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ECONOMICS OF POLYSILICON PROCESSES

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ABSTRACT

New technologies are being developed to provide lower cost polysilicon material for solar cells. Existing technology which normally provides semiconductor industry polysilicon material is undergoing changes and also being used to provide polysilicon material for solar cells.

Economics of new and existing technologies are presented for producing polysilicon. The economics are primarily based on the preliminary process design of a plant to produce 1,000 metric tons/year of silicon. The polysilicon processes include: Siemens process (hydrogen reduction of trichlorosilane); Union Carbide process (silane decomposition) and Hemlock Semiconductor process (hydrogen reduction of dichlorosilane). The economics include cost estimates of capital investment and product cost to produce the polysilicon via the technology. Sensitivity analysis results are also presented to disclose the effect of major parameters such as utilities, labor, raw materials and capital investment.

## 1. INTRODUCTION

A typical sequence for process selection is presented in Figure 1. The process evaluation activities are shown in relation to their usefulness in the comparison of processes and in the selection for scale-up to pilot plant and large scale plant. The process evaluation activities which primarily involve chemical engineering and economic analyses are useful in the evaluation of alternate processes under consideration for the production of polysilicon. Specifically, the process evaluation provides technical and economic data which may be used for the identification of those processes having good potential for producing polysilicon within the cost goals of the project.

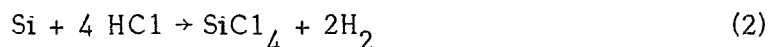
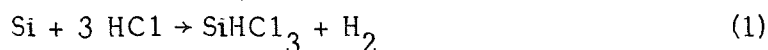
The objective of the present work is to provide initial economics for several processes for the production of polysilicon. The results are intended to be semi-quantitative and useful in initial project studies.

## 2. SIEMENS PROCESS

### Process Description And Design

The flowsheet (1, 2, 33, 34, 36, 39, 41-43, 45) for the Siemens process, consisting of several major processing operations of hydrochlorination, condensation, distillation and chemical vapor deposition, is shown in Figure 2.

Initially, metallurgical grade silicon (MGSi) is reacted with anhydrous hydrogen chloride (HCl) in a fluidized bed (300-350°C) to produce a mixture of chlorosilanes. The mixture is primarily trichlorosilane (TCS) and silicon tetrachloride (TET) which are produced via the representative reactions:



Since the reactions are highly exothermic, heat transfer for removal of heat of reaction is required to maintain reaction temperature control.

The mixture of chlorosilanes from the reaction is condensed and subjected to several distillations to separate by-products and remove impurities. Representative results for boron impurity removal from TCS are shown in Figure 2A. Figure 2B presents representative results for phosphorous impurity removal.

The purified TCS is reacted with hydrogen ( $\text{H}_2$ ) in a rod reactor to obtain polysilicon deposition via the representative reaction:



The deposition reaction occurs on the surface of a hot rod (1000-1100°C) which is heated by passage of electrical current through the rod. Large electrical energy requirements are necessary because of the endothermic reaction, radiation heat

