaction rates with the slaked lime slurry, the design of a continuous process should be relatively straightforward.

3.2 Double Alkali CO_2 Fixation Process

3.2.1 Description of the double alkali fixation process

A schematic diagram of the double alkali process is given in Fig. 3.2. As in the direct fixation process, the pebble lime is pulverized in a ball mill and stored. When needed, the lime is slaked with the filtrate from the filter, according to reaction (1). However, in the double alkali flow-sheet, the filtrate contains approximately 4 N NaOH and 0.2 N Na_2CO_3 along with small amounts of Ca(OH)_2 and CaCO_3 . At the same time, the gas stream entering the packed fixation tower is being contacted with 4 N NaOH to form approximately 3.2 N Na_2CO_3 according to reaction (3). The NaOH concentration exiting the tower is about 1 N. The 3.2 N Na_2CO_3 solution is then combined with the slaked lime slurry in the causticization tank to form the CaCO_3 , and to regenerate the 4 N NaOH. This CaCO_3 slurry is subsequently filtered, and the filter cake washed. A portion of the filtrate is returned to the lime slaking tank. The remainder of the filtrate is augmented by make-up water and NaOH, and returns to the CO_2 fixation tower. The filter cake is moved by screw conveyor to the dry-package-transport system.

3.2.2 Double alkali process flow rates

The gas flow rates are as described in Fig. 2.1. Lime (CaO) is required at a rate of 17,457 kg/day, as in the direct fixation process. The flow rate of liquid into the lime slaking reactor is about 9 gpm and the flow rate of liquid through the CO_2 absorption tower is about 36 gpm, giving a liquid flow rate of about 45 gpm out of the causticization reactor. The wash water flow rate is about 3.45 gpm. The make-up water flow rate is about 0.38 gpm, and the NaOH make-up rate is estimated to be 1,873 kg/day. The filter cake contains approximately the following:

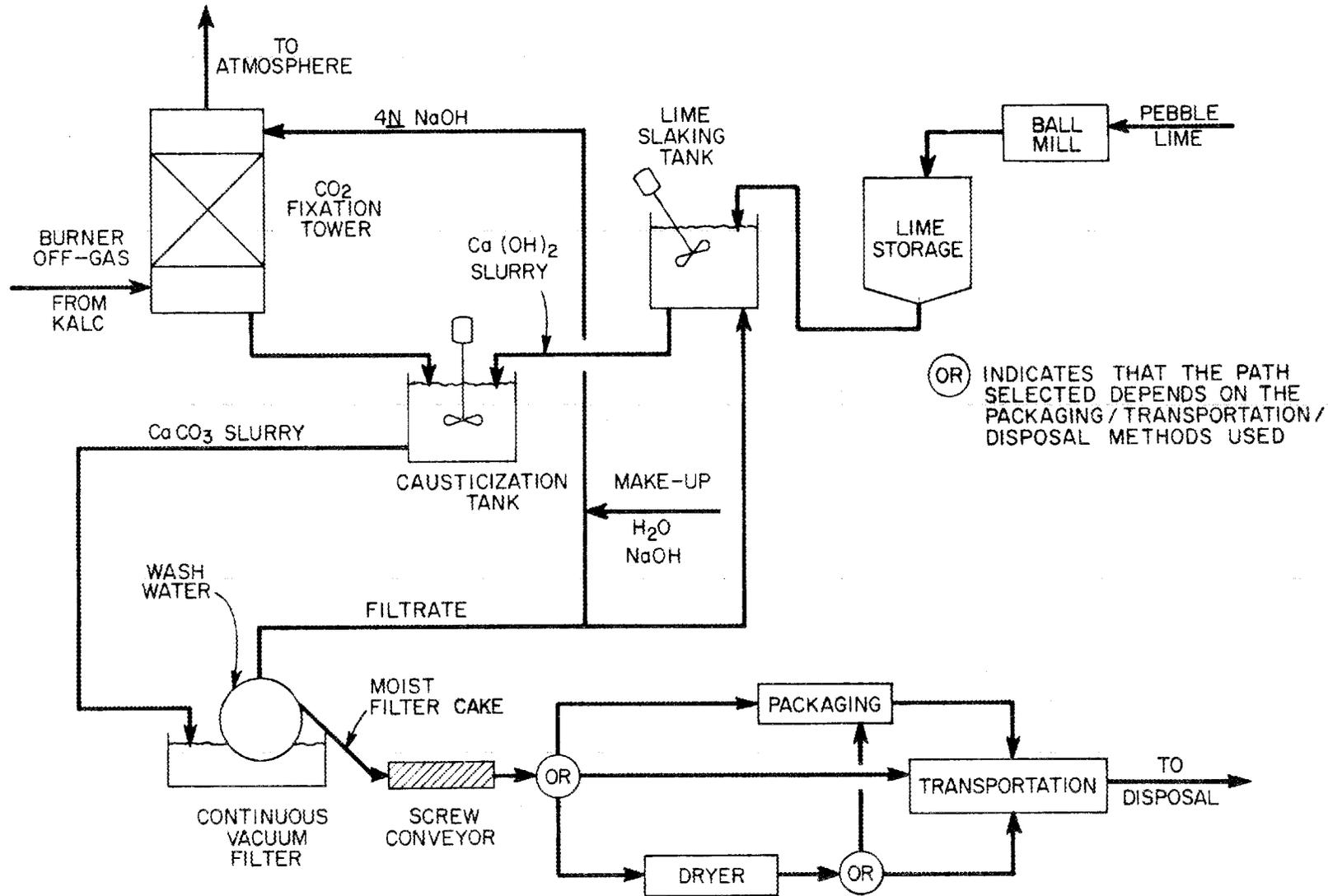


Fig. 3.2. Schematic of the double alkali CO₂ fixation process.

CaCO ₃	29,689 kg/day
Ca(OH) ₂	1,099 kg/day
H ₂ O	20,525 kg/day
NaOH	1,831 kg/day
Na ₂ CO ₃	113 kg/day

3.2.3 Discussion of the double alkali fixation process

The double alkali CO₂ fixation process is not used industrially as such, since the calcium carbonate produced using this process requires extensive washing to remove residual NaOH.⁴ However, most of the operations in the double alkali process have been proven as a result of their inclusion in other processes.

The most direct analogy with the double alkali CO₂ fixation system described herein is the double alkali SO₂ fixation system for removing SO₂ from stack gases.⁷ In this process, the NaOH solution is used to scrub the SO₂ from the stack gas, forming Na₂SO₄, Na₂SO₃, and NaHSO₃. These species are then causticized with slaked lime to regenerate the NaOH, forming CaSO₄ and CaSO₃ precipitates. However, the low SO₂ concentration has resulted in double alkali flowsheets with NaOH concentrations ranging from 0.08 to 0.5 N. The double alkali CO₂ flowsheet proposed herein uses much higher NaOH concentrations to reduce the possibility of flooding while still reacting with the required amount of CO₂.

Both the lime grinding, storage, and slaking and the filtration, drying, and packaging steps are very nearly the same as in the direct CO₂ fixation process. The only difference is the presence of NaOH in the water used for lime slaking.

The causticization reaction is carried out industrially in the manufacture of NaOH from soda ash (Na₂CO₃) and slaked lime [Ca(OH)₂].⁸ The causticization reaction is slightly endothermic (ΔH = 0.44 kcal/mole) and will proceed more rapidly when heated. The equilibrium constant for the causticization reaction is given by

$$K = \frac{[\text{OH}^-]^2}{[\text{CO}_3^{2-}]}$$

The 4 N NaOH concentration used in the double CO₂ fixation process results in the causticization reaction being approximately 90% complete at equilibrium.^{8,9} This, in turn, results in Na₂CO₃ being present in the filtrate and in the entrained water in the filter cake. The Na₂CO₃ in the filtrate which is used for lime slaking will react in the slaking reactor to form CaCO₃. The remainder of the filtrate, plus the make-up H₂O-NaOH, is recirculated through the CO₂ fixation tower.