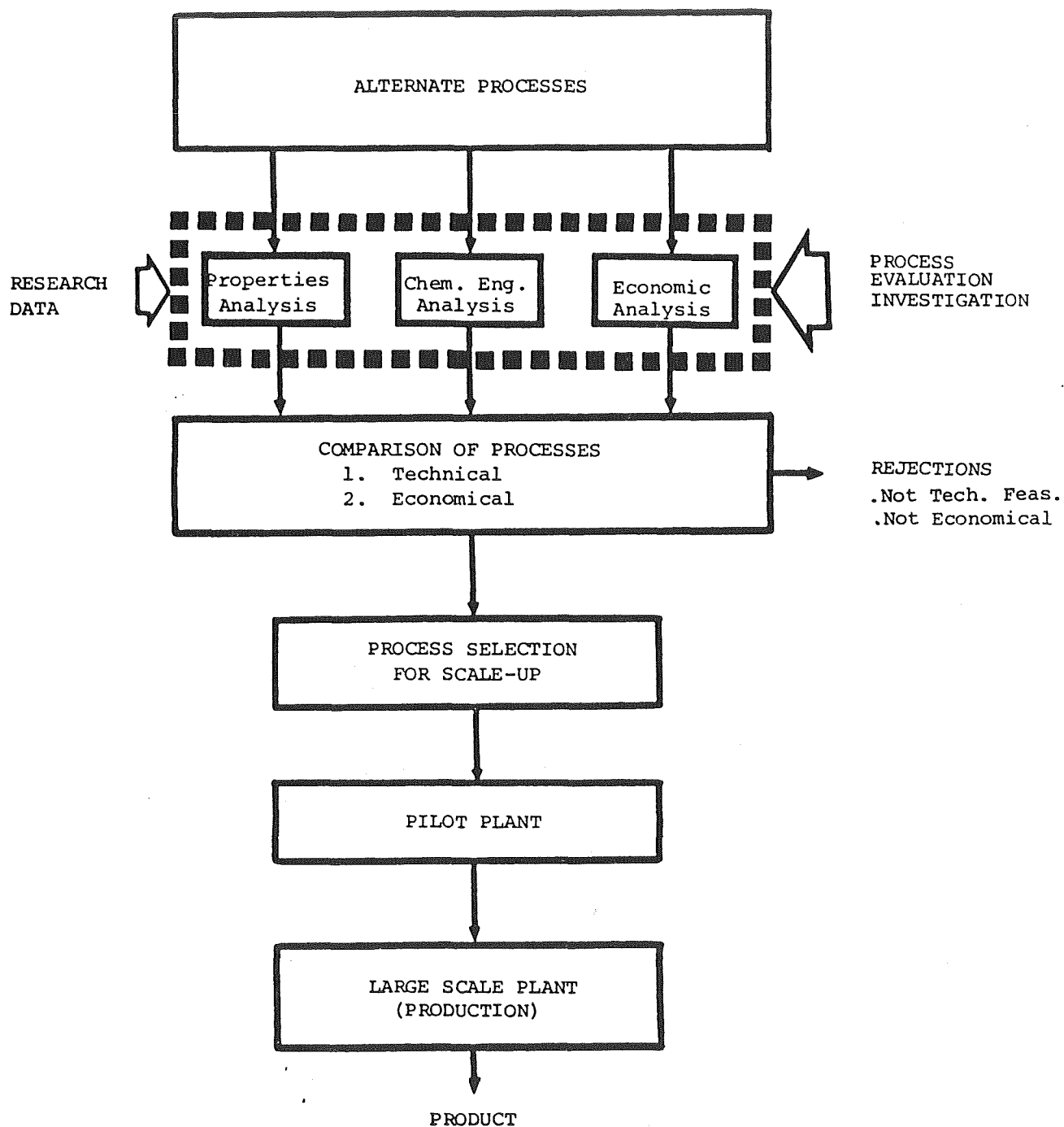


FIGURE 1. TYPICAL SEQUENCE FOR PROCESS SELECTION

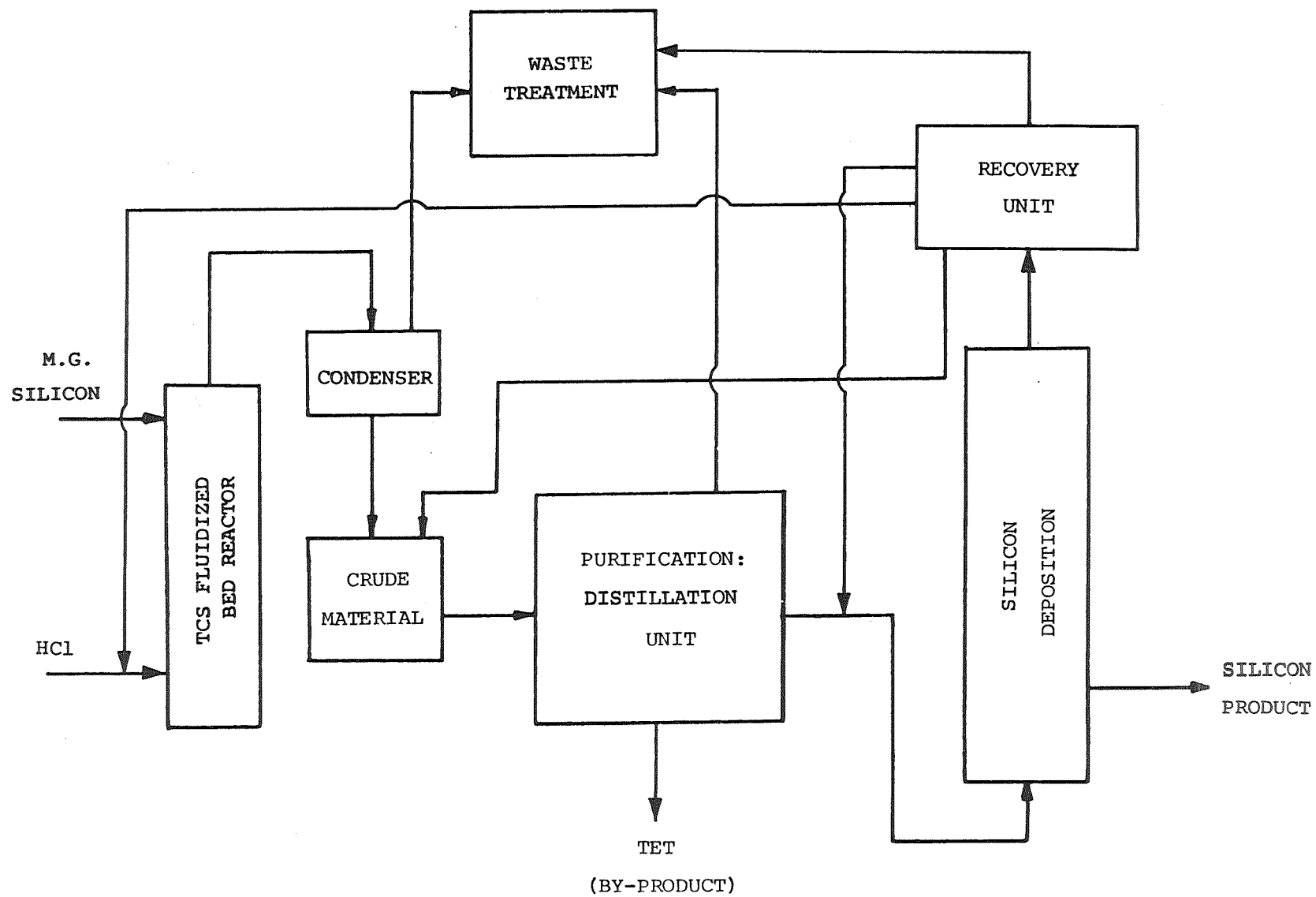


FIGURE 2. FLOWSHEET FOR SIEMENS PROCESS

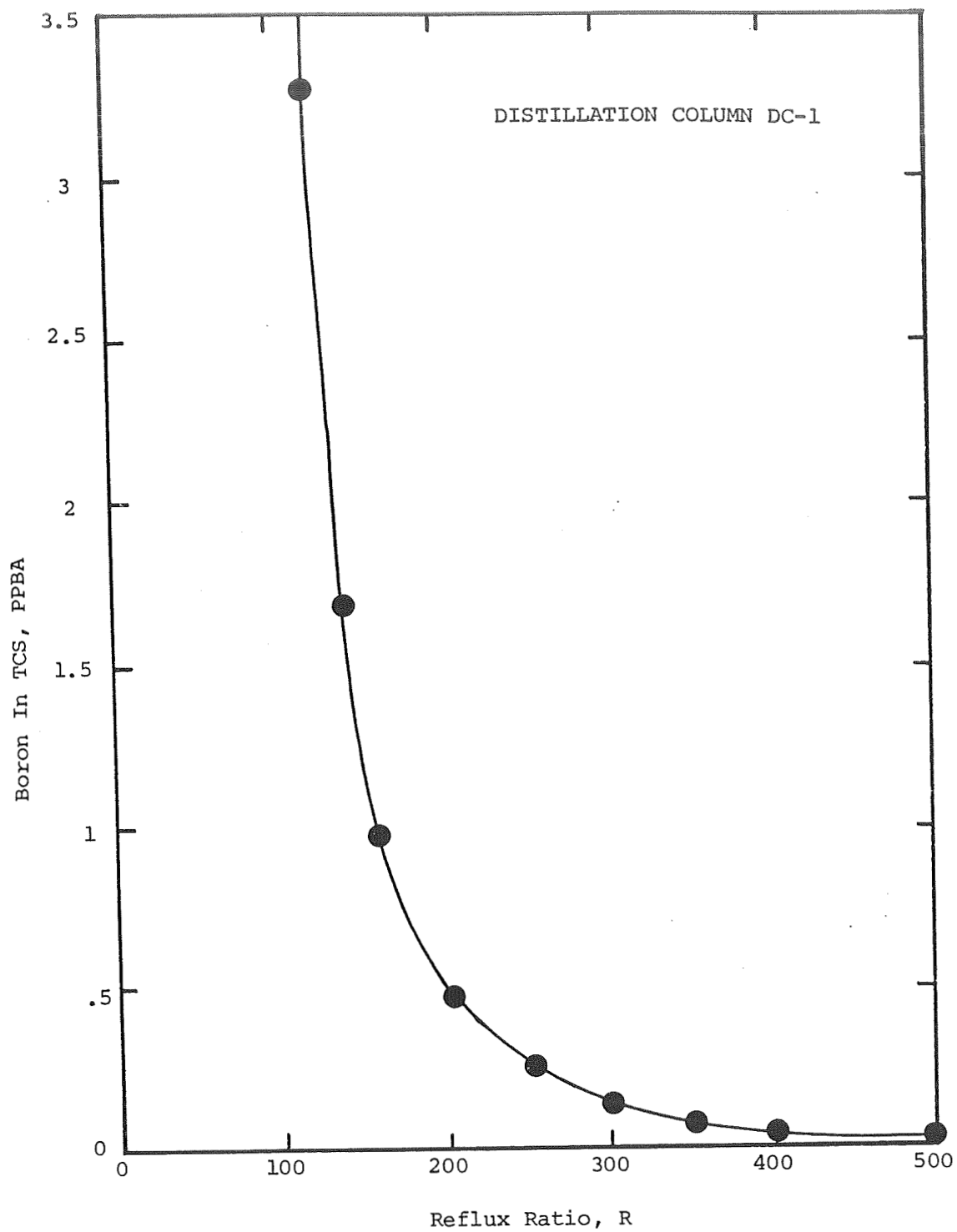


FIGURE 2A. REPRESENTATIVE RESULTS FOR BORON IMPURITY REMOVAL

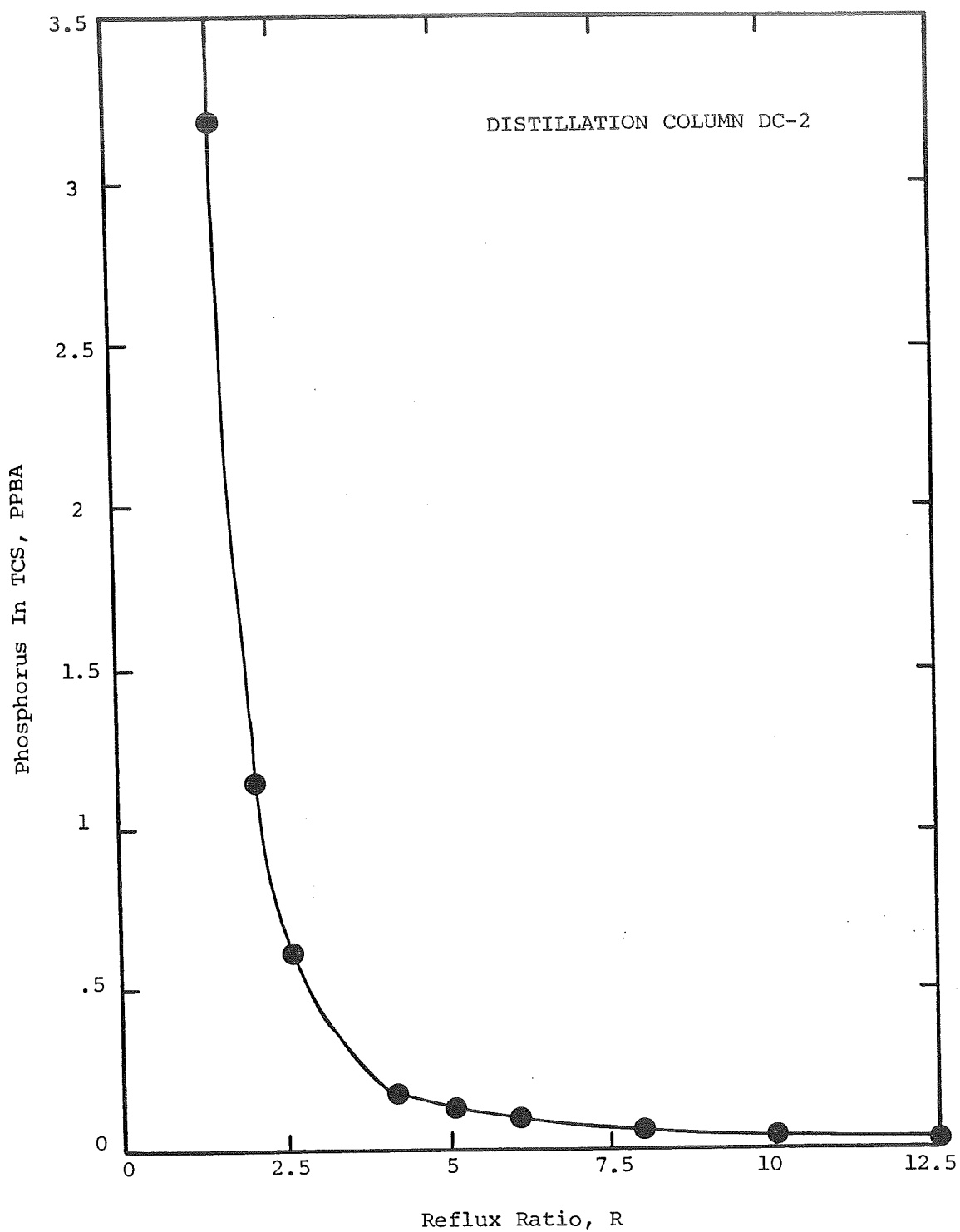


FIGURE 2B. REPRESENTATIVE RESULTS FOR PHOSPHORUS IMPURITY REMOVAL

losses and incomplete conversion of the TCS. Unreacted chlorosilanes, hydrogen chloride and hydrogen are separated and recycled.

A representative polysilicon deposition reactor using trichlorosilane as the silicon source material is shown in Figure 3.

In the chemical engineering analysis of the process, a process design was performed to obtain data for the cost analysis. The design was based on a plant for the production of 1,000 metric tons/yr of polysilicon via the Siemens process. The detailed design included TCS production in a fluidized bed; TCS purification by distillation; silicon production by chemical vapor deposition in a Siemens type rod reactor; recycle of chlorosilanes, hydrogen chloride, and hydrogen; waste treatment provisions to meet environmental quality; and storage considerations for feed, in-process and product materials.

The process design provided detailed data for raw materials, utilities, major process equipment and production labor requirements which are necessary for polysilicon production.

### Cost Analysis

The cost analysis results for producing silicon by this technology are presented in Table 1 including costs for raw materials, labor, utilities and other items composing the product cost (total cost of producing silicon). The tabulation summarizes all of these items to give a total product cost without profit of 29.49 \$/kg Si (1985 dollars). This product cost without profit includes direct manufacturing cost, indirect manufacturing cost, plant overhead and general expenses.

The economic summary for the process is given in Table 2. Results for process, plant size, plant product, plant investment, profitability analysis and sensitivity analysis are displayed in the tabulation.

A sensitivity analysis was performed to determine the influence of cost parameters on the economics of producing silicon by this technology. The cost sensitivity results are given in Figure 4 in which product cost (\$/kg Si) is plotted vs variation (-100 to 0 to +100 percent) of the primary cost parameters (raw materials, labor and utilities). The 0 per cent variation represents the base case. The -100 per cent variation corresponds to the case of no costs for the parameter; and the +100 per cent represents the case for a doubling of cost for each parameter. The plot illustrates that product cost is greatly influenced by utilities (electrical energy).

The variation of product cost (\$/kg Si) with electrical energy requirements (kw-hr/kg Si) is shown in Figure 5. The present study which is based on electrical energy requirements of 120 kw-hr/kg of Si and 5 \$/kw-hr is shown as the darkened circle in the figure. If electrical energy requirements are increased from 120 to 220 kw-hr, the product cost increases from \$29.5 to \$34.5 per kg of Si. The increase is even more pronounced at 7.5 \$/kw-hr electricity.

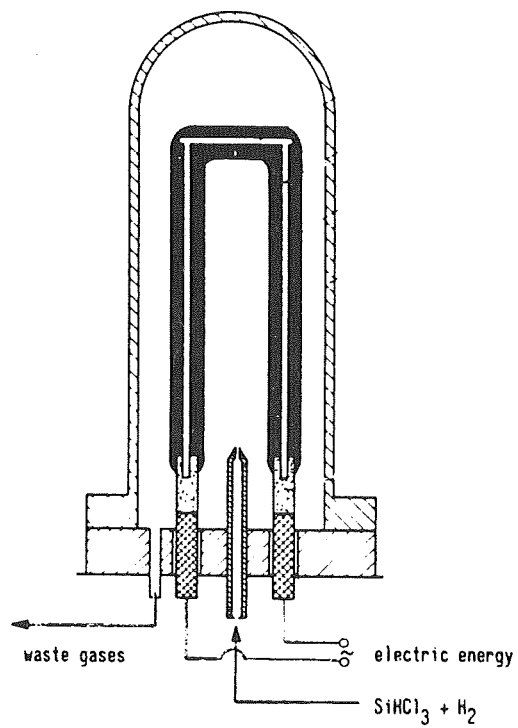


FIGURE 3. REPRESENTATIVE POLYCRISTALLINE DEPOSITION REACTION FOR TRICHLOROSILANE  
( GOVERNMENT REPORT: CISZEK (2) )



TABLE 1

## ESTIMATION OF PRODUCT COST FOR SIEMENS PROCESS

	COST \$/kg of Si -----
1. Direct Manufacturing Cost.....	16.09
Raw Materials	
Direct Operating Labor	
Utilities	
Supervision and Clerical	
Maintenance and Repairs	
Operating Supplies	
Laboratory Charge	
2. Indirect Manufacturing Cost.....	7.80
Depreciation	
Local Taxes	
Insurance	
3. Plant Overhead.....	1.76
4. General Expenses.....	3.85
Administration	
Distribution and Sales	
Research and Development	
	-----
5. Product Cost without Profit.....	29.49

Note: 1985 Dollars

TABLE 2

## ECONOMIC SUMMARY: COST ANALYSIS FOR SIEMENS PROCESS

1. Process ----- SIEMENS PROCESS
2. Plant Size ----- 1000 MT/yr
3. Plant Product ----- POLYSILICON
4. Plant Investment ----- \$ 69.00 Million

Fixed Capital ---- \$ 60.00 Million  
 Working Capital -- \$ 9.00 Million  
 Total Capital ---- \$ 69.00 Million

## 5. Profitability Analysis

Return on Original Investment after Taxes (% ROI)  
 Discounted Cash Flow Rate of Return after Taxes (% DCF)

Return	Sales Price \$/kg of Si	Return	Sales Price \$/kg of Si
-----	-----	-----	-----
0 % ROI	29.49	0 % DCF	29.49
10 % ROI	42.27	10 % DCF	38.13
20 % ROI	55.05	20 % DCF	48.22
30 % ROI	67.83	30 % DCF	59.32
40 % ROI	80.60	40 % DCF	71.08
50 % ROI	93.38	50 % DCF	83.25
60 % ROI	106.16	60 % DCF	95.66
70 % ROI	118.94	70 % DCF	108.21
80 % ROI	131.71	80 % DCF	120.85
90 % ROI	144.49	90 % DCF	133.54
100 % ROI	157.27	100 % DCF	146.27

Based on 10 year project life and 10 year straight  
 line depreciation. Tax rate (federal) of 46 %.

## 6. Sensitivity Analysis

	Product Cost, \$/kg of Si					
	-100%	-50%	BASE	+50%	+100%	DELTA
	-----	-----	-----	-----	-----	-----
Raw Materials	25.40	27.45	29.49	31.54	33.58	4.09
Labor	27.60	28.54	29.49	30.44	31.39	1.90
Utilities	21.76	25.63	29.49	33.36	37.22	7.73
Plant Investment (Fixed Capital)	17.93	23.71	29.49	35.27	41.05	11.56

Note: 1985 Dollars

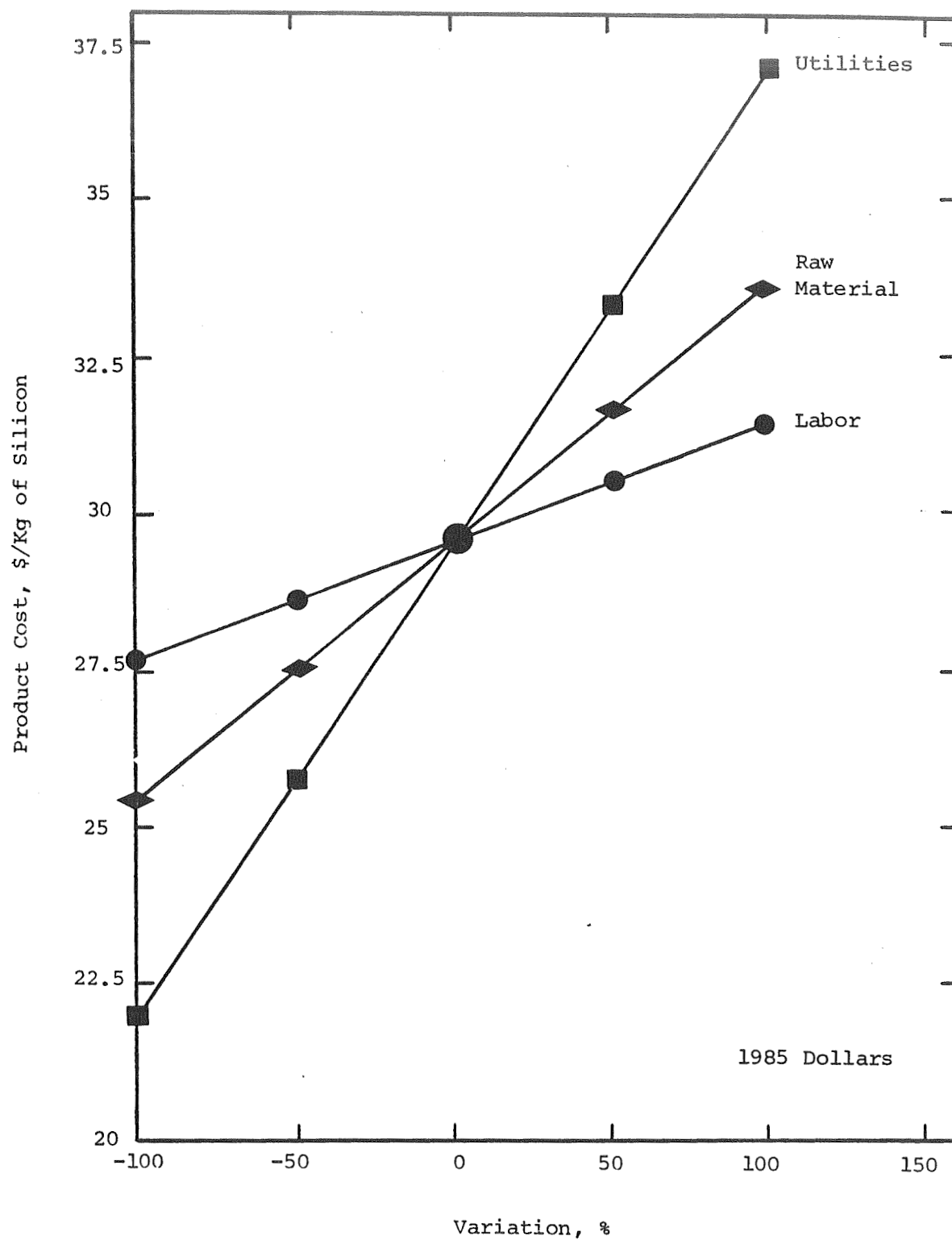


FIGURE 4. SENSITIVITY PLOT FOR SIEMENS PROCESS

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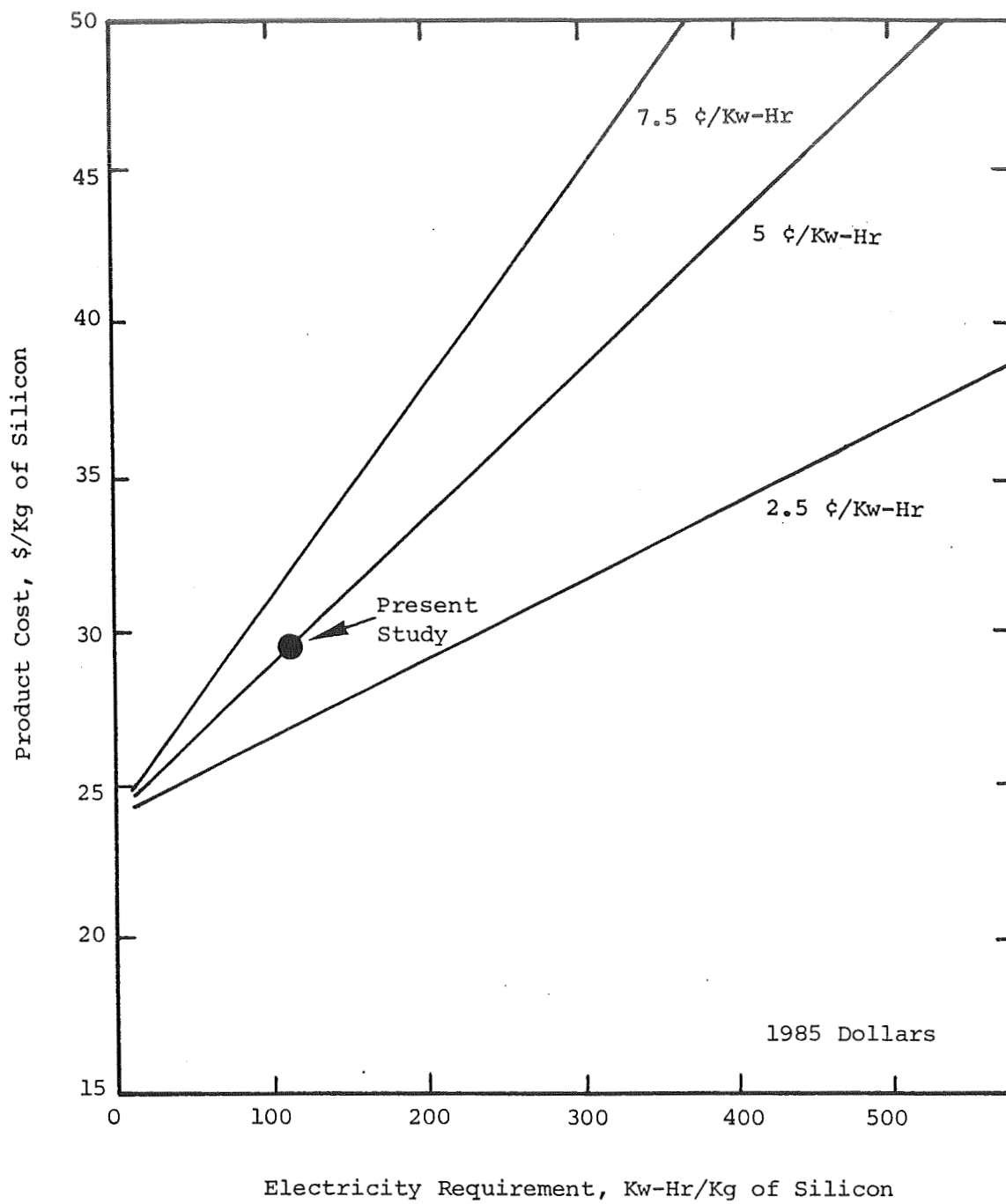


FIGURE 5. PRODUCT COST VS. ELECTRICITY REQUIREMENTS FOR SIEMENS PROCESS

## 2. UNION CARBIDE PROCESS

### Process Description And Design

The Union Carbide process (7-9, 27, 36, 39) for silicon involves several processing operations of hydrogenation-hydrochlorination reaction, stripping, distillation, redistribution reaction, silane purification, and silicon deposition. The process flowsheet is shown in Figure 6.

Hydrogen, silicon tetrachloride, and metallurgical grade silicon are fed to the hydrogenation reactor (fluidized bed, 500C, 515 psia, copper catalyst) to produce a mixture of chlorosilanes. The mixture of chlorosilanes from the hydrogenation reaction is condensed and subjected to several distillations to separate components and remove impurities.

Initially, the condensed liquid mixture is sent to D-01 stripper (90 psia) to remove inert gases and volatile impurities. The stripper bottoms go to D-02 distillation (55 psia) which separates TCS (trichlorosilane) and TET (silicon tetrachloride). The TCS redistribution reactor (liquid phase, 85 psia, 140°F catalyst) is used to produce DCS (dichlorosilane). The separation of DCS and TCS is achieved in D-03 distillation (320 psia). The overhead goes to DCS redistribution reactor (liquid phase, 510 psia, 140°F, catalyst) to produce silane ( $\text{SiH}_4$ ). The silane is purified by separation from trace impurities (such as  $\text{B}_2\text{H}_6$ ) by D-04 distillation (355 psia). Representative results for diborane impurity removal are shown in Figure 6A.

The purified silane is mixed with hydrogen and then introduced into the deposition reactor (Komatsu license, 7) to produce silicon via the representative reaction:



The reaction occurs in a deposition reactor which is heated by passage of electrical current through the silicon rods to attain a temperature in the 800-900C range (21, 22). Deposition rates of 4-8 micrometers/min of silicon on the rod surface are reported in the Komatsu patents.

For the deposition reactor, the homogeneous decomposition reaction resulting in silicon dust formation is not desirable. The heterogeneous decomposition reaction resulting in silicon deposition on the rod surface is desirable. The temperature dependence of critical silane concentration for homogeneous and heterogeneous decomposition regions has been studied by Iya (20) and others (28). Results from the recent report of Dudukovic (5) are shown in Figure 7. At 800C, the heterogeneous decomposition region appears to be in the 1-2% concentration range for silane in hydrogen.

A representative polysilicon deposition reactor for silane is shown in Figure 8 (Komatsu patent: 21, 22). The thermal insulator in the reaction chamber is obvious in both the side and top views. The thermal insulators provide for radiation benefits (individual silicon rods from radiation from the other red-heated silicon rods) and for temperature control benefits for reduction of homogeneous reaction in the gas phase.

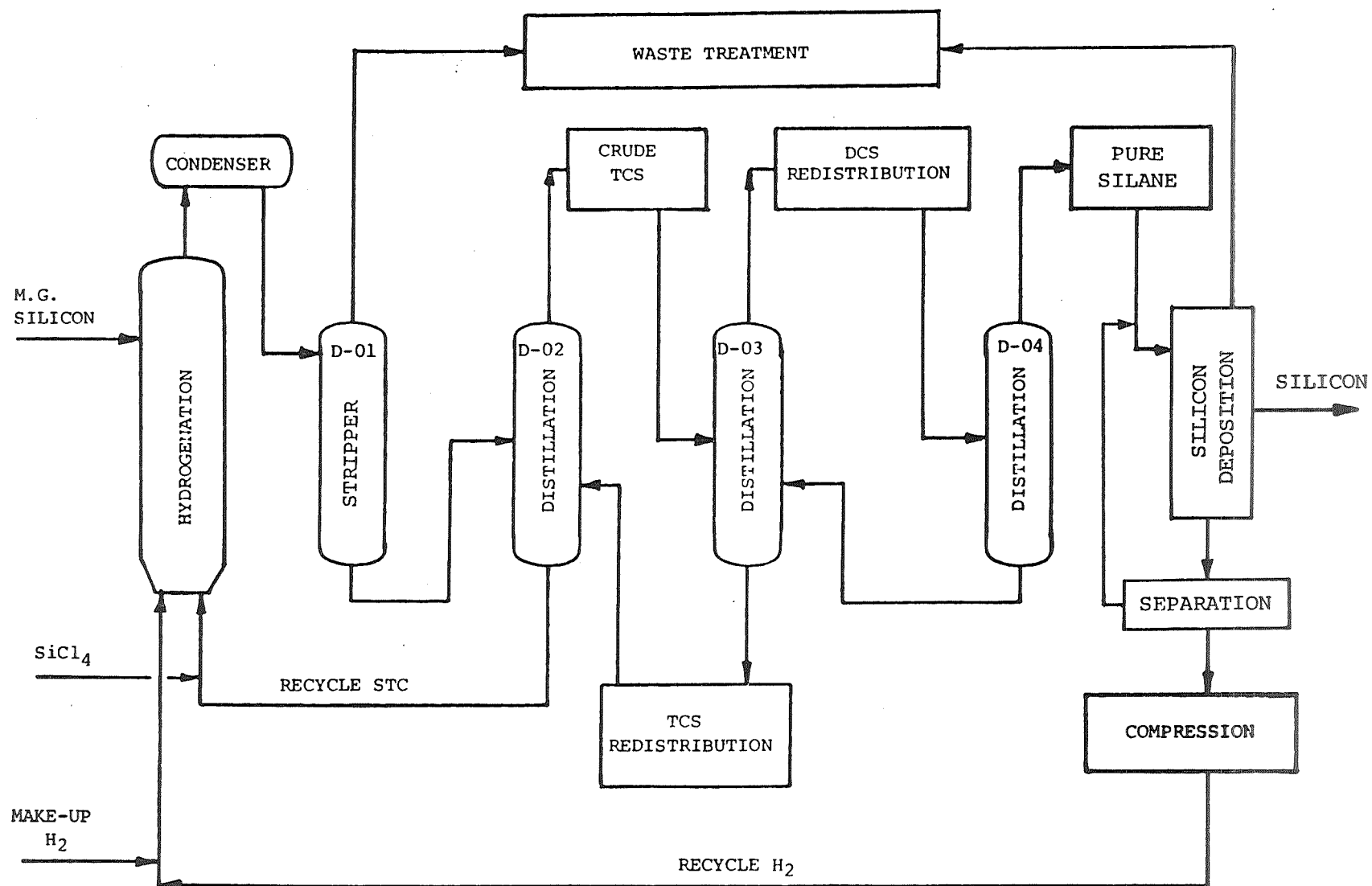


FIGURE 6. FLOWSHEET FOR UNION CARBIDE PROCESS

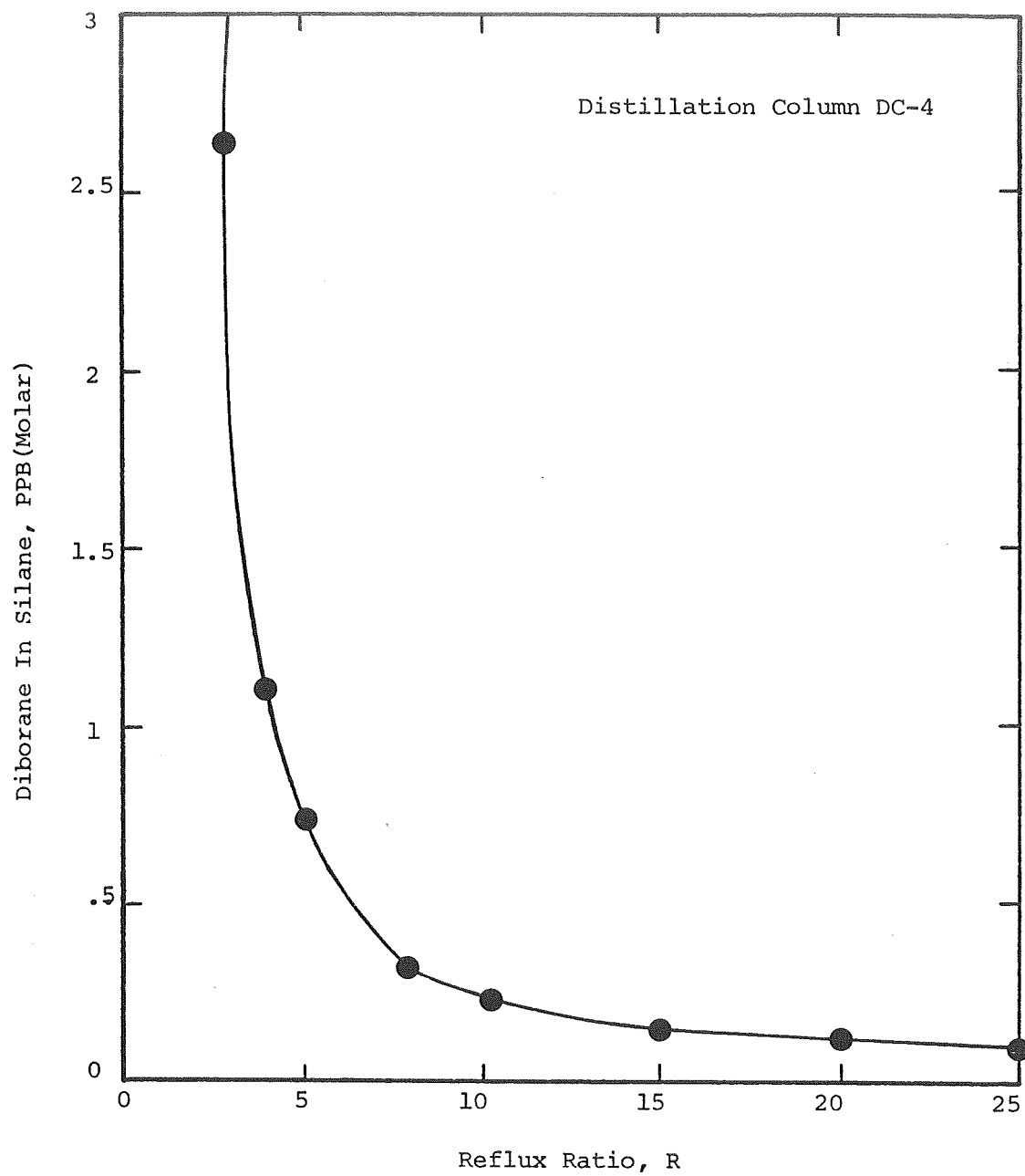


FIGURE 6A REPRESENTATIVE RESULTS FOR DIBORANE IMPURITY REMOVAL

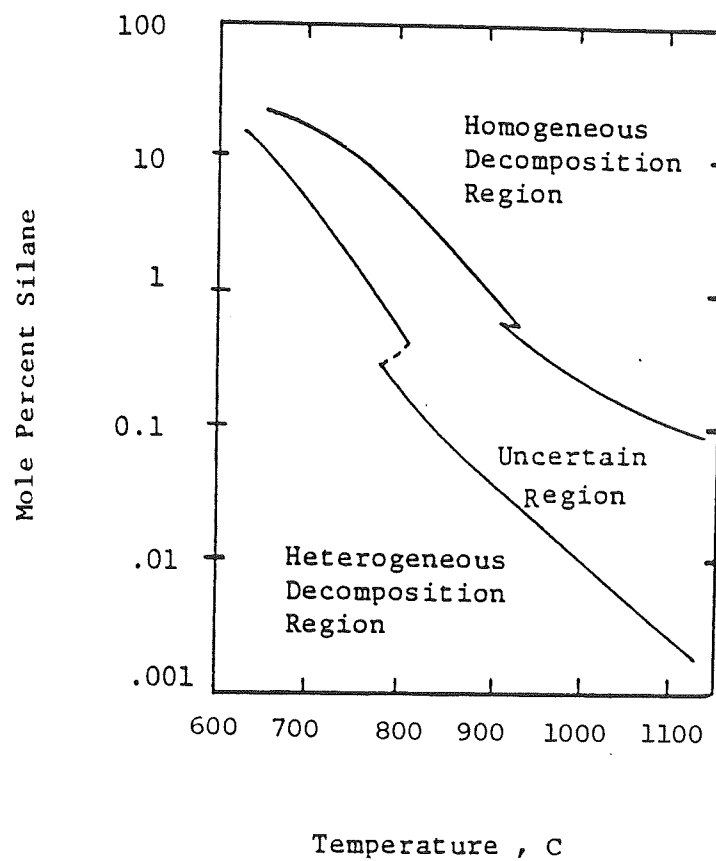


FIGURE 7. TEMPERATURE DEPENDENCE OF CRITICAL SILANE CONCENTRATION  
( GOVERNMENT REPORT: IYA (20), DUDUKOVIC (5) )