The fact that the water in the filter cake contains substantial amounts of NaOH and lesser amounts of Na₂CO₃ requires that NaOH be continuously input to the process. If a water balance is to be maintained, the volume of wash water must approximately equal the volume of water in the filter cake, resulting in a poor NaOH recovery. It is estimated that 50% of the NaOH present in the water entrained in the CaCO₃ product can be recovered, resulting in 4.65% of the NaOH entering the filter and being lost to the filter cake. The alternative to this is to increase the wash volume and use an evaporator to maintain the water balance. This option was not investigated explicitly, but the extreme corrosivity of hot, concentrated caustic solutions indicates that this modification would not be desirable.

3.3 Comparative Evaluation of the Direct and Double Alkali CO₂ Fixation Processes

The following paragraphs will give a qualitative comparison of the most significant aspects of the two CO₂ fixation processes.

3.3.1 Availability of design data

At the present time, the double alkali process has an advantage over the direct process with respect to design data availability since data on the reaction rate of CO₂ with aqueous NaOH, causticization, and lime slaking are available in the open literature.⁹⁻¹² Although design data also exist for the direct CO₂ fixation system,³ the fact that they are not readily available in the open literature militates against the direct process.

3.3.2 Complexity

Clearly, the direct fixation process is superior in this respect. The double alkali process is more complex than the direct process physically because of the additional loop required and more complex chemically because of the introduction of NaOH and Na₂CO₃ into the system. The use of NaOH also requires that the filter cake be washed and that the lime be slaked with 4 N NaOH.

3.3.3 Solids

Both CO₂ fixation systems involve the handling of solids or slurries. The only piece of equipment which does not involve slurries or solids is the CO₂ fixation tower in the double alkali process, which should contain only soluble sodium compounds. It might, however, be possible for CaCO₃ or Ca(OH)₂ precipitation or scaling to occur in the double alkali fixation tower, depending on specific conditions, impurities, etc. Verification or negation of this possibility must await detailed experimental work.

3.3.4 Corrosion

The presence of concentrated NaOH in the double alkali process could result in corrosion problems. The expected corrosion rate of carbon steel in 4 N NaOH is about 0.004 in. per year, which is relatively low. However, there is a risk of stress-corrosion cracking when carbon steels are used with concentrated caustic solutions. As a result, the use of higher alloy steels or stainless steels may be required.

The small OH⁻ concentration, resulting from the low Ca(OH)₂ solubility, eliminates the possibility of stress-corrosion cracking, making mild steel a quite acceptable construction material for the direct process.

3.3.5 Economics

The detailed economic analyses of the CO₂ fixation processes are given in Sect. 8. However, for comparative purposes, the capital costs of the competing systems appear to be approximately equal, assuming that carbon steel can be used as the construction material for the double alkali CO₂

fixation process. The use of stainless steel could increase the capital cost of the double alkali process by as much as 33% [about \$0.65/(kg heavy metal)]. The operation, maintenance, and raw material costs are expected to be about the same for both systems, except for the 1873-kg/day NaOH make-up required of the double alkali process. At the current price of about \$0.14/lb, this amounts to \$578/day or \$1.61/kg heavy metal, a value almost equal to the estimated fixed charges on the capital cost of the fixation equipment.

3.3.6 Process recommendation

The following briefly summarizes the preceding discussion:

<u>Criterion</u>	<u>Favored Process</u>
1. Design data availability	Double alkali
2. Complexity	Direct
3. Solids	Neither
4. Corrosion	Direct
5. Economics	Direct

These considerations lead to the conclusion that the direct CO₂ fixation process is superior to the double alkali process and should therefore be considered to be the reference process. Fortunately, since the products of the two processes are nearly identical, the portion of this project pertaining to the CaCO₃ packaging, transportation, and disposal will be applicable to either of the CO₂ fixation processes.

4. CONCRETION AND PACKAGING OF THE CaCO₃ PRODUCT

4.1 Concretion

The CaCO₃ was assumed to be concreted with cement, a common practice for many types of low- and intermediate-level wastes. The monolithic product was assumed to contain 35 wt % cement, 35 wt % water, and 30 wt % CaCO₃, a typical composition for the concretion of inert solids with cement. The bulk density of the concreted product was taken to be 100 lb/ft³.

Concretion costs via cement were obtained,¹³⁻¹⁵ but proved to be relatively difficult to interpret. They typically included concretion, packaging, and disposal as a single cost. Even when a cost breakdown was given, the labor cost for all three steps was given as a single number. However, for low-level material (no significant fission product contamination), a cost of \$.40 per gallon of 46 wt % CaCO_3 slurry is probably appropriate. For higher-activity material (e.g., graphite with some fission product contamination), the concretion cost was taken to be \$.60/gal.

There are, of course, other concretion media available. Two media which may be of interest in this application are a polymer-type concretion agent (e.g., urea formaldehyde) and asphalt. Both should allow the incorporation of more CaCO_3 within a given concreted product mass. The polymer weighs less than either asphalt or cement but is relatively expensive. Asphalt should be able to contain 50 wt % CaCO_3 and has excellent properties for reducing the leachability of the contained waste. Asphalt is, however, flammable when subjected to moderate heating.

4.2 Packaging

When packaging was required for either the unconcreted or concreted product, it was assumed that standard 55-gal (7.5-ft^3) steel drums would be used. Available economics¹⁵ indicate that mass-produced drums have the lowest cost per unit of contained volume. Used drums sell for about \$8 each, while new drums are about \$10. Packaging costs, including labor and equipment, were taken to be \$12 per drum for low-level wastes and \$15 per drum for somewhat higher-level wastes. For those cases where unconcreted CaCO_3 is packaged at the reprocessing plant and unpackaged for concretion or bulk burial at the disposal site, an additional charge of \$4 per drum was assessed to cover the extra handling steps required.

5. TRANSPORTATION OF THE CaCO_3 PRODUCT5.1 Characterization of the CaCO_3 Product Being Transported

The CaCO_3 product that will be the basis for discussion in this section is the moist filter cake containing 60 wt % solids [$\text{CaCO}_3 + \text{Ca}(\text{OH})_2$] and 40 wt % H_2O . As was previously stated, the moist filter cake from the direct process is comprised of:

29,689 kg CaCO_3 /day,
 1,099 kg $\text{Ca}(\text{OH})_2$ /day,
 20,525 kg H_2O /day.

If the double alkali CO_2 fixation process is used, the cake also contains approximately

1,831 kg NaOH /day,
 113 kg Na_2CO_3 /day,

with both of the sodium salts being dissolved in the entrained water. Based on a moist filter cake bulk density of 102.2 lb/ft^3 (specific gravity = 1.64),² the resulting volume is $1107 \text{ ft}^3/\text{day}$ for the direct process, where no sodium salts are present. The packed density of dry commercial CaCO_3 ranges between 31.8 and 46.2 lb/ft^3 (specific gravity = 0.51 to 0.74).¹⁶ It is expected that a somewhat higher dry CaCO_3 packed density could be obtained in the CO_2 fixation process since a larger particle size can be produced. However, compaction will probably be required to attain a dry CaCO_3 product density equivalent to the effective moist filter cake density of 61.3 lb/ft^3 (specific gravity = 0.98).

Based on a volume of $1107 \text{ ft}^3/\text{day}$ (36 fuel blocks per day) and a 42,000-lb payload limit, approximately 2.7 truck shipments would be required each day. If rail transport were used, approximately one-third boxcar per day would be required.