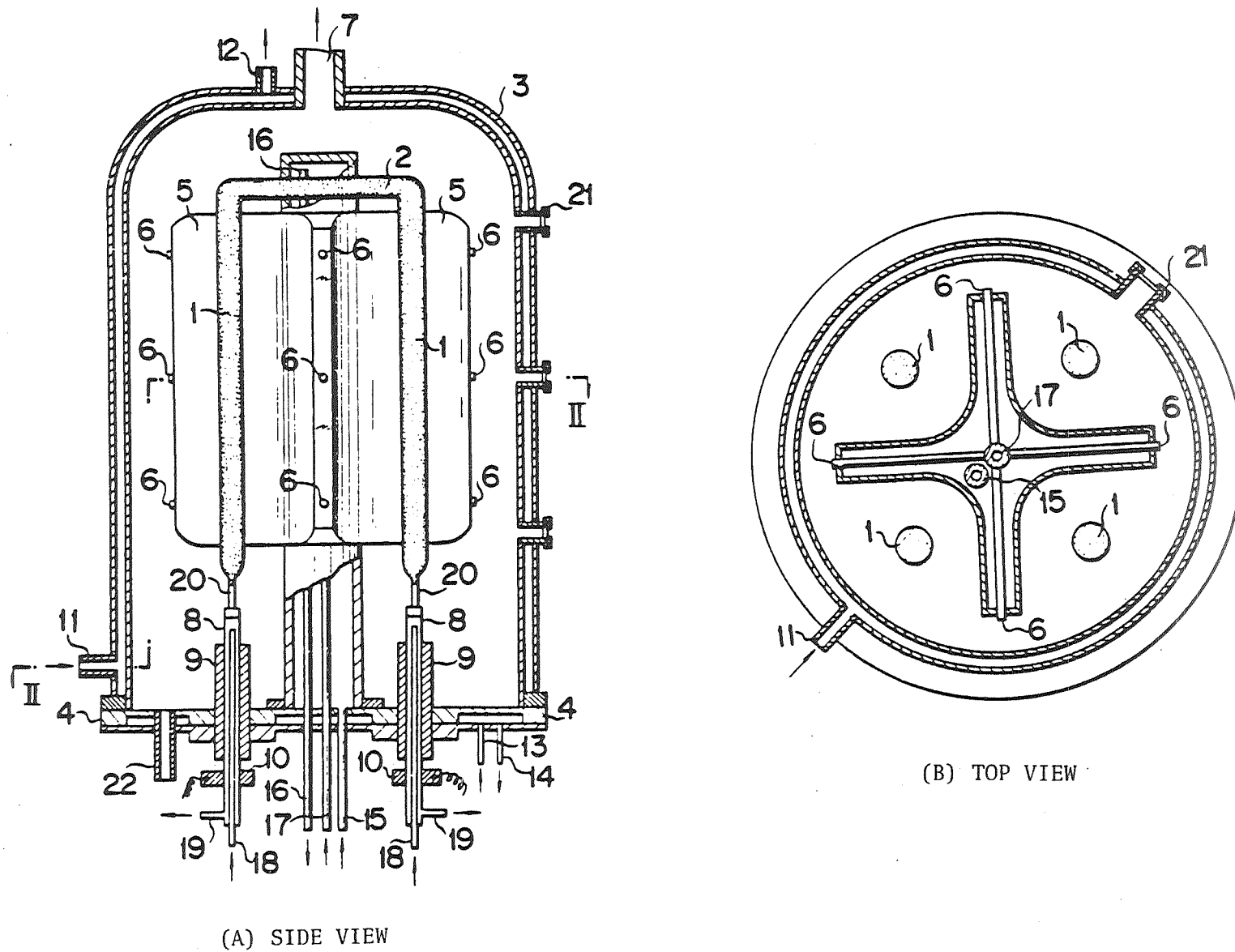


FIGURE 8. REPRESENTATIVE POLYCRYSTALLINE SILICON DEPOSITION REACTOR FOR SILANE  
 ( KOMATSU: U.S. PATENT 4,150,168 (REF. 22 ) )

In the chemical engineering analysis of the Union Carbide process, a process design was performed for a plant to produce 1,000 metric tons/yr of polysilicon.

### Cost Analysis

Cost analysis results for producing polysilicon via the Union Carbide process with chemical vapor deposition reactors are given in Table 3. The results indicate a product cost without profit of 24.65 \$/kg Si (1985 dollars).

Table 4 presents the economic summary of results for process, plant size, plant product, plant investment, profitability analysis and sensitivity analysis. The capital investment of \$74.75 million of the present study for 1,000 metric ton plant is in the range of the \$85 million for a 1,500 metric ton plant reported by Union Carbide in a news release (9) and \$90 million reported for a doubling of capacity to 2,400 metric tons (8).

The sensitivity analysis plot is shown in Figure 9. The product cost is influenced most by utilities (electrical energy).

## 4. HEMLOCK SEMICONDUCTOR PROCESS

### Process Description and Design

The process flowsheet for Hemlock Semiconductor process for polysilicon is shown in Figure 10. The process involves major processing operations of hydrochlorination, separation, several distillation units, redistribution, boron removal, silicon deposition, recovery unit and waste treatment.

Metallurgical grade silicon is hydrochlorinated in the presence of hydrogen and silicon tetrachloride in a fluidized bed reactor (500C, 515 psia). In the process, the reaction product issuing from the hydrochlorination reactor (hydrochlorination-hydrogenation reaction) is cooled and undergoes a vapor-liquid flash separation. The vapor fraction containing the hydrogen from the flash is recycled back to the hydrochlorination reactor. The liquid fraction containing the chlorosilanes and dissolved gases is fed to the initial distillation column.

The function of the initial distillation column (D-01, stripper column, 90 psia) in the process is to remove volatile gases (such as hydrogen and nitrogen) which are dissolved in liquid chlorosilanes. For the engineering design, TCS (trichlorosilane) was selected as the heavy key component for the separation.

The second distillation column (D-02, TCS column, 90 psia) in the process separates TCS (trichlorosilane) and TET (silicon tetrachloride). The distillation column has three feeds (bottoms from the third distillation, chlorosilanes from the recovery unit and bottoms from the initial distillation). The TET from the distillation is recycled to the hydrochlorination reactor for additional conversion.

TABLE 3

## ESTIMATION OF PRODUCT COST FOR UNION CARBIDE PROCESS

	COST \$/kg of Si -----
1. Direct Manufacturing Cost.....	11.19
Raw Materials	
Direct Operating Labor	
Utilities	
Supervision and Clerical	
Maintenance and Repairs	
Operating Supplies	
Laboratory Charge	
2. Indirect Manufacturing Cost.....	8.45
Depreciation	
Local Taxes	
Insurance	
3. Plant Overhead.....	1.80
4. General Expenses.....	3.21
Administration	
Distribution and Sales	
Research and Development	
	-----
5. Product Cost without Profit.....	24.65

Note: 1985 Dollars

TABLE 4

## ECONOMIC SUMMARY: COST ANALYSIS FOR UNION CARBIDE PROCESS

1. Process -----	UNION CARBIDE PROCESS
2. Plant Size -----	1000 MT/yr
3. Plant Product -----	POLYSILICON
4. Plant Investment -----	\$ 74.75 Million

Fixed Capital ----	\$ 65.00 Million
Working Capital --	\$ 9.75 Million
Total Capital ----	\$ 74.75 Million

## 5. Profitability Analysis

Return on Original Investment after Taxes (% ROI)  
Discounted Cash Flow Rate of Return after Taxes (% DCF)

Return	Sales Price \$/kg of Si	Return	Sales Price \$/kg of Si
-----	-----	-----	-----
0 % ROI	24.65	0 % DCF	24.65
10 % ROI	38.49	10 % DCF	34.00
20 % ROI	52.33	20 % DCF	44.93
30 % ROI	66.17	30 % DCF	56.96
40 % ROI	80.02	40 % DCF	69.70
50 % ROI	93.86	50 % DCF	82.88
60 % ROI	107.70	60 % DCF	96.33
70 % ROI	121.54	70 % DCF	109.93
80 % ROI	135.39	80 % DCF	123.62
90 % ROI	149.23	90 % DCF	137.37
100 % ROI	163.07	100 % DCF	151.15

Based on 10 year project life and 10 year straight  
line depreciation. Tax rate (federal) of 46 %.

## 6. Sensitivity Analysis

	Product Cost, \$/kg of Si					
	-100%	-50%	BASE	+50%	+100%	DELTA
	-----	-----	-----	-----	-----	-----
Raw Materials	21.74	23.19	24.65	26.10	27.55	2.91
Labor	22.75	23.70	24.65	25.59	26.54	1.90
Utilities	20.78	22.71	24.65	26.58	28.51	3.87
Plant Investment (Fixed Capital)	12.13	18.39	24.65	30.91	37.17	12.52

Note: 1985 Dollars

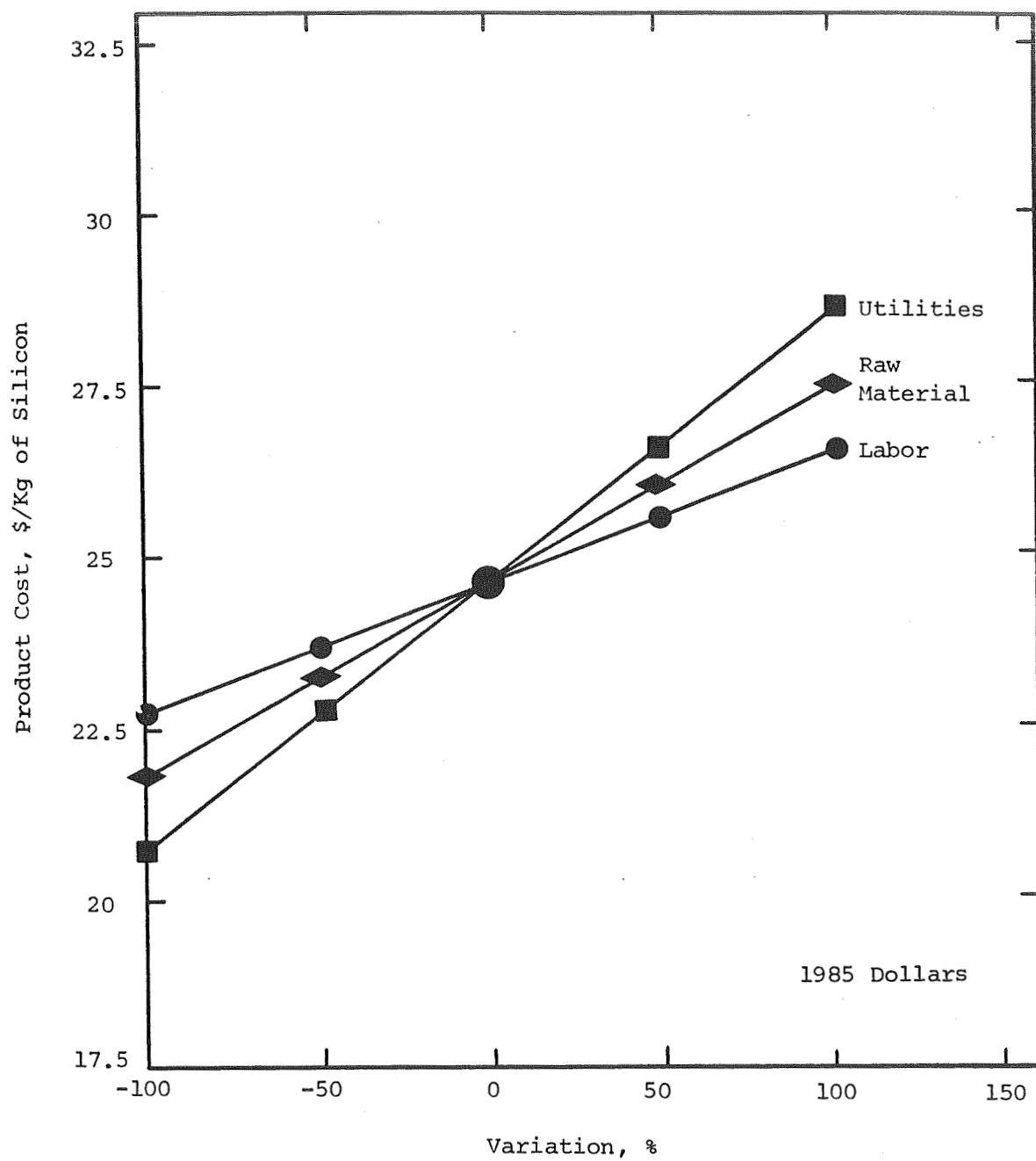


FIGURE 9. SENSITIVITY PLOT FOR UNION CARBIDE PROCESS

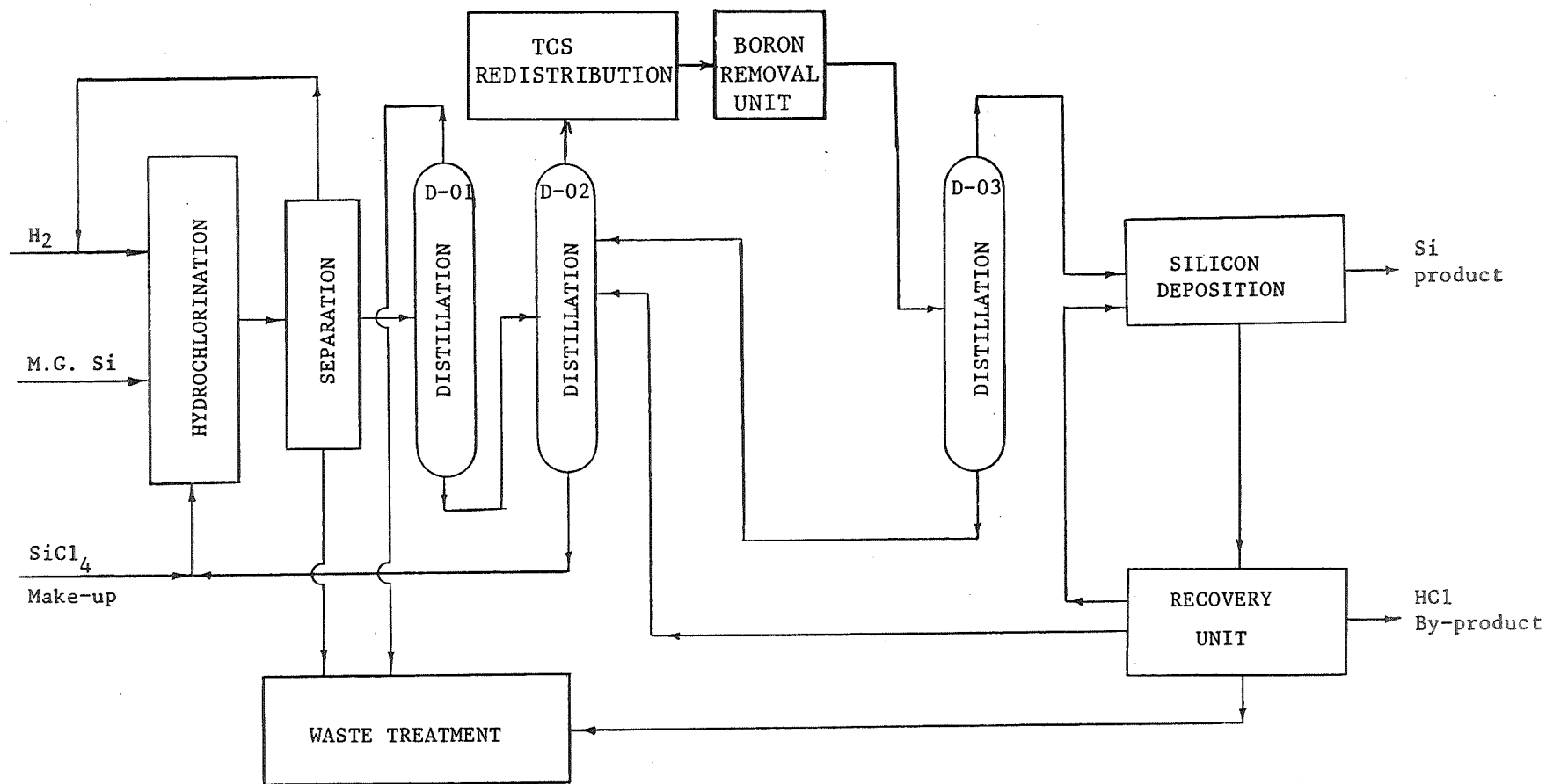
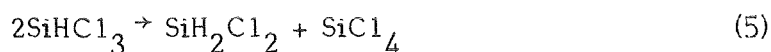


FIGURE 10. FLOWSHEET FOR HEMLOCK SEMICONDUCTOR PROCESS

The TCS from the second distillation is sent to the redistribution reactor (liquid phase, 80 psia, 80C) where TCS is redistributed to DCS and TET according to the representative chemical reaction equation:



The conversion from pure TCS is about 10.5% to DCS.