One of the most important characteristics of the CaCO_3 product with respect to transportation is its radioactivity. Information related to the ^{14}C and ^{85}Kr activities present in the product is given in Table 5.1. The two principal sources of ^{14}C are the $^{14}\text{N}(\text{n,p})^{14}\text{C}$ and $^{13}\text{C}(\text{n},\gamma)^{14}\text{C}$

Table 5.1. Radioactivity of CaCO₃ product^a

Case No.	¹⁴ N (ppm)	¹⁴ C (Ci per fuel element ^b)	¹⁴ C (ppb)	⁸⁵ Kr (Ci per fuel element ^c)	^μ Ci/g product ^a			Ci/day		
					¹⁴ C	⁸⁵ Kr	Total	¹⁴ C	⁸⁵ Kr	Total
I ^d	0	0.030	69.2	5.31	0.0211	3.71	3.73	1.08	191	192
II ^d	26	0.160	359	5.31	0.1120	3.71	3.82	5.76	191	197
III ^d	100	0.530	1212	5.31	0.3710	3.71	4.08	19.08	191	210
IV ^e	0	0.030	69.2	531	0.0211	371	371	1.08	19,080	19,081
V ^e	26	0.160	359	531	0.1120	371	371	5.76	19,080	19,086
VI ^e	100	0.530	1212	531	0.3710	371	371	19.08	19,080	19,099

^a 60% solids, 40% H₂O.

^b Ref. 1.

^c Ref. 17.

^d Cases I-III assume the CO₂ fixation process follows KALC.

^e Cases IV-VI assume the CO₂ fixation process precedes KALC.

reactions. Since the amount of ^{13}C in a fuel block is fixed (natural abundance = 1.11%), the production rate of ^{14}C from ^{13}C is constant at 0.030 Ci/block.¹ The production rate of ^{14}C from ^{14}N is variable, depending on the amount of nitrogen initially present in the fuel block. The ^{14}N concentration in an HTGR fuel block has been estimated to be less than 30 ppm.¹ However, in Table 5.1 the calculated ^{14}C activities are given parametrically as a function of the ^{14}N concentration.

It was assumed that all of the Kr entering the CO_2 fixation process (see Fig. 2.1) was contained in the product. In Table 5.1, Cases I-III represent ^{85}Kr levels when the CO_2 fixation process follows the KALC process. The information given in Table 5.1 clearly indicates that, even when the "low" (Cases I-III) Kr cases are considered, the ^{85}Kr activity predominates over the ^{14}C activity at all ^{14}N concentrations examined. The ^{14}N concentration would have to be about 1000 ppm for the ^{14}C activity to equal the ^{85}Kr activity in the "low" Kr case. Without concretion to a monolithic form, the Kr is retained in the CaCO_3 product only by dissolution in the entrained water and by sorption. The ^{85}Kr can, therefore, be released by simply heating the product, or possibly by compressing the product.

5.2 Categorization of the CaCO_3 Product

5.2.1 Department of Transportation

The regulations governing the transport of radioactive materials, as set forth by the Department of Transportation (DOT),¹⁸ are quite complex. However, it is clear that the CaCO_3 product, as characterized in Table 5.1, can quite readily be transported in Type A packaging under existing regulations. The fact that Type A packaging can be used implies the following:

1. The product may be packaged in a wide variety of readily available metal or fiber drums, wooden boxes, or fiberboard boxes
2. The packaged product does not have to meet the stringent hypothetical accident conditions (1475°F for 30 min, water immersion for 8 hr, etc.), but must instead meet conditions principally consisting of various drop tests.

The high density of the product would probably negate the use of fiber-board containers if the product is classified as Type A.

It is desirable to have the product classified under the least stringent category as is possible since this maximizes the CaCO_3 product concretion, transportation, and disposal options from which an overall ^{14}C disposal scheme may be selected. It is probable that the CaCO_3 product may also be categorized as Low Specific Activity (LSA). The only question here is what the maximum allowable concentration of ^{85}Kr is in the product, since there is presently no limit given for the amount of ^{85}Kr which may be present in LSA material. The DOT regulations assign radioactive nuclides to "Groups," with ^{14}C being in Group IV and ^{85}Kr in Group VI. The activity limits to qualify for LSA are as follows:

Group I	<0.1 $\mu\text{Ci/g}$
Group II	<5 $\mu\text{Ci/g}$
Groups III, IV	<300 $\mu\text{Ci/g}$
Groups V-VII	Not specified

The fact that in the "high" Kr case (371 μCi of ^{85}Kr per gram) the ^{85}Kr activity is approximately equal to the activity limit for Groups III and IV, combined with the fact that the activity limits are becoming less stringent with increasing Group numbers, indicates that the CaCO_3 product (including the ^{85}Kr) can probably be considered as LSA material. LSA materials, when transported in vehicles assigned for the sole use of the consignor, must ". . . be packaged in strong, tight packages so that there will be no leakage of radioactive material under conditions normally incident to transportation."¹⁸ More significantly, there are no test conditions (drops, puncture, etc.) to be met by the packaged material. Thus, classification of the product as LSA could greatly reduce the packaging requirements for transport.

Considerable additional flexibility could be obtained if the LSA material could qualify for bulk (unpacked) shipment. The principal criterion here is that the product must have an activity less than 1.0 $\mu\text{Ci/g}$. Under this criterion, none of the products represented by the six cases in Table 5.1 would qualify for bulk shipment, the problem being the ^{85}Kr activity. This criterion could be met (for Cases I-III) by:

- (1) increasing the Kr recovery in the KALC process to 99.8% (DF = 500) or
- (2) showing that less than one-fourth of the Kr entering the fixation process will be present in the product as a result of either krypton's physical properties or degassing the product.

The advantages of bulk shipment will be discussed in Sect. 6.

5.2.2 IAEA Regulations

At the present time the DOT regulations regarding radioactive waste transportation are being rewritten to conform much more closely to current IAEA transportation regulations.¹⁹⁻²¹ Assuming that the rewritten DOT regulations are the same as the present IAEA regulations, the criteria which define LSA material are given in Table 5.2. The most restrictive portion of paragraph 121 (Table 5.2) for the case at hand is item 121(e). This item means that, if the radioactivity might be concentrated during transport, then the resulting concentrated activity must still be less than $10^{-4} A_2/g$. The appropriate values of A_2 are 100 Ci for ^{14}C and 1000 Ci for uncompressed ^{85}Kr . Thus, when concentrated, the specific activities must be less than 0.01 Ci/g for ^{14}C and less than 0.1 Ci/g for ^{85}Kr . It is difficult to conceive of a process for further concentrating the ^{14}C activity, since the $CaCO_3$ occupies nearly the total transport volume and is quite stable. However, the small amount of Kr present with the $CaCO_3$ is only fixed by physical forces (adsorption or dissolution) and presumably could be released by lowering the external pressure or heating. This could result in the formation of Kr bubbles at some point in the shipping package. These bubbles would have an activity of about 24.5 Ci per gram of krypton, far exceeding the specified limit of 0.1 Ci/g. This problem can be alleviated somewhat by using smaller shipping packages which contain a total ^{85}Kr activity less than 24.5 Ci. For example, a single 55-gal drum would contain only 1.29 Ci of ^{85}Kr , assuming the ^{14}C fixation process follows the Kr removal process. However, reduction of the ^{85}Kr activity to less than 0.1 Ci per package by decreasing the package size does not appear to be practical, since the required package size is about 4.25 gal (0.58 ft³). Thus, the moist filter cake cannot readily be shown to be LSA material under

Table 5.2. IAEA definition of low specific activity material^a

121. Low specific activity material (LSA) shall mean any of the following:

(a) Uranium or thorium ores and physical or chemical concentrates of those ores.

(b) Unirradiated natural or depleted uranium or unirradiated natural thorium.

(c) Tritium oxide in aqueous solutions, provided the concentration does not exceed 10 Ci/litre.

(d) Materials in which the activity, under normal transport conditions, is, and remains, uniformly distributed and in which the average estimated specific activity does not exceed $10^{-4} A_2/g$.^b

(e) Materials in which the activity is uniformly distributed and which, if reduced to the minimum volume under conditions likely to be encountered in transport, such as dissolution in water with subsequent recrystallization, precipitation, evaporation, combustion, abrasion, etc., would have an average estimated specific activity of no more than $10^{-4} A_2/g$.^b

(f) Objects of non-radioactive material contaminated with radioactive material, provided the non-fixed surface contamination does not exceed ten times the values given in Table XI and the contaminated object or the contamination on the object, if reduced to the minimum volume under conditions likely to be encountered in transport, such as dissolution in water with subsequent recrystallization, precipitation, evaporation, combustion, abrasion, etc., would have an average estimated specific activity of no more than $10^{-4} A_2/g$.^b

(g) Objects of non-radioactive material contaminated with radioactive material, provided that the radioactive contamination is in a non-readily dispersible form and the level of contamination averaged over 1 m^2 (or the area of the surface if this is less than 1 m^2) does not exceed $1 \mu\text{Ci}/\text{cm}^2$ for beta and gamma emitters and for low toxicity alpha emitters;^c $0.1 \mu\text{Ci}/\text{cm}^2$ for other alpha emitters.

^a Taken from ref. 20.

^b $A_2 = 100 \text{ Ci}$ for ^{14}C ; $A_2 = 1000 \text{ Ci}$ for uncompressed ^{85}Kr .

^c ^{235}U ; ^{238}U ; ^{232}Th ; ^{228}Th and ^{230}Th when diluted to a specific activity of the same order as that of natural uranium and thorium; radionuclides with a half-life of less than 10 days.

projected DOT regulations. If the CaCO_3 product is not to be shipped in the more restrictive Type A packaging, then some or all of the following actions may be required:

- (1) experimentally demonstrate that less than 0.1 Ci of ^{85}Kr per package will be desorbed/evolved ". . . under conditions likely to be encountered during transport . . .,"¹⁵
- (2) dry the CaCO_3 product to reduce the ^{85}Kr content to less than 0.1 Ci per package,
- (3) fix the Kr in place with, for example, cement to prevent its concentration during transport, or
- (4) increase the Kr recovery in the KALC process from 99 to 99.93%.

Bulk transport would be out of the question, since the larger mass of CaCO_3 and Kr in a single package would simply aggravate the existing ^{85}Kr concentration problem.

5.2.3 Summary of transportation regulations

The net result of Sects. 5.2.1 and 5.2.2 is that the CaCO_3 product (including 3.71 Ci of ^{85}Kr per gram of product) can probably be considered to be LSA material under present DOT regulations. Furthermore, this same CaCO_3 product can probably be made to conform to the projected LSA material definition by experimental demonstration and process modifications (product drying and/or concretion). Thus, in the remainder of this report, it is assumed that only the minimal packaging requirements described briefly in Sect. 5.2.1 need be met.

5.3 Radiation Doses During Transportation and Handling

The radiation level of the CaCO_3 product is of interest during both the handling and the transportation of the product. The charge for burial at commercial burial grounds increases stepwise depending on the radiation level of the material being buried. The product radiation level also defines the shielding requirements for shipping. The controlling requirement in this case is that the dose rate must be less than 10 mrem/hr and 6 ft from the external surface of the transport vehicle.¹⁸

The dose rates at the surface of 55-gal steel drums containing CaCO_3 product for the six cases defined in Table 5.1 are given in Table 5.3. The product was assumed to have a bulk density of 102.2 lb/ft^3 and to contain 40 wt % water. The dose rate calculations were performed using the ANISN code,²² and the bremsstrahlung spectrum and intensity were based on information given in refs. 23 and 24. As is evident from the information given in Table 5.3, ^{85}Kr accounts for virtually all of the radiation dose rate at the drum surface in all of the six cases examined. The dose rates in Cases I-III ("low Kr") are small enough so as to incur no additional burial charges and so as to present minimal handling problems. The dose rates in Cases IV-VI are relatively large, and in these cases considerable care would be required when handling the product. The burial charges in Cases IV-VI are about $\$3.25/\text{ft}^3$ as compared with $\$1.30/\text{ft}^3$ for less than 200 mrem/hr per package.

The dose rate from a vehicle carrying the CaCO_3 product was approximated by assuming the vehicle was an infinite plane source 56.8 cm (1 drum) thick. Since the ^{85}Kr gamma rays have a relatively low energy (0.514 MeV from 0.41% of all decay events), the outermost row of drums should be an effective shield for the ^{85}Kr gamma rays emanating from the drums in the interior of the truck. The results show that about 0.3 cm (0.12 in.) of iron would be required to reduce the dose rate in Cases I-III to 10 mrem/hr at a point 6 ft from the vehicle surface. For Cases IV-VI, about 7.35 cm (2.9 in.) of iron would be required. In either case, the drum thickness of 0.15 cm may be subtracted to get the required shielding thickness. These thicknesses may be put in perspective by assuming that 42,000 lb of product is to be shipped in a cubical container, which results in the minimum surface area for a rectangular-type container. For Cases I-III, the required iron shielding would weigh about 1600 lb, while for Cases IV-VI the iron shielding would weigh about 39,500 lb. Thus, the total weight shipped in Cases IV-VI is nearly double that in Cases I-III.

5.4 Transportation Costs

The transportation costs used in this study, given in Table 5.4,²⁵ are based on existing Interstate Commerce Commission (ICC) tariffs with Oak Ridge

Table 5.3. Dose rate at the surface of a 55-gal drum containing CaCO₃ product^a with ¹⁴C and ⁸⁵Kr

Case No.	¹⁴ N (ppm)	⁸⁵ Kr (Ci per fuel element)	¹⁴ C		⁸⁵ Kr		Total	
			mCi per drum	mrem per hr	Ci per drum	mrem per hr	Ci per drum	mrem per hr
I	0	5.31	5.38	2.75 x 10 ⁻³	0.949	22.34	0.954	22.34
II	26	5.31	28.7	1.47 x 10 ⁻²	0.949	22.34	0.978	22.35
III	100	5.31	94.9	4.85 x 10 ⁻²	0.949	22.34	1.04	22.39
IV	0	531	5.38	2.75 x 10 ⁻³	94.9	2,234	94.9	2,234
V	26	531	28.7	1.47 x 10 ⁻²	94.9	2,234	95.0	2,234
VI	100	531	94.9	4.85 x 10 ⁻²	94.9	2,234	95.0	2,234

^a Moist filter cake containing 60% solids and 40% H₂O.

Table 5.4. Transportation costs from Oak Ridge to various destinations

Destination	One-way distance from Oak Ridge (miles)	One-way cost (\$/100 lb)
Morehead, Ky.	188	0.93
Barnwell, S.C.	339	2.13
West Valley, N.Y.	630	2.13
Los Alamos, N.M.	1403	4.28
Hanford, Wash.	2366	6.09

as the originating point. If a standard vehicle owned by a commercial carrier is used, the one-way tariff is applicable, since the vehicle need not be returned to the originating point. If a specially designed CaCO_3 product transport vehicle is used with a commercial carrier, the vehicle will have to be returned empty to the originating point, and the rates in Table 5.4 would be increased. In the latter case, it would probably be more economical to ship the product in specially designed vehicles which are owned by the reprocessor, thus circumventing the ICC tariffs. Indications are that the round-trip cost of a self-owned vehicle is roughly the same as the one-way ICC tariff.

The cost of transporting the product by rail was not investigated. Representatives of the commercial burial grounds indicated that burial grounds are often inaccessible to rail traffic. Additionally, the railroads in some regions of the country, most notably the Northeast, have virtually refused to carry radioactive cargo. However, it is expected that the cost of transporting the product by rail would be roughly the same as transportation by road vehicles.

Shipping the product by barge should reduce shipping costs to about 50% of those for road or rail transport on a per-mile basis. The limitation here is to ensure that both the reprocessing plant and disposal sites are accessible to barge traffic. This may be difficult to accomplish, since most burial grounds are purposely removed from water sources to minimize the possibility of contamination of surface water and seepage into circulating groundwater.

6. DISPOSAL OF THE CaCO_3 PRODUCT

6.1 Introduction

The method by which the CaCO_3 product is disposed is extremely important since this not only determines the required characteristics of the CaCO_3 product, but may also dictate the packaging and transportation requirements. The principal problem encountered when trying to define the best method for disposing of the product is that the regulations regarding

the disposal of ^{14}C are nonexistent, and the regulations regarding the management of low-level wastes, in general, are variable and are being continuously reevaluated.