After redistribution the stream is sent to the boron removal unit and the third distillation. The third distillation column (D-03, DCS column, 90 psia) in the process separates DCS (dichlorosilane) and TCS (trichlorosilane). DCS from the distillation is sent to the silicon deposition reactors.

The purified DCS is reacted with hydrogen ( $\text{H}_2$ ) in a rod reactor to obtain polysilicon deposition via the following representative chemical reaction equation:



The above reaction equation may include several reaction steps. Chemical equilibrium is involved and in reality, several chlorosilanes (such as  $\text{SiH}_2\text{Cl}_2$ ,  $\text{SiHCl}_3$  and  $\text{SiCl}_4$ ) are also present in the gas phase by-products.

The chemical vapor deposition reaction with DCS is very fast and occurs on the surface of a hot rod (1000-1200C) which is heated by passage of electrical current through the rod. Deposition rates and conversions for dichlorosilane are approximately twice (2X) those for the usual trichlorosilane process. Other benefits include higher molar silicon conversion and lower power consumption (25, 26).

In this process using dichlorosilane as the silicon source material, wall deposits resulting from the dichlorosilane deposition reaction are not desirable. Representative bell jar silicon deposition is shown in Figure 11. The homogeneous gas phase reaction resulting in the formation of solid silicon particles (not on the rod surface) is also not desirable. The reduction of wall deposits has been investigated by Hemlock Semiconductor (26). One approach involved an advanced decomposition reactor with a cool bell jar temperature (300C) as compared with hot bell jar reactor (750C). The study also encompassed screening of lower cost materials of construction such as stainless steels and other metallic alloys.

A process design was performed to obtain data for a cost analysis of a process plant to produce 1,000 metric tons/yr of polysilicon via the Hemlock Semiconductor process.

### Cost Analysis

Cost analysis results for producing polysilicon by the Hemlock Semiconductor process are displayed in Table 5 including raw materials, labor, utilities and other items. A total product cost without profit of 19.48 \$/kg Si (1985 dollars) is indicated.

The economic summary for the process is provided in Table 6 including plant investment, profitability analysis and sensitivity analysis.

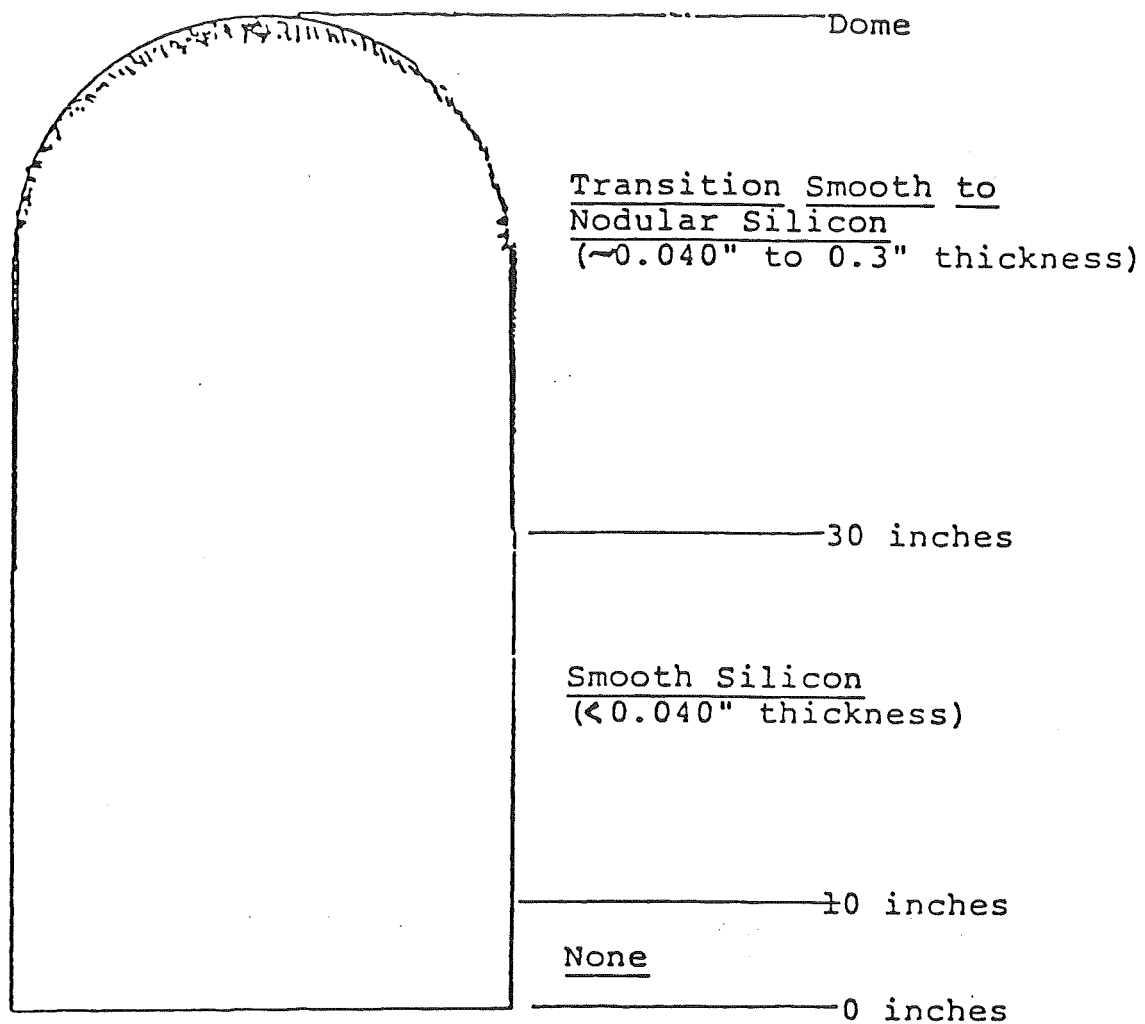


FIGURE 11. REPRESENTATIVE BELL JAR SILICON DEPOSITION ( GOVERNMENT REPORT:HEMLOCK SEMICONDUCTOR, REF.25, PAGE 79 )



TABLE 5

## ESTIMATION OF PRODUCT COST FOR HEMLOCK SEMICONDUCTOR PROCESS

	COST \$/kg of Si -----
1. Direct Manufacturing Cost.....	11.05
Raw Materials	
Direct Operating Labor	
Utilities	
Supervision and Clerical	
Maintenance and Repairs	
Operating Supplies	
Laboratory Charge	
2. Indirect Manufacturing Cost.....	4.68
Depreciation	
Local Taxes	
Insurance	
3. Plant Overhead.....	1.21
4. General Expenses.....	2.54
Administration	
Distribution and Sales	
Research and Development	
	-----
5. Product Cost without Profit.....	19.48

Note: 1985 Dollars

TABLE 6

## ECONOMIC SUMMARY: COST ANALYSIS FOR HEMLOCK SEMICONDUCTOR PROCESS

1. Process -----	HEMLOCK SEMICONDUCTOR PROCESS
2. Plant Size -----	1000 MT/yr
3. Plant Product -----	POLYSILICON
4. Plant Investment -----	\$ 41.40 Million

Fixed Capital ---- \$ 36.00 Million

Working Capital -- \$ 5.40 Million

Total Capital ---- \$ 41.40 Million

## 5. Profitability Analysis

Return on Original Investment after Taxes (% ROI)

Discounted Cash Flow Rate of Return after Taxes (% DCF)

Return	Sales Price \$/kg of Si	Return	Sales Price \$/kg of Si
-----	-----	-----	-----
0 % ROI	19.48	0 % DCF	19.48
10 % ROI	27.15	10 % DCF	24.67
20 % ROI	34.82	20 % DCF	30.72
30 % ROI	42.48	30 % DCF	37.38
40 % ROI	50.15	40 % DCF	44.44
50 % ROI	57.82	50 % DCF	51.74
60 % ROI	65.48	60 % DCF	59.18
70 % ROI	73.15	70 % DCF	66.72
80 % ROI	80.82	80 % DCF	74.30
90 % ROI	88.48	90 % DCF	81.91
100 % ROI	96.15	100 % DCF	89.55

Based on 10 year project life and 10 year straight line depreciation. Tax rate (federal) of 46 %.

## 6. Sensitivity Analysis

	Product Cost, \$/kg of Si					
	-100%	-50%	BASE	+50%	+100%	DELTA
	-----	-----	-----	-----	-----	-----
Raw Materials	16.15	17.81	19.48	21.15	22.82	3.34
Labor	18.11	18.80	19.48	20.17	20.85	1.37
Utilities	14.63	17.06	19.48	21.91	24.34	4.85
Plant Investment (Fixed Capital)	12.55	16.02	19.48	22.95	26.42	6.93

Note: 1985 Dollars

The sensitivity analysis results for primary cost parameters is displayed in Figure 12. The product cost is influenced much by utilities (electrical energy).

## 5. COST COMPARISON

A cursory cost comparison for capital investment is made in Figure 13 for the Siemens process. In 1980, Wacker (12) announced a \$13 million capital investment for a 500 MT/yr polysilicon plant expansion (1,200–1,300 to 1,800 MT/yr). The first block in the figure shows this data. The second block with a capital investment of \$31.2 million represents this data adjusted to 1,000 MT/yr expansion and 1985 dollars. The third block presents the present study. The capital investment of \$69 million (complete plant) of the present study is higher than the \$31.2 million (plant expansion).

Results for capital investment for the Union Carbide process are displayed in Figures 14 and 15. In 1984, Union Carbide (9) announced its plans to start silane production in its \$85 million polysilicon facility (Moses Lake, Washington) capable of producing 1,500 MT/yr. The first block in Figure 14 presents this data. The second block shows the adjusted data of \$56.7 million (1,000 MT/yr, 1985 dollars). The result of \$74.8 million (1,000 MT/yr, 1985 dollars) for the present study is higher. In 1985, capital investment of \$90 million was reported (8) to double the capacity of the polysilicon facility to 2,400 MT annually. The adjusted value for a 1,000 MT/hr is about \$75 million (plant expansion). The adjusted value is about the same as the result of \$74.75 (complete plant) of the present study.

Capital investment results for the Hemlock Semiconductor process are presented in Figure 16. A capital investment of \$25.21 million (1980 dollars) for 1,000 MT/yr polysilicon plant using dichlorosilane as the silicon source material is given in the Hemlock Semiconductor report (25). The adjusted value is \$30.25 million (1985 dollars). The results of \$41.40 million (1985 dollars) of the present study are higher.

The results for product cost are shown in Figure 17 for the Hemlock Semiconductor process. A product cost for polysilicon of 15.60 \$/kg (1980 dollars) is presented in the Hemlock semiconductor report (25). The adjusted value is about 18.72 \$/kg (1985 dollars). The product cost of 19.48 \$/kg (1985 dollars) of the present study is slightly higher. Both results suggest product cost without profit is in the 20 \$/kg range.

Results for capital investment are shown in Figure 18 for Siemens, Union Carbide and Hemlock Semiconductor processes. The cursory comparison suggests that capital investment of \$41.4 million for the Hemlock Semiconductor process is lower than the \$69 million and \$74.75 million for Siemens and Union Carbide processes.

The results for product cost are given in Figure 19. The product cost per kg of polysilicon of \$29.49 for Siemens process appears to be the highest. The \$24.65 for Union Carbide process appears to be intermediate. The \$19.49 for Hemlock Semiconductor process appears to be lowest.