The variability of the regulations is a result of their being defined on a state-by-state basis instead of being defined by Federal authorities. For example, the state of Kentucky appears to be rather conservative, strictly limiting the waste forms it will accept, while South Carolina appears to be more flexible and may be willing to consider exemptions for specific waste types.

The fact that the regulations concerning low-level waste management are in a continuous state of flux is a symptom of the current difficulties being encountered in all phases of waste management. Waste management technology is presently the subject of intensive research and development, as well as widespread debate, in both public and private forums. It appears that the research, development, and debate have just begun and, therefore, that resolution of the problems concerning waste management regulations, including low-level wastes, will be several years in the future. The only point that most people who are experienced in low-level waste management agree upon is that the trend is increasingly toward concretion of all radioactive wastes.

The remainder of Sect. 6 will be devoted to a discussion of the following CaCO_3 product disposal options:

- (1) shallow-land burial
- (2) partial-block burning and burial
- (3) hydraulic fracturing
- (4) deep-sea disposal
- (5) geologic repository emplacement

6.2 Shallow-Land Burial Options

Shallow-land burial of low-level radioactive wastes typically involves burial of the waste in surface trenches having dimensions of 500 ft x 40 ft x 20 ft deep. When radioactive waste has accumulated to a depth of 15 to 17 ft, the trench is backfilled with previously excavated earth. Drainage

is provided for the trench, and provisions are made for monitoring the water (if any) draining from the trench. Commercial shallow-land burial sites are:

<u>Location</u>	<u>Operated by</u>
Sheffield, Ill.	Nuclear Engineering Co., Morehead, Ky.
Richland, Wash.	
Beatty, Nev.	
Morehead, Ky.	
Barnwell, S. C.	Chem-Nuclear Services, Inc., Bellevue, Wash.
West Valley, N. Y.	Nuclear Fuel Services, West Valley, N. Y.

The Nuclear Fuel Services' burial ground is not presently accepting wastes for burial.

Noncommercial shallow-land burial facilities are maintained by the Federal government at ORNL, SRL, NRTS (Idaho), Hanford, and LASL.²⁶

Charges for burial appear to be quite uniform, and are \$1.30/ft³ for a dose rate less than 200 mrem/hr at the surface of the package. If the dose rate at the surface is between 200 and 5000 mrem/hr, the charge is \$3.25/ft³. These rates may be reduced if large volumes of waste (greater than 100,000 ft³/yr) are being buried, as would be in the case with the CaCO₃ product disposal, which amounts to about 450,000 ft³ of uncreted material per year from the demonstration size reprocessing plant. The volume discount could be expected to be about 10%. Surcharges are levied for large packages (greater than 15 tons per package) and for high-density materials (greater than 200 lb/ft³).

The following four shallow-land burial options have been identified:

- (1) burial of unpackaged, uncreted CaCO₃ product,
- (2) burial of packaged, uncreted CaCO₃ product,
- (3) burial of unpackaged, ccreted CaCO₃ product,
- (4) burial of packaged, ccreted CaCO₃ product.

Low-level waste burial charges were discussed previously. Concretion and packaging costs are discussed in Sect. 4. The following discussion will concern the viability of the four shallow-land burial concepts in the future.

6.2.1 Burial of unpackaged, unconcreted CaCO_3 product

This disposal method was included principally for the sake of completeness since it is the fourth possible combination of the packaged-unpackaged and uncreted-concreted product choices. This practice is not permitted at any of the commercial burial sites. The ready solubility of CaCO_3 in even weakly acid water and the susceptibility of unpackaged, uncreted material to unintentional disturbance by virtually any type of excavation indicate that there is little hope that this disposal method will be permitted in the future unless there are radical reverses in the direction of present regulatory policy.

A slight variation of this concept was suggested by Brooks et al.;¹ the plan was to bury the CaCO_3 in layers adjacent to the reprocessing site, with each layer being covered by some waterproof material. This disposal plan is subject to the same problems cited before. In addition, the selection of the reprocessing plant location would be restricted by the availability of sites with an acceptable geology for burial of the CaCO_3 product.

To summarize, shallow-land burial of unpackaged, uncreted, ¹⁴C-contaminated CaCO_3 is not acceptable under present regulations, and probably will not be acceptable under future regulations. The implementation of this plan would probably require that the CaCO_3 product be given a special exemption from low-level waste disposal regulations, an arduous and uncertain course at best.

6.2.2 Burial of packaged, uncreted CaCO_3 product

Shallow-land burial of packaged, uncreted CaCO_3 product consists of packaging either the moist CaCO_3 filter cake or the dry CaCO_3 product in containers, assumed to be standard 55-gal drums in this study.

Current practice regarding shallow-land burial of packaged, unconcreted low-level wastes varies depending on which commercial burial site is being considered. For example, the burial site at Morehead, Kentucky, will not accept unconcreted CaCO_3 product, dry or moist. The burial site at Barnwell, South Carolina, will probably accept packaged, unconcreted solids, but only if the solids are dry (i.e., a moist filter cake is not acceptable).

As usual, the future regulations regarding the burial of packaged, unconcreted low-level wastes are open to speculation. In the short term (i.e., for the demonstration plant), the disposal of packaged, unconcreted CaCO_3 product will probably be acceptable. In the longer term, the fact that the CaCO_3 product presumably requires containment for tens of thousands of years indicates that unconcreted burial will probably not be acceptable for a commercial HTGR economy since the containers will probably not retain their integrity for even a few hundred years. This option might be rendered somewhat more acceptable if cement storage containers were used.

6.2.3 Burial of unpackaged, concreted CaCO_3 product

This option would involve mixing the CaCO_3 product with appropriate amounts of cement and water and placing the wet mixture directly in the shallow-land burial trench. The result will be massive (about 40 ft x 15 ft x 500 ft), unpackaged blocks of concreted CaCO_3 . This disposal method is not presently acceptable at any of the commercial burial grounds.

The principal advantage in this disposal method (other than the elimination of packaging costs) follows from the realization that any packaging used will probably retain its integrity for, at the most, several centuries. After the packaging has been breached, the loss of large amounts of ^{14}C -contaminated CaCO_3 to the environment is prevented by the low rate at which the CaCO_3 is leached from the concrete. Thus, for long-term safety it may be more desirable to have a minimum surface-to-volume ratio rather than packaging, since the leach rate is proportional to exposed surface area.

Technically, the burial of unpackaged, concreted product would seem superior to the burial of packaged, concreted waste (Sect. 6.2.4) because

the former is more economical (no packaging cost), and it has a lower long-term leach rate due to its lower surface-to-volume ratio. However, the emotionalism surrounding the disposal of radioactive wastes might well lead to the required use of packaging to placate those who perceive short-term risks more acutely than long-term risks. Since this method of waste disposal has not been extensively used, the "propose it and see what happens" approach appears to be the only way to determine the acceptability of the unpackaged burial of concreted CaCO_3 .

6.2.4 Burial of packaged, concreted CaCO_3 product

This burial option consists of packaging and burying the CaCO_3 product which has been mixed with appropriate amounts of cement and water so as to result in the product being concreted in a monolithic form. The volumes, masses, and costs involved in concretion are discussed in Sect. 4. The CaCO_3 may be concretized at either the reprocessing plant or the burial site.

Shallow-land burial of concreted, packaged low-level wastes is presently acceptable (and preferable) at all of the commercial burial grounds. Additionally, this burial option can probably be expected to remain acceptable in the future. The difficulty with this option is one of economics, since the twin burdens of concretion and packaging are present. The large mass of the concreted product can result in considerable additional transportation cost. This will be discussed in detail in Sect. 8.

6.3 Partial Block Burning and Concretion

This method of disposing of the large volume of ^{14}C -contaminated graphite is a variation on the burial options described in Sect. 6.2. Here, the graphite matrix rods containing the heavy metal (principally U and Th) and the fission products are removed from the HTGR fuel block by either (core) drilling them out or by pushing them out. The graphite matrix and the heavy metal which have been removed from the graphite block are subjected to the standard reprocessing sequence (burn, leach, etc.), with the removed graphite matrix ultimately being fixed as CaCO_3 . The

remainder of the graphite block is packaged with the CaCO_3 product with no further processing, and the whole mass is disposed of as a unit.

The principal advantage resulting from this disposal method is a marked reduction in the volume and mass of ^{14}C -contaminated waste which must be buried. In the case where the fuel rods are pushed out of the graphite block, the waste volume and mass are reduced to about 50% of the CaCO_3 volume resulting from whole block burning. The waste volume and mass are reduced to about 80% of the original volume when the fuel rods are drilled out. The difference between drilling and pushing the rods out results from the required use of an oversized core drill. These reductions in volume and mass are significant in view of the substantial costs of concretion, packaging, transportation, and burial.

Unfortunately, both of the partial block burning schemes have several other disadvantages. Firstly, the hardware that would be required to push or drill the fuel rods out of the graphite block promises to further complicate the already complex and expensive operations involved in the HTGR head-end process. Secondly, the graphite block which is not burned and processed contains a multitude of adsorbed fission products from broken fuel particles as well as activation products. The radiation from these adsorbed species is substantial and would result in the unburned block having to be treated as an intermediate- or high-level waste. Thus, what was once very low-level waste now requires remote handling, shielded transport, and quite probably burial in deep geological strata (e.g., a salt mine repository). The economic penalties associated with these disadvantages, combined with the still large volume of waste (75 to 120 drums per day), lead to the conclusion that partial block burning should not be considered as a method for disposing of ^{14}C -contaminated graphite.

6.4 Disposal of CaCO_3 by Hydraulic Fracturing

This method consists of mixing the aqueous CaCO_3 waste with preblended dry solids containing principally cement, and then pumping the resulting slurry down a well and out into a conformable, nearly horizontal fracture in a thick shale formation at the desired depth. The cased well is prepared

for the injection by perforating the casing at the desired depth and pressurizing the well with water. This induces a fracture in the rocks, which is further extended as the slurry is pumped into it. After the pumping phase is completed, the cement slurry is allowed to harden under pressure, thereby forming a thin, horizontal grout sheet. This procedure can be repeated successively up the well, creating a stack of horizontal grout sheets.²⁶

Assuming that a 30 wt % CaCO_3 slurry is injected, a volume of 22,500 gal/day (700 gal per fuel element per day) would have to be injected. Cost estimates for hydraulic fracturing range from \$0.50 to \$1.00/gal.²⁷ A value of \$0.50/gal was used in this study because economies of scale would accrue in a hydraulic fracturing operation of this magnitude.

The principal advantage of this process is that the wastes are injected and immobilized at depths (360 to 900 ft) well below the zone of circulating water. This greatly reduces the possibility of any leached ^{14}C -contaminated CaCO_3 reaching the environment.

The disadvantages of the hydraulic fracturing are as follows:

1. The capacity of an injection well is limited. The total waste volume injected at ORNL up to the present time is about one million gallons, as compared with the required HTGR demonstration reprocessing plant injection rate of 5 to 10 million gallons per year. Thus, many injection wells will probably be required.
2. Hydraulic fracturing requires rather specific geologies to ensure horizontal fracturing and the absence of circulating water. This restricts the available reprocessing plant sites.
3. The uncertainties inherent in the waste placement may make licensing this process difficult.

To summarize, the unfavorable economics, the potential uncertainties, and the restrictions inherent in the licensing and operation of the hydraulic fracturing concept make it unsuitable in this application.

6.5 Deep-Sea Disposal of the CaCO_3 Product

Deep-sea disposal of virtually every type of radioactive waste produced has been suggested during the last several years. A proposal to dispose of the CaCO_3 product in the deep sea receives further impetus from the fact that the deep sea contains about 90% of the earth's ^{14}C inventory.¹

Active sea disposal of radioactive waste is centered in the western European nations, with considerable interest in Japan. There has been no viable commercial sea disposal activity in the United States for over 10 years, due principally to unfavorable economics. The long transportation distances required, combined with the high cost of concretion, would also appear to make the disposal of CaCO_3 in the deep sea uneconomical.

The regulatory climate regarding radioactive waste disposal in the deep sea is also unfavorable. The following appears in the Draft Environmental Statement on the Management of Commercial High-Level and Transuranium-Contaminated Radioactive Wastes:²⁸

It may be assumed that in administering sea disposal regulations, the EPA will closely follow a prior policy recommendation by the Council on Environmental Quality (CEQ) as follows:

"The policy recommended would continue the practice of prohibiting high-level radioactive wastes in the ocean. Dumping other radioactive materials would be prohibited, except in a very few cases for which no practical alternative offers less risk to man and his environment."

---Ocean Dumping, a National Policy,
A Report to the President prepared
by the Council on Environmental
Quality, October 1970.

In view of both the unfavorable economics and the unfavorable regulatory climate, the concept of deep-sea disposal of the CaCO_3 product should probably not be considered.

6.6 Geologic Repository Emplacement of the CaCO_3 Product

Because ^{14}C has a relatively long half-life (5730 years), there is a significant chance that the CaCO_3 product may have to be placed in a geologic repository which is principally designed for non-heat-generating wastes contaminated by long-lived alpha emitters.

Logically, the burial of ^{14}C -contaminated waste in a geologic repository should not be required, since ^{14}C has a much lower radiotoxicity than the alpha emitters. The magnitude of this difference is evident from Table 6.1, wherein the radiotoxicity of ^{14}C , as measured by the ingestion Radionuclide Concentration Guide (RCG), is compared to the RCGs of several alpha emitters with comparable half-lives and to that of tritium. The least toxic of the alpha emitters is 200 times as toxic as ^{14}C , whereas ^{14}C is only 3.75 times as toxic as tritium. As a result of this low toxicity, ^{14}C should not have to be treated as an alpha emitter, assuming that the regulations are written in some logical fashion. The only caveat here is, again, that the emotionalism surrounding the management of radioactive wastes might force ^{14}C to be treated like an alpha emitter, even though it is much less toxic than the alpha emitters. Unofficial conversations with a representative of the NRC indicate that there is a significant chance that geologic repository emplacement of the CaCO_3 product will be required.

Costs for waste burial in a geologic repository are not well known, but it is felt that a charge of \$24.50/rectilinear ft^3 would be appropriate for present comparative purposes.

6.7 Summary of the Status of the Options Available for Disposal of the CaCO_3 Product

The most economical of the CaCO_3 product disposal options which would probably be environmentally acceptable, in decreasing order of preference, are:

- (1) shallow-land burial of packaged, unconcreted CaCO_3 product,
- (2) shallow-land burial of unpackaged, concreted CaCO_3 product,
- (3) shallow-land burial of packaged, concreted CaCO_3 product.

Table 6.1. Comparison of the radiotoxicity of ^{14}C with that of other nuclides

Nuclide	Half-Life (years)	Decay mode(s)	Radionuclide Concentration Guide ^a (ingestion, $\mu\text{Ci}/\text{m}^3 \text{H}_2\text{O}$)
^3H	12.33	β^- (0.0186 MeV max)	3.0×10^{-3}
^{14}C	5730	β^- (0.156 MeV max)	8.0×10^{-4}
^{226}Ra (soluble)	1600	α (4.78 MeV max)	3.0×10^{-8}
^{240}Pu (soluble)	6540	α (5.17 MeV max), SF ^b	5.0×10^{-6}
^{243}Am (soluble)	7370	α (5.28 MeV max)	4.0×10^{-6}
^{245}Cm (soluble)	8500	α (5.36 MeV max)	4.0×10^{-6}
^{246}Cm (soluble)	4760	α (5.39 MeV max), SF ^b	4.0×10^{-6}

^a Values taken from ref. 29.

^b SF = Spontaneous fission.

Shallow-land burial of unconcreted, unpackaged product was rejected because of its anticipated environmental unacceptability. Additionally, if, as discussed in Sect. 5, the bulk shipment of the CaCO_3 product is not allowed because of the presence of krypton, Option 2 (shallow-land burial of unpackaged, concreted CaCO_3) would be possible only if the CaCO_3 were shipped in packages which were emptied at the burial site. The other disposal options (partial block burning, hydraulic fracturing, deep-sea burial, geologic repository burial) were rejected on the basis of some combination of environmental, technical, or economic (see Sect. 8) grounds.