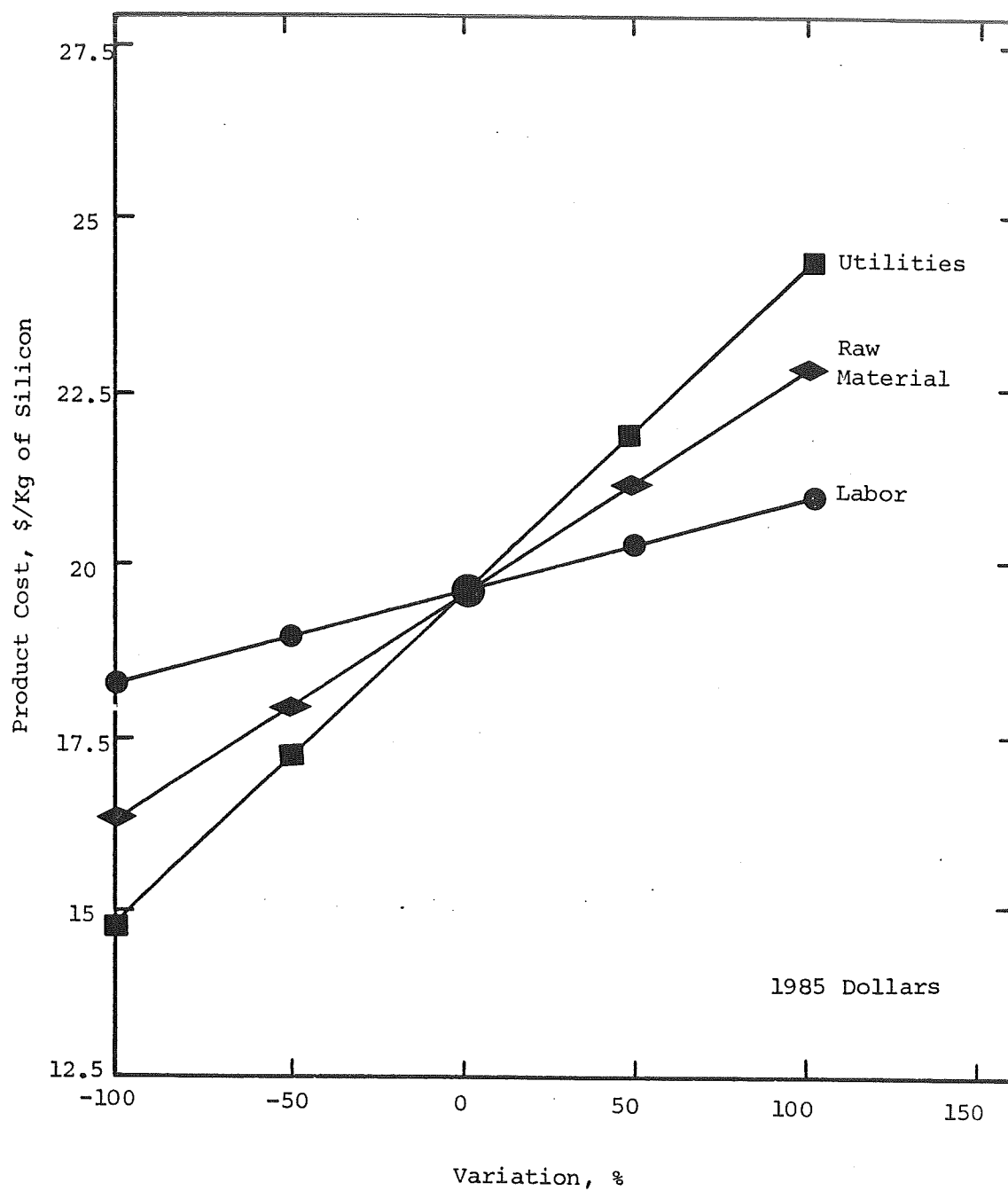


FIGURE 12. SENSITIVITY PLOT FOR HEMLOCK SEMICONDUCTOR PROCESS

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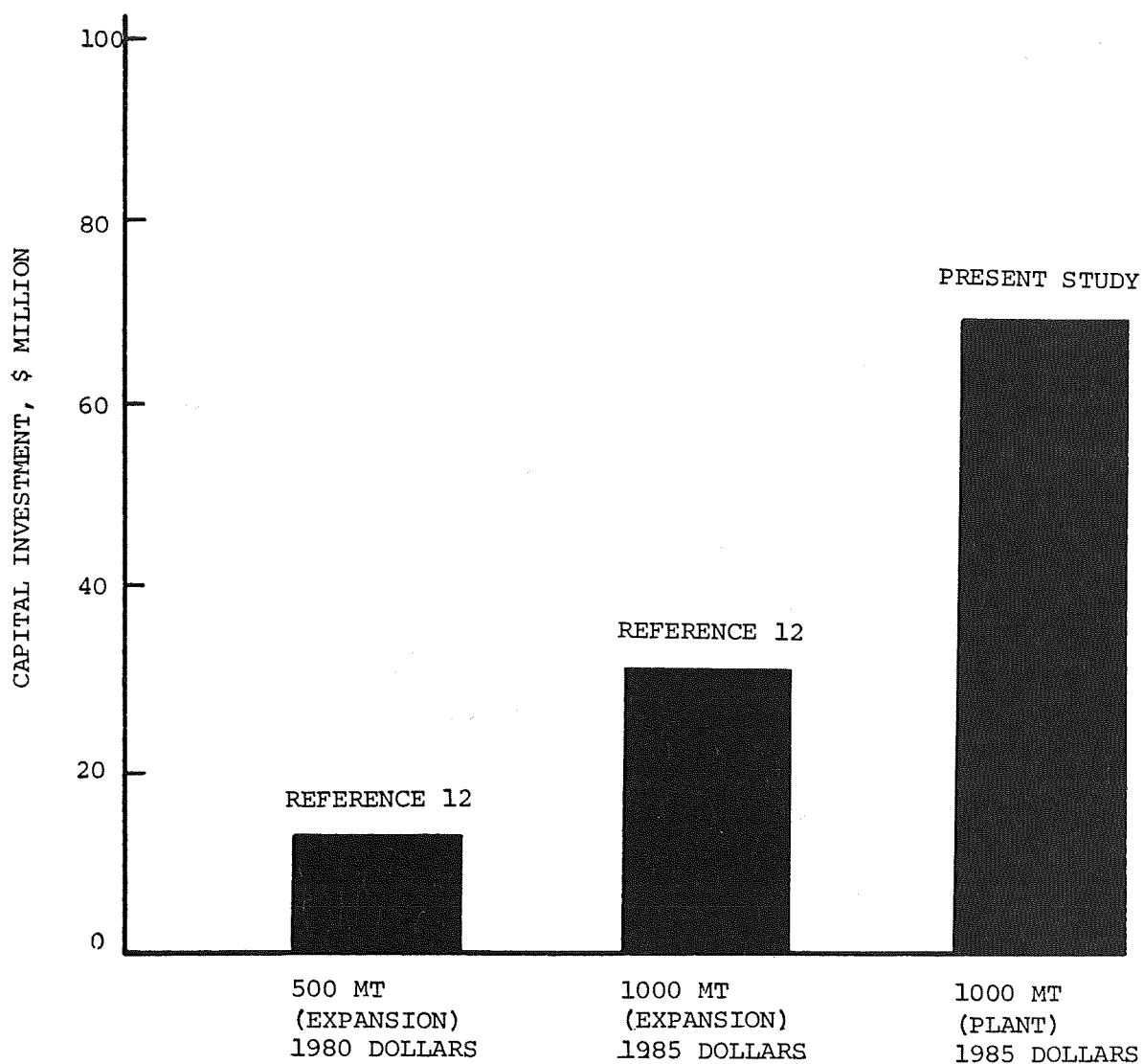