Finally, it should be noted that at the present time the evaluation of various CaCO_3 disposal options is necessarily quite subjective. Therefore, a continuing effort should be made to seek out individuals with expertise in the waste management area with an eye toward verifying the above discussion and monitoring current developments regarding waste management.

7. LOCATION OF THE CO_2 FIXATION SYSTEM WITH RESPECT TO THE KALC PROCESS

It has been suggested³⁰ that the KALC system for removing ^{85}Kr might possibly be minimized with regard to size, or eliminated altogether, by placing the CO_2 fixation system before the KALC process in the off-gas cleanup system.

7.1 Elimination of KALC

If the KALC system is to be eliminated, at least 99% of the Kr in the burner off-gas stream must be entrained with the CaCO_3 product. To ensure the fixation of the Kr with the CaCO_3 product, the following modifications must be made to the direct CO_2 fixation process (Fig. 3.1):

1. The continuous vacuum filter must be replaced by a thickener. The air being drawn through the filter cake would desorb Kr, and the reduced pressures would cause Kr which was dissolved in the water to be evolved.

2. The product cannot be dried, since the major portion of the sorbed and dissolved Kr would be released by the heat.
3. To prevent the release of the relatively large amounts of Kr in the CaCO_3 product during transportation and disposal operations, it is expected that immediate product concretion will be required.

Additionally, there may be a problem with retaining the Kr in aqueous solution within the CO_2 fixation tower. In the original direct CO_2 fixation process, the flow rate of water through the tower is about 50 times larger (at 140°F) than the theoretical amount required to dissolve all of the Kr entering the tower, assuming that the solubility of Kr in water is unaffected by the presence of other solutes and suspended solids. However, as the Kr concentration in the water increases (i.e., the driving force for dissolution and sorption decreases), it may prove difficult to limit the amount of Kr leaving the tower to less than 1% of that entering the tower.

The following factors combine to make this option one of the most expensive options considered:

1. The CaCO_3 -Kr must be concreted and packaged.
2. Both the CaCO_3 -Kr and the inert concretion agent must be transported, increasing the transportation cost.
3. The radioactivity of the Kr will require substantial shielding during concretion, packaging, and transportation.
4. The large volume of the concreted CaCO_3 -Kr will result in increased burial costs.
5. The radioactivity of the Kr will result in increased burial costs.

When all of these factors are taken into consideration (see Sect. 8.2), the resulting ^{14}C disposal cost is equivalent to a charge of \$86.20/kg heavy metal. As a result of the excessive cost and the difficulty in making sure that less than 1% of the Kr is released, it is recommended that this option not be pursued.

7.2 Size Reduction of the KALC Process

If, on the other hand, the size of the KALC system is only to be minimized, then

- (1) the CaCO_3 product must contain less than 1% (preferably 0.1%) of the Kr originally in the off-gas stream to facilitate packaging, transportation, and disposal of the CaCO_3 and,
- (2) the effluent stream containing the Kr should contain less than 1% of the CO_2 originally present in the off-gas stream and should be diluted as little as possible by other gas streams to minimize the size of the KALC system.

Assuming there is no dilution of the effluent stream containing Kr, the gas volume the KALC system must handle is reduced approximately sixfold (see Fig. 2.1) by placing the CO_2 fixation process before the KALC system.

The modifications required in the direct CO_2 fixation flowsheet (Fig. 3.1) to allow the size of the KALC system to be reduced are as follows:

1. The vacuum filter must be replaced by a thickener to eliminate dilution of the effluent Kr by air passing through the filter cake.
2. The product must be dried to drive off Kr adsorbed on the CaCO_3 and dissolved in the water in the filter cake.

If a carrier gas is used in the dryer, it must be routed through the KALC system to remove the Kr desorbed and evolved from the CaCO_3 product during drying, increasing the size of the KALC system. Dryers which minimize the amount of carrier gas used (e.g., vacuum dryers) are typically rather expensive to operate and have a limited throughput.

Thus, it appears that minimizing the size of the KALC system would entail many process complexities and uncertainties, as well as significant additional costs. In view of the fact that a Kr removal system must be developed for this case anyway, the additional complexities, uncertainties, and costs do not appear to be worth pursuing.

8. ECONOMICS

8.1 Cost of Fixing CO₂ as CaCO₃

8.1.1 Capital cost

An estimate of the capital cost of the CO₂ fixation process is given in Table 8.1. The installed capital costs were obtained from standard sources for chemical engineering cost data³¹⁻³⁵ and were escalated to a constant, 1975 dollar basis using the M&S Equipment Cost Index. The double alkali process was used as the basis for capital cost estimation because of the ready availability of reaction rate data which enabled the fixation tower to be designed. However, the capital cost of the direct fixation process should be very nearly the same, since most of the process equipment is identical. A "nuclear contingency factor" of 50% has been included to account for the requirement that this entire system be "tighter" than a normal industrial system to prevent leaks and to facilitate maintenance.

8.1.2 Total CO₂ fixation cost

The total cost of producing dry CaCO₃, including fixed charges, labor, utilities, and raw material costs, is given in Table 8.2. The cost of NaOH make-up was not included, since the estimate in Table 8.2 is intended to pertain to the direct fixation process where no NaOH is required. However, at the current price of about 14¢/lb NaOH, the NaOH make-up cost would be about \$169,000/year (\$1.56/kg heavy metal).

8.2 Total Cost of Producing and Disposing of ¹⁴C-Contaminated CaCO₃

The individual cost components comprising the total cost of producing and disposing of ¹⁴C-contaminated CO₂ have been discussed previously in the sections where the components were described. In this section, these individual cost components will be combined to yield total costs for several different sequences of packaging, concretion, transportation, and disposal. Toward this end, nine cases have been defined based on the options

Table 8.1. Estimated capital cost of fixing CO₂ as CaCO₃^a

Item	Capital cost (\$)
Fixation tower	38,700
Pumps	6,700
Causticization tank and agitator	44,600
Ball mill	27,800
Solids storage	900
Conveyors	20,200
Vacuum filter	70,700
Screw conveyor	8,400
Dryer and pneumatic system	<u>37,900</u>
Total installed equipment	255,900
Building and facilities	<u>260,900</u>
Base capital cost	516,800
Normal contingency @ 15%	77,500
Nuclear contingency @ 50%	<u>258,400</u>
Total capital cost	852,700

^a Double alkali process, 1975 dollars; capital cost of direct process assumed to be the same.

Table 8.2. Estimated total cost of fixing CO₂ as CaCO₃ using the direct fixation process

Component	Cost (\$/year)
Fixed charges @ 25% of total capital cost/year	213,000
Labor (10 men @ \$15,000/year)	150,000
Utilities, supplies, replacements (estd.)	75,000
Lime @ \$27.50/ton	<u>159,000</u>
Total fixation cost	\$597,000/year
Equivalent unit charge ^a	\$5.53/kg heavy metal

^a 10 kg heavy metal/block; 36 blocks/day; 300 days/year.

discussed in Sects. 4 through 7. The reprocessing plant is assumed to be located at Oak Ridge, Tennessee, in all cases.

Case 1

The dry CaCO_3 is assumed to be transported in 55-gal drums to a burial site in the vicinity of Barnwell, South Carolina, where it is buried as an unpackaged, unconcreted solid. The drums are recovered for reuse.

Case 2

The dry CaCO_3 is assumed to be packaged (unconcreted) in 55-gal steel drums and transported to the burial site in the vicinity of Barnwell, South Carolina, where it is buried without concretion.

Case 3

The dry CaCO_3 is assumed to be transported in 55-gal steel drums to the burial site in the vicinity of Barnwell, South Carolina, where it is mixed with cement and water and poured into burial trenches without packaging. The drums are recovered for reuse.

Case 4

The dry CaCO_3 is packaged into 55-gal drums and transported by truck to a burial site in the vicinity of Barnwell, South Carolina, where it is concreted and subjected to shallow-land burial.

Case 5

The CaCO_3 is assumed to be mixed with cement and water into 55-gal drums at the reprocessing plant and transported by truck to the burial site in the vicinity of Barnwell, South Carolina, where it is subjected to shallow-land burial.

Case 6

The CaCO_3 is assumed to be mixed with an appropriate amount of cement and water and then pumped into a deep geological stratum (at the reprocessing site) far removed from circulating ground water.

Case 7

The CaCO_3 is assumed to be mixed with cement and water into 55-gal drums, loaded onto barges, and transported to the deep sea (3000 miles round trip assumed), where the concreted mass is dumped.

Case 8

The dry CaCO_3 is assumed to be packaged (unconcreted) in 55-gal drums and transported to a geologic repository in the vicinity of Barnwell, South Carolina, where it is unpackaged, concreted, repackaged, and buried in the repository.

Case 9

The CaCO_3 containing 99+ % of the Kr initially present in the CO_2 is assumed to be mixed with cement and water into 55-gal drums and transported in a shielded truck (0.5 lb of shielding per pound of payload) to the vicinity of Barnwell, South Carolina, where it is subjected to shallow-land burial.

The overall costs of producing and disposing of ^{14}C -contaminated CaCO_3 for the nine cases described above are given in Table 8.3. Table 8.4 briefly summarizes the economic bases used in Table 8.3. Table 8.5 is a summary of the packaging, concretion, transportation, and disposal options which define each of the nine cases examined. The costs are given in both dollars/day and dollars/kg heavy metal (U + Th) to facilitate comparison with existing fuel cycle cost information. For an HTGR, a levelized fuel cycle cost of 2.0 mills/kWhr(e) is approximately equivalent to a charge of \$1750/kg heavy metal.

Before discussing the results given in Table 8.3, it would be useful to briefly recapitulate the results of the section discussing the waste disposal options (Sect. 6). Referring to Table 8.5, the three recommended CaCO_3 disposal options, in decreasing order of desirability, are:

- (1) Case 2,
- (2) Case 3, and
- (3) Cases 4 and 5.

Cases 1 and 7 were rejected principally on the basis of probable regulatory unacceptability. Cases 6, 8, and 9 were considered likely to be accepted from a regulatory viewpoint.

Table 8.3. Overall cost of producing and disposing of ^{14}C -contaminated CaCO_3 ^a

Cost Component	Case 1		Case 2		Case 3		Case 4		Case 5	
	\$/day	\$/kg HM ^b								
CaCO_3 production	1,991	5.53	1,991	5.53	1,991	5.53	1,991	5.53	1,991	5.53
Packaging	590	1.64	1,771	4.92	590	1.64	4,208	11.69	3,618	10.05
Concretion	0	0	0	0	5,010	13.92	5,010	13.92	5,010	13.92
Transportation	1,446	4.02	1,446	4.02	1,446	4.02	1,446	4.02	6,432	17.87
Disposal	<u>1,439</u>	<u>4.00</u>	<u>1,439</u>	<u>4.00</u>	<u>2,942</u>	<u>8.17</u>	<u>2,942</u>	<u>8.17</u>	<u>2,942</u>	<u>8.17</u>
Total	5,466	15.19	6,647	18.47	10,476	29.11	15,597	43.33	19,993	55.54

Cost Component	Case 6		Case 7		Case 8		Case 9	
	\$/day	\$/kg HM ^b	\$/day	\$/kg HM ^b	\$/day	\$/kg HM ^b	\$/day	\$/kg HM ^b
CaCO_3 production	1,991	5.53	1,991	5.53	1,991	5.53	1,991	5.53
Packaging	0	0	3,618	10.05	4,208	11.69	4,523	12.56
Concretion	0	0	5,010	13.92	5,010	13.92	7,515	20.88
Transportation	0	0	9,695	26.93	1,446	4.02	9,648	26.80
Disposal	<u>11,250</u>	<u>31.25</u>	<u>0</u>	<u>0</u>	<u>88,641</u>	<u>246.23</u>	<u>7,354</u>	<u>20.43</u>
Total	13,241	36.78	20,314	56.43	101,296	281.39	31,031	86.20

^a See Table 8.4 for a summary of the economic bases of this table; see Table 8.5 for a summary of the case characteristics.

^b 2.0 mills/kWhr(e) \approx \$1750/kg heavy metal.

Table 8.4. Bases for Table 8.3

Operation	Bases
CaCO ₃ production	See Tables 8.1 and 8.2 10 kg heavy metal/block; 36 blocks/day; 300 days/year
Packaging	55-gal (7.5-ft ³) drums @ \$12 each \$15/drum for Case 8 because of shielding \$4/drum for handling in Cases 1, 3, 4, and 8
Concretion	40¢/gal of 46 wt % CaCO ₃ slurry Concreted product 30 wt % CaCO ₃ , 100 lb/ft ³ 60¢/gal for Case 8 to account for shielding
Transportation	Truck Oak Ridge, Tenn. to Barnwell, S. C. \$2.13/100 lb Shielded truck 0.5 lb shield/lb payload Barge \$4.28/100 lb (round trip)
Disposal	Shallow-land burial \$1.30/ft ³ for < 200 mR/hr \$3.25/ft ³ for 200 to 5000 mR/hr Geologic repository \$24.50/ft ³ (rectilinear volume, 12 ft ³ /drum) Hydraulic fracturing \$0.50/gal, 30 wt % CaCO ₃

Table 8.5. Summary of case definitions for Table 8.3

Case number	Packaging	Concretion	Transportation	Disposal
1	No	No	Truck	Shallow-land burial
2	Yes	No	Truck	Shallow-land burial
3	No	At burial site	Truck	Shallow-land burial
4	Yes	At burial site	Truck	Shallow-land burial
5	Yes	At reproc. plant	Truck	Shallow-land burial
6	No	No	No	Hydraulic fracturing
7	Yes	At reproc. plant	Barge	Deep sea
8	Yes	At burial site	Truck	Geologic repository burial
9	Yes	At reproc. plant	Shielded truck	Shallow-land burial

The costs for the "recommended" CaCO_3 disposal options (Cases 2-5) range from \$18.47 to \$55.54/kg heavy metal. Based on the equivalency cited above, this range corresponds to about 0.022 to 0.063 mill/kWhr(e), or from 1.1 to 3.2% of a 2.0-mill/kWhr(e) fuel cycle cost. Case 1, considered to be unacceptable from a regulatory standpoint, is the most economical option, at \$15.19/kg heavy metal.

The hydraulic fracturing method of disposal is estimated to cost \$36.78/kg heavy metal based on a hydraulic fracturing cost of \$0.50 per gallon of CaCO_3 slurry. The \$0.50/gal unit cost corresponds to the lower end of the \$0.50 to \$1.00/gal cost estimate given in ref. 27. However, it is felt that the lower cost is applicable in this case because of the economies of scale inherent in injecting 5 to 10 million gallons per year.

The cost of deep-sea disposal is also relatively high (\$56.43/kg heavy metal), principally reflecting the required long-distance transportation of the large mass of concreted CaCO_3 .

The high cost of geologic repository burial (\$281.39/kg heavy metal) reflects the high burial charge (\$24.50/rectilinear ft³) for a deep geologic facility.

The unit cost of \$86.20/kg heavy metal in Case 9 gives an indication of the high cost of disposing of non-low-level material, since the material being disposed of was assumed to contain 100% of the ⁸⁵Kr originally in the discharged fuel. The cost penalties result from the additional cost of transporting heavy shielding and the additional charges for burying higher-level wastes. The disposal cost for the graphite- CaCO_3 resulting from partial block burning and concretion (Sect. 6.3) can also be expected to be above \$75/kg heavy metal, since the block has a higher activity level than that in Case 9.

There is, of course, a possibility that the CaCO_3 disposal facility may be located adjacent to the reprocessing plant. This would result in substantial savings, since the transportation cost of \$4 to \$27/kg heavy metal would be reduced to very nearly zero.

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