FIGURE 13 RESULTS FOR CAPITAL INVESTMENT: SIEMENS PROCESS

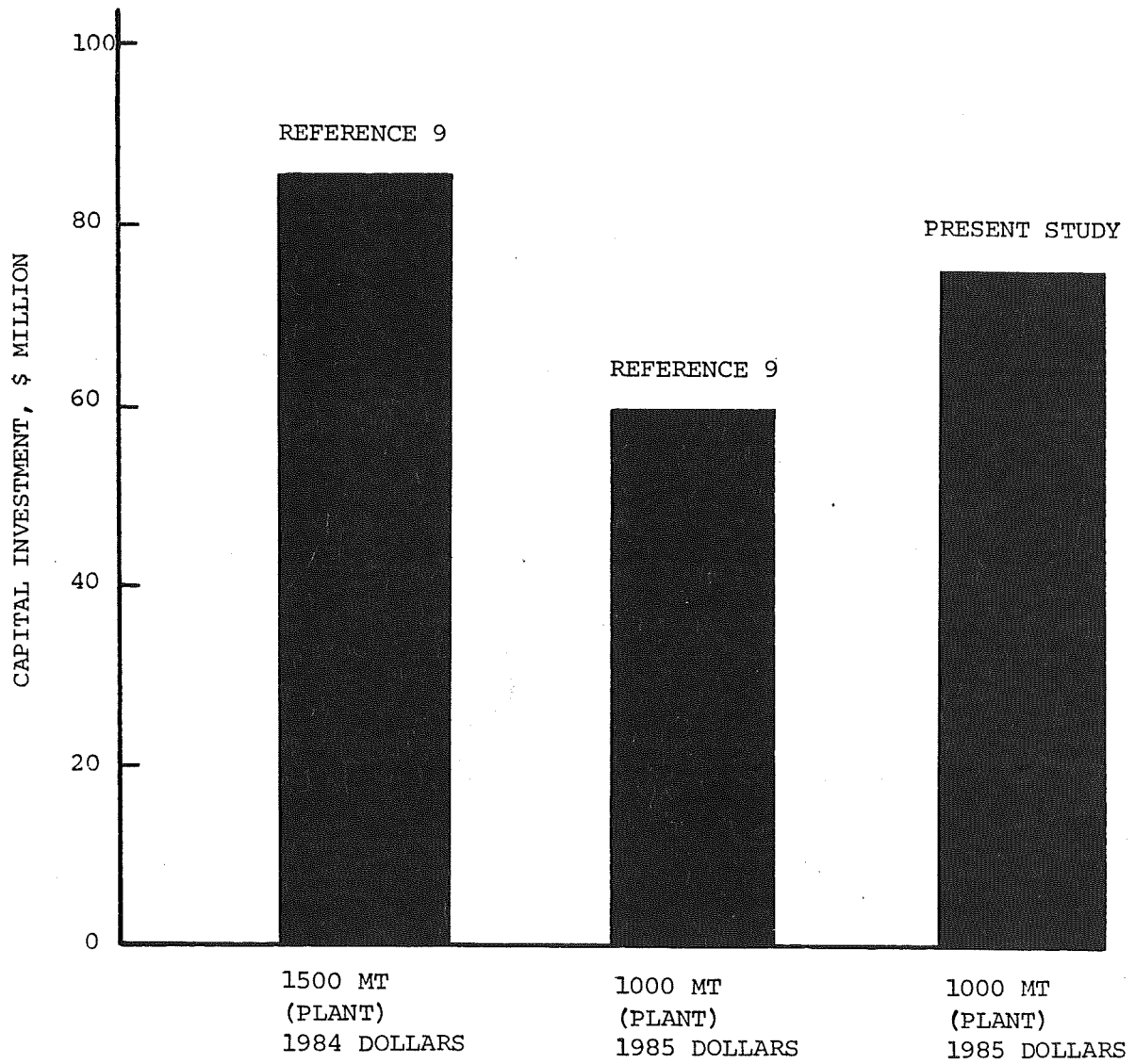


FIGURE 14 RESULTS FOR CAPITAL INVESTMENT: UNION CARBIDE PROCESS

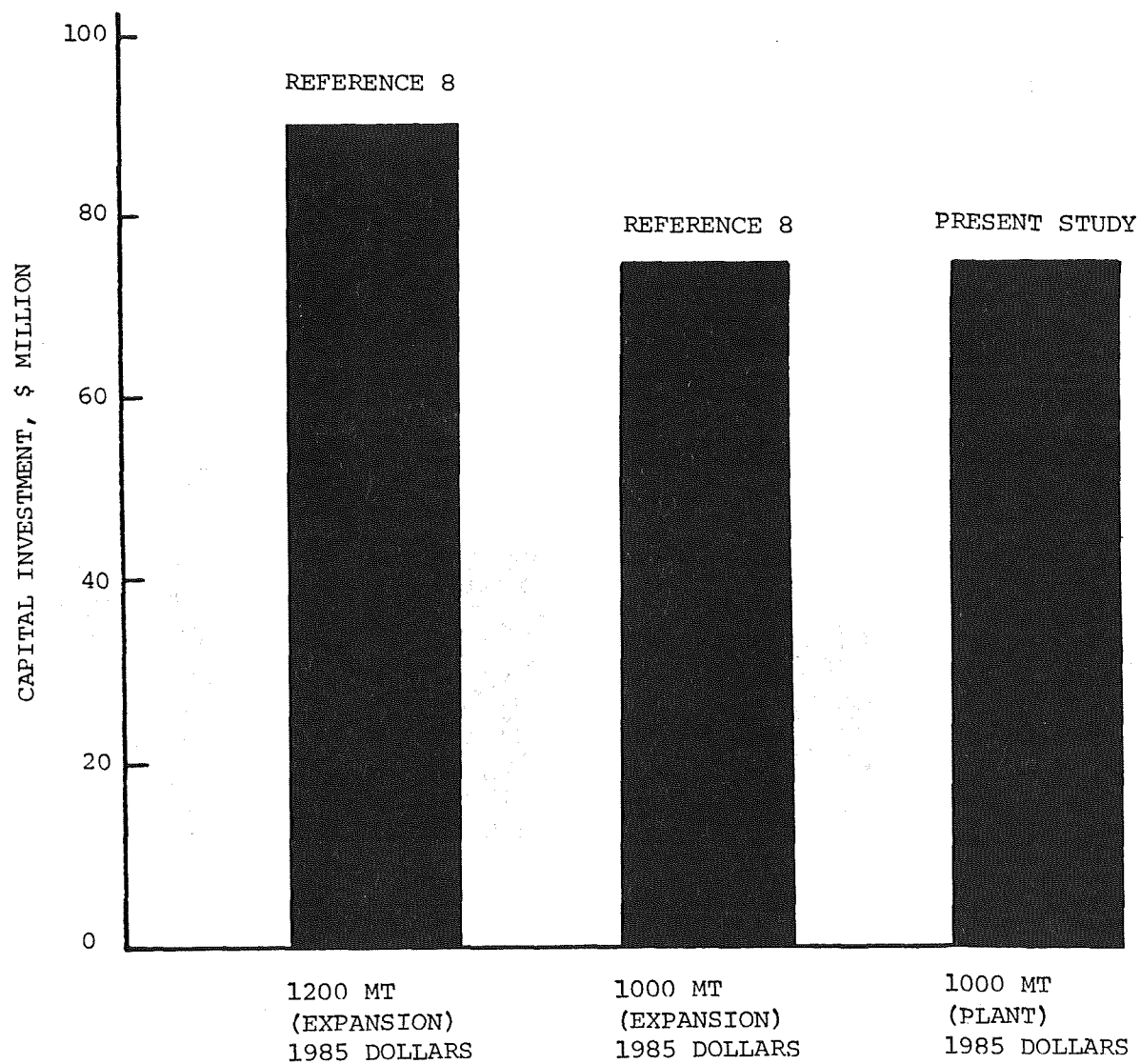


FIGURE 15 RESULTS FOR CAPITAL INVESTMENT: UNION CARBIDE PROCESS

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OF POOR QUALITY

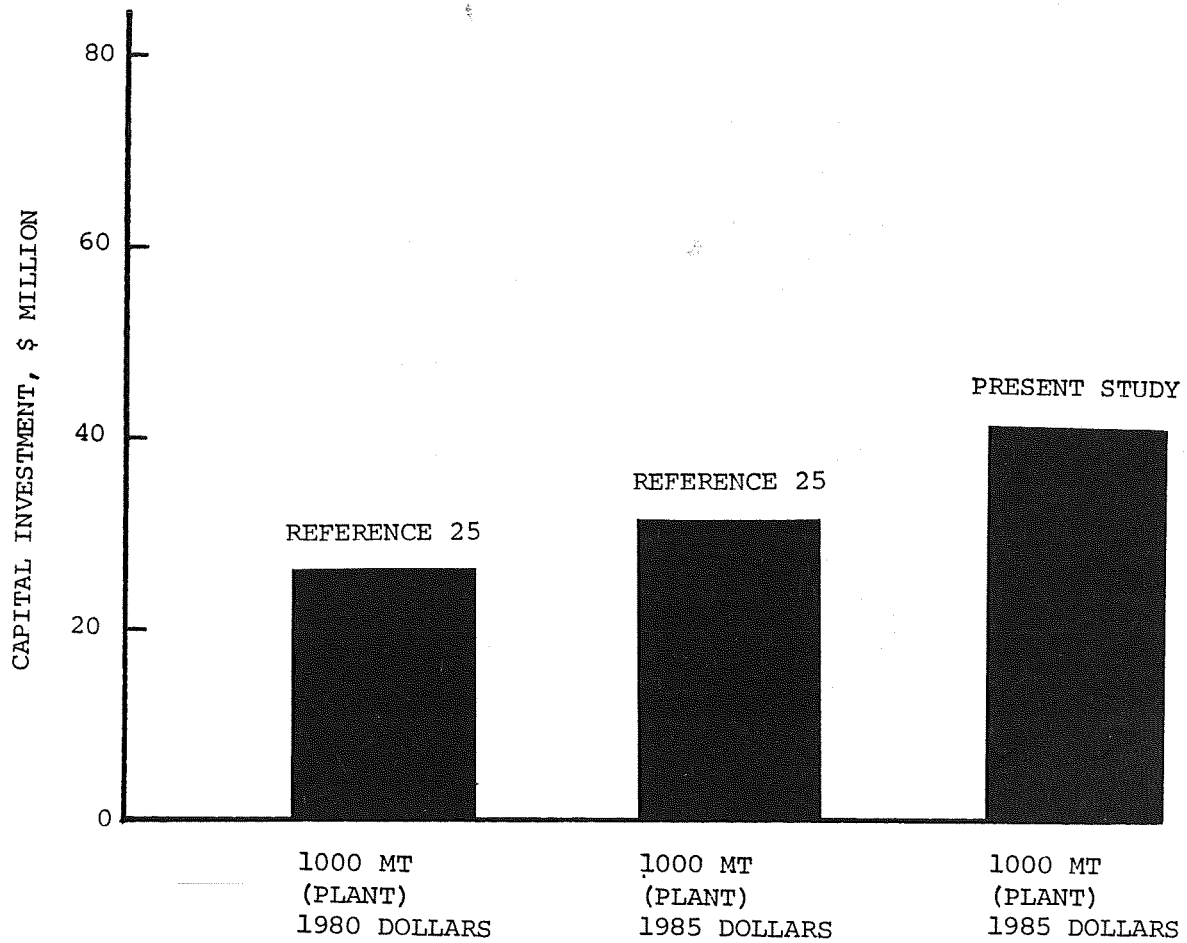


FIGURE 16 RESULTS FOR CAPITAL INVESTMENT: HEMLOCK SEMICONDUCTOR PROCESS



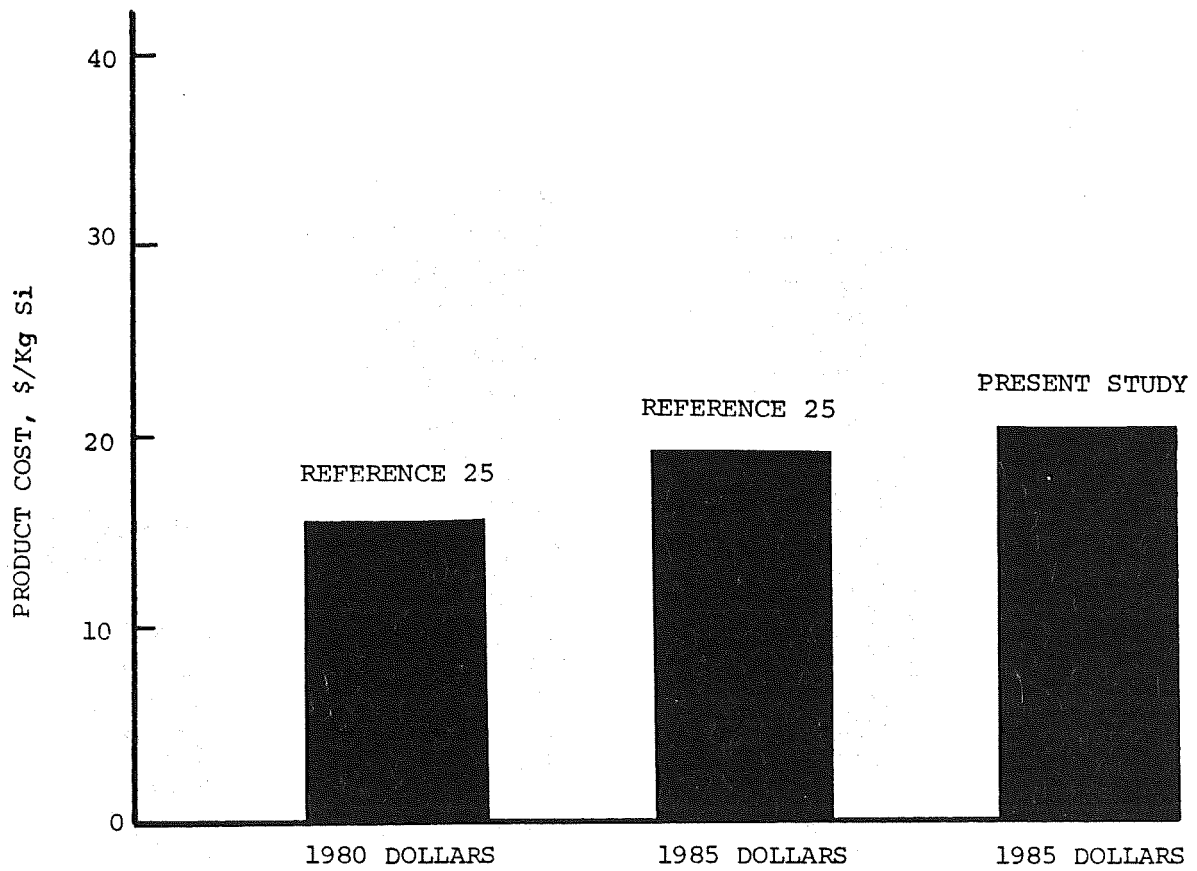


FIGURE 17 RESULTS FOR PRODUCT COST: HEMLOCK SEMICONDUCTOR PROCESS

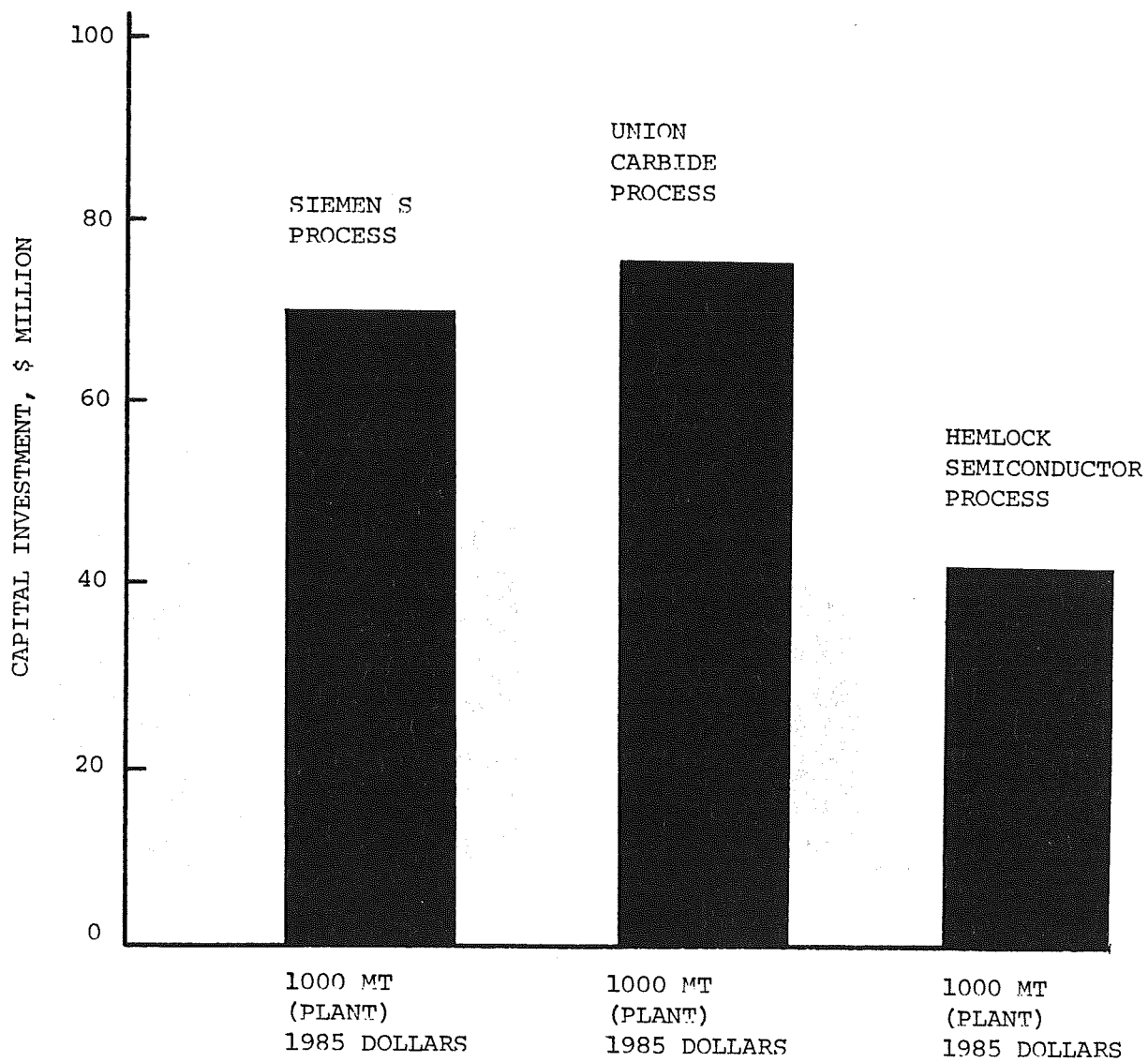


FIGURE 18 RESULTS FOR CAPITAL INVESTMENT

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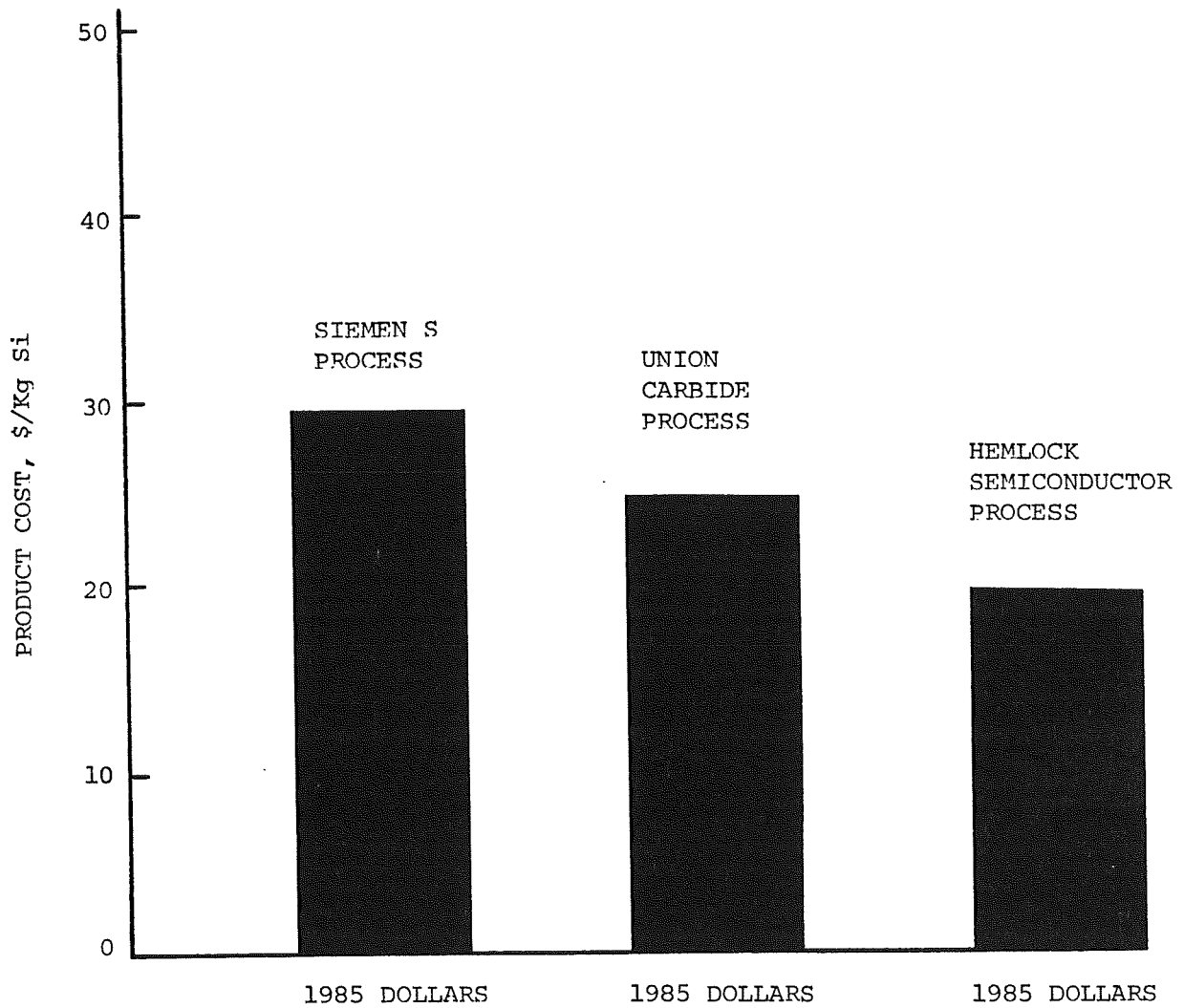


FIGURE 19 RESULTS FOR PRODUCT COST

## 6. OTHER PROCESSES

Other processes that depart from conventional technology are also under consideration for the production of polysilicon:

1. Zinc Reduction of Silicon Tetrachloride (Batelle Columbus Laboratories)
2. Bromosilane Process (J. C. Schumacher Co.)
3. Sodium Reduction of Silicon Tetrafluoride (SRI International, Inc.)
4. Sodium Reduction of Silicon Tetrachloride (AeroChem Research Laboratories, Inc., and Universal Silicon, Inc.).
5. Direct-Arc Furnace Process (Dow Corning Corporation)
6. Silicon Difluoride Transport Process (Motorola Inc.)
7. Carbothermic Reduction of Silicon Dioxide (Texas Instruments, Inc.)
8. Rotary Chamber Reactor for Use in a Closed-Cycle Process (Texas Instruments, Inc.)
9. High-Capacity Arc Heater Process (Westinghouse Electric Corporation)
10. Gaseous Melt Replenishment System (Energy Materials Corporation)
11. FBR Process (Osaka Titanium Co.)
12. Refining of Metallurgical-Grade Silicon (Heliotronic, GmbH)
13. Solar Cell Grade Silicon Prepared By Carbothermic Reduction of Silica (Siemens Research Laboratories)
14. A Metallurgical Route to Solar Grade Silicon (Elkem)
15. Solar Silicon From Directional Solidification of MG Silicon Produced Via The Silicon Carbide Route (Enichemico)
16. Silane Based Polysilicon Process (Eagle-Picher Industries, Inc.)
17. Silicon Purification Using A Cu-Si Alloy Source (Solar Energy Research Institute)

Initial economics of process 1 are given by Yaws (38, 39). Several recent news releases (16-18) relate to process 2. A good summary of processes 1-9 is provided by Lutwack (24). Process 10 is reported by Jewett, Bates and Hill (47). The remaining processes 11-17 are discussed in the proceedings of this meeting.

## 7. SUMMARY AND CONCLUSIONS

The following summary and conclusions are made as a result of the present study:

1. The economics of producing polysilicon in a 1,000 MT/yr plant are presented for the Siemens process (hydrogen reduction of trichlorosilane), Union Carbide process (silane decomposition) and Hemlock Semiconductor process (hydrogen reduction of dichlorosilane). The economics include estimates of capital investment and product cost to produce the polysilicon.
2. For the Siemens process using trichlorosilane, the product cost without profit is estimated to be 29.49 \$/kg Si (1985 dollars). This product cost includes provisions for hydrogen recycle, hydrogen chloride recycle and chlorosilane recycle. Without such raw material recycle, the product cost will be higher. The product cost also includes low electrical usage (120 kw-hr/kg Si). If higher

electrical usage is required, the product cost will be higher.

3. For the Union Carbide process using silane in a hot rod reactor (Komatsu), the product cost without profit is estimated to be 24.65 \$/kg Si (1985 dollars). This product cost assumes reasonable resolution of homogeneous gas phase decomposition reaction. Electrical usage is estimated at 62.4 kw-hr/kg Si.

4. For the Hemlock Semiconductor process using dichlorosilane, the product cost without profit is estimated at 19.48 \$/kg Si. This product cost assumes reasonable resolution of wall deposits and homogeneous reaction. An overall electrical power consumption of 82 kw-hr/kg Si is used.

5. A cursory cost comparison is made for capital investment cost. For the Siemens process, the comparison suggests that the capital investment of \$69 million of the present study may be high. For the Union Carbide process, the comparison indicates that the capital investment of the present study may be slightly high or equivalent. For the Hemlock semiconductor process, the comparison suggests that the capital investment of the present study may be slightly high.

6. The polysilicon economics (capital investment, product cost) presented in the present study are intended for use in initial project studies.

#### Acknowledgement

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## REFERENCES

1. Bawa, M. S., R. C. Goodman and J. K. Truitt, "Kinetics and Mechanism of Deposition of Silicon by Reduction of Chlorosilanes with Hydrogen", Chem. Vap. Dep. 4th Int. Conf., Electro. Chem. Soc., Princeton, N. J., pgs. 63-73, (1973).
2. Ciszek, T. F., "Silicon for Solar Cells", Government Report, SERI/TR-212-2084, DE 84004503, Solar Energy Research Institute (April, 1984).
3. Costogue, E. N., R. R. Ferber and R. Lutwack, "Polycrystalline Silicon Study", Government Report, DOE/ET/20356-17, DE 85000327, Jet Propulsion Laboratory (May, 1984).
4. Costogue, E. N. and R. Pellin, "Polycrystalline Silicon Material Availability and Market Pricing Outlook Study for 1980 to 88", Government Report, DOE/JPL-1012-79A, DE 84001226, Jet Propulsion Laboratory (February, 1983).
5. Dudukovic, M. P., P. A. Ramachandran and S. Lai, "Modeling of Fluidized-Bed Reactors for Manufacture of Silicon from Silane", Government Report, DOE/JPL-956737-8517, Distribution Category UC-63, Washington University (September, 1985).
6. Ecco High Frequency Corp., Personal Communication, North Bergen, N.J. (1977)
7. Electronic News, "Carbide Gets Silane License (Komatsu)", 58 (May 11, 1981).
8. Electronic News, "UC to Double Moses Lake Capacity", \$90 million for double capacity to 2,400 tons annually, 71 (January 28, 1985)
9. Electronic News, "Carbide to Start Silane Production", \$85 million for 1,500 tons per year, 15 (December 24, 1984)
10. Electronic News, "Mitsubishi Buys 12.25% Hemlock Stake", \$47 million for plant expansion of 1,000-1,200 tons annually, 15 (December 24, 1984).
11. Electronic News, "Say Poly Shortage Possible by 1987", \$47 million to increase production by 1,000 tons annually, 47 (January 7, 1985).
12. Electronic News, "Wacker in \$13M Poly Plant Expansion", \$13 million for increase of capacity from 1,200-1,300 to 1,800 metric tons per year, 69 (October 20, 1980).
13. Electronic News, "Siltronic (Wacker) to Build \$80 M Polysilicon Plant", \$80 million for polysilicon plant to be capable of manufacturing 5,000 metric tons per year, 70 (October 6, 1984).

14. Electronic News, "Toxic Substances Delay Building of Wacker Poly Plant", 63 (June 24, 1985).
15. Electronic News, "GE to Sell Poly Plant to NKK, Exit Market", \$16 million for poly plant with reported 200 metric tons capacity, 62 (October 7, 1985).
16. Electronic News, "Schumacher Names Materials Mgnt.", process of choosing sites for 1,000 tons/year polysilicon facility, 61 (January 21, 1985).
17. Electronic News, "Schumacher Breaks Out Chemical Div.", Semiconductor Materials Division for capital intensive polysilicon plants, 41 (April 15, 1985).
18. Electronic News, "Schumacher Plans Wafer Mfg. Facilities", company expects to start up pilot poly plant (50-75 metric tons), 54 (September 30, 1985).
19. Electronic News, "Site Selection", 3 (September 23, 1985).
20. Iya, S. K., R. N. Flagella and F. S. DiPaolo, J. Electrochem. Soc., 129 (7), 1531 (July, 1982).
21. Komatsu (Assignee, Japan), Yatsurgi, Y., A. Yusa and N. Takahashi (Inventors), U.S. Patent 4, 147, 814 (April 3, 1979).
22. Komatsu (Assignee, Japan), Yatsurgi Y., A. Yusa and N. Takahashi (Inventors), U.S. Patent 4, 150, 168 (April 17, 1979).
23. Levenspiel, O., et. al., "Preliminary Study of a Radiantly Heated Fluidized Bed for the Production of High Purity Silicon", Government Report, DOE/JPL-956133-83-1, DE 83016569, Oregon State University (August, 1983).
24. Lutwack, R., "A Review of the Silicon Material Task", Government Report, DOE/JPL-1012-96, DE 84014599, Jet Propulsion Laboratory (February, 1984).
25. McCormick, J. R., et. al., "Development of a Polysilicon Process Based on Chemical Vapor Deposition (Phase 1 and Phase 2)", Government Report, DOE/JPL-955533-83/7, DE 84004975, Hemlock Semiconductor Corporation (August, 1982).
26. McCormick, J. R., et. al., "Development of a Polysilicon Process Based on Chemical Vapor Deposition of Dichlorosilane in an Advanced Siemens Reactor", Government Report, DOE/JPL-955533-83, DE 84017155, Hemlock Semiconductor Corporation (July, 1983).
27. Mui, J.Y.P., "Investigation of the Hydrochlorination of  $\text{SiCl}_4$ ", Government

- Report, DOE/JPL-956061-7, DE 83015173, Solarelectronics (April, 1973).
28. Murthy, T.U.M.S., N. Miyamoto, M. Shimbo and J. Nishiyawa, J. of Crystal Growth, 33, 1 (1976).
  29. Reiter, L. J., "A Probabilistic Analysis of Silicon Cost", Government Report, DOE/JPL/1012-93, DE 84005741, Jet Propulsion Laboratory (November, 1983).
  30. Rogers, Leo, "Polycrystalline Silicon Plants", News Release, Polycrystalline Silicon Technology Corporation (P.S.T.), Mesa, Arizona (June 15, 1985).
  31. Smiel, "Silicon for the Semiconductor Industry", Company Brochure, MONTEDISON Group (May, 1979).
  32. TREI, "Silicon Production Process Evaluations", Government Report, DOE/JPL-956045-82/5, Distribution Category UC-63 (July, 1982).
  33. Wacker (Assignee, Germany), Koppl, F. and H. Hamster (inventors), U.S. Patent 4,173,944 (Nov., 1979).
  34. Yaws, C.L., T. G. Digges, Jr., M. A. Drews and B. J. Boggs, "High Purity Silicon Manufacturing Facility", Government Report AFRL-TR-71-130, Texas Instruments (July 1971).
  35. Yaws, C. L., J. W. Miller, Jr., R. Lutwack and G. Hsu, "Electricity from Sunlight: Low Cost Silicon for Solar Cells", Energy and the Environment-Proceedings of the Fifth National Conference, A.I.Ch.E.-A.P.C.A., p. 329, Cincinnati, Ohio (Oct. 31-Nov. 3, 1977).
  36. Yaws, C. L., J. W. Miller, Jr., R. Lutwack, and G. Hsu, Solid State Technology, 63 (January, 1979).
  37. Yaws, C. L., F. C. Jelen, K. Y. Li, P. M. Patel and C. S. Fang, Solar Energy, 22 (No. 6), 547 (1979).
  38. Yaws, C. L., K. Y. Li, C. S. Fang, R. Lutwack, S. Hsu and H. Leven, Solar Energy, 24 (No. 6), 359 (1980).
  39. Yaws, C. L., et. al., "Process Feasibility Study in Support of Silicon Material Task I", Government Report, Jet Propulsion Laboratory, Contract No. 954343, DOE/JPL-954343-81, Distribution Category UC-63, Lamar University (February, 1981).
  40. Yaws, C. L., K. Y. Li, T. C. T. Chu, C. S. Fang, R. Lutwack and A. Briglio, Solar Energy, 27 (No.6), 539 (1981).
  41. Chen, C.C., M. M. Chang, F. R. Shu and C. S. Liu, J. Ch. I. Ch. E., 5, 93 (1974).



42. Chen, C.C., M. M. Chang, C. S. Liu, C. T. Chang and F. R. Shu, J. Ch. I. Ch. E., 5, 99 (1974).
43. Chen, C.C., M. M. Chang, F. R. Shu and C. S. Liu, J. Ch. I. Ch. E., 5, 107 (1974).
44. Peters, M. S., and K. D. Timmerhaus, "Plant Design and Economics for Chemical Engineers," 3rd edition, McGraw-Hill Book Co., N.Y. (1980).
45. Ingle, W. M. and R. D. Darnell, J. Electrochem. Soc., 1236 (May, 1985)
46. Ingle, W. M. and R. D. Darnell, J. Electrochem. Soc., 1240 (May, 1985).
47. Jewett, D. N., H. E. Bates and D. M. Hill, "Gaseous Melt Replenishment System," Government Report, DOE/JPL-955269-80/1, Distribution Category VC-63, Energy Materials Corporation (April, 1980).

## APPENDIX A

### COSTS USED IN ECONOMIC ANALYSIS

#### 1. Raw Material Costs

<u>Raw Material</u>	<u>Raw Material Cost</u>
1. M. G. Silicon	.763 \$/lb
2. Silicon Tetrachloride	.227 \$/lb
3. Liquid Hydrogen	9 \$/1000 ft <sup>3</sup>
4. Copper Catalyst	1.548 \$/lb
5. Hydrate Lime	.0252 \$/lb
6. Hydrogen Chloride	.168 \$/lb
7. Nitrogen	4.08 \$/1000 ft <sup>3</sup>

Source: Hemlock Semiconductor (ref. 25), page 129, adjusted to 1985 dollars

#### 2. Utility Costs

<u>Utility</u>	<u>Cost of Utility</u>
1. Electricity	5 /kw-hr
2. Steam	1.89 \$/mmBTU
3. Hot Oil	1.89 \$/mmBTU
4. Cooling Water	.144 \$/mgal
5. Process Water	.680 \$/mgal
6. Refrigerant (-40°F)	18.68 \$/mmBTU
7. Refrigerant (34°F)	3.75 \$/mmBTU
m = 1,000	
mm = 1,000,000	

Source: Hemlock Semiconductor (ref. 25), page 128, and Peters and Timmerhaus (ref 43), page 881, adjusted to 1985 dollars

#### 3. Labor Cost 12.00 \$/hr

Source: Electronic News (ref. 19)

# APPENDIX B

## CHECKLIST FOR CHEMICAL ENGINEERING ANALYSIS

<u>Prel. Process Design Activity</u>	<u>Status</u>	<u>Prel. Process Design Activity</u>	<u>Status</u>
1. Specify Base Case Conditions	●	7. Equipment Design Calculations	●
1. Plant Size	●	1. Storage Vessels	●
2. Product Specifics	●	2. Unit Operations Equipment	●
3. Additional Conditions	●	3. Process Data (P, T, rate, etc.)	●
		4. Additional	●
2. Define Reaction Chemistry	●		
1. Reactants, Products	●	8. List of Major Process Equipment	●
2. Equilibrium	●	1. Size	●
		2. Type	●
3. Process Flow Diagram	●	3. Materials of Construction	●
1. Flow Sequence, Unit Operations	●		
2. Process Conditions (T, P, etc.)	●	8a. Major Technical Factors	●
3. Environmental	●	(Potential Problem Areas)	●
4. Company Interaction	●	1. Materials Compatibility	●
(Technology Exchange)		2. Process Condition Limitations	●
		3. Additional	●
4. Material Balance Calculations	●		
1. Raw Materials	●	9. Production Labor Requirements	●
2. Products	●	1. Process Technology	●
3. By-Products	●	2. Production Volume	●
5. Energy Balance Calculations	●	10. Forward for Economic Analysis	●
1. Heating	●		
2. Cooling	●		
3. Additional	●		
6. Property Data	●	0 Plan	
1. Physical	●	● In Progress	
2. Thermodynamic	●	● Complete	
3. Additional	●		

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## DISCUSSION

SHIMIZU: This morning, I presented a paper about the same subject and I obtained quite different conclusions. Would you comment on my paper?

YAWS: Although I didn't write down complete notes on your paper, the order in which the processes was placed is about the same as in my analysis. I think you had the Hemlock Semiconductor process lower than the Union Carbide process with the Komatsu reactor, and you had the conventional Siemens process at a higher cost than Union Carbide.

SHIMIZU: Also, this morning, I compared those processes and others. In the case of silane, I compared two processes. One was the fluidized-bed reactor process, and the other was the Komatsu reactor.

YAWS: The comparison showed that the Union Carbide process with the Komatsu reactor had a lower production cost than the Siemens process.

SHIMIZU: In my estimate, the cost is about 80% that of the conventional Siemens process.

YAWS: Your table showed that the Siemens process is the most expensive. Union Carbide, using the Komatsu reactor, is second. The third is Hemlock using dichlorosilane. Those are the same results that I got. Now the numbers may be a little different, but I think our ranking orders are the same. I didn't look at as many processes as you did.

WRIGHT: You used a scaling factor for production levels from about 1500 MT/year down to 1000 MT/year. Was that a linear scaling factor? What was your justification for using that particular scaling factor?

YAWS: For normal chemical industry equipment, the factor is about 0.6, or a scaling factor which has been determined to be best for a specific type of equipment can be used. I assumed that the major cost was in reactors so that a linear factor was used for additional reactors.

PRINCE: Did you make the calculations for the Union Carbide process with the fluidized-bed reactor?

YAWS: No. That was not part of this study. The calculations were for the Union Carbide process operating with Komatsu deposition reactors.

AULICH: Your calculations were for 1000 MT/year. Since 1000 MT/year capacity plants are not needed now for the photovoltaic industry, what is the analysis of the cost of these processes if smaller units of about 200 MT/year are used?

YAWS: Smaller plants would result in higher costs. Someone would have to do a study to get the answers. I can't respond to your question now, because I haven't done the analysis, but it can be done.