# Towards a Profitable Means of Municipal Refuse Disposal

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#### ABSTRACT

Refuse disposal processes which can generate revenue from the sale of by-products are studied. Economic evaluation of several alternatives reveals ethanol production from the wastepaper content of the refuse to have strong profit potential. This is studied in depth and a design proposed with sufficient flexibility to enable the process to function profitably despite chemical kinetic uncertainties. Further work is recommended to take ethanol production from refuse to the pilot-plant stage with the ultimate objective of full-scale municipal installation for refuse processing.

#### INTRODUCTION

The magnitude of the urban refuse collection and disposal problem may be grasped when one considers the major urban areas of the United States. Roughly one quarter of the population lives in places with 120,000 or more inhabitants. It has been established [1] that a median value for urban refuse production is of the order 4.1 lb of ordinary refuse per capita per calendar day (a dry-weight basis is used in this paper) with 25 percent greater production in the maximum week. A "typical" 120,000 strong community generates approximately 1700 tons of refuse per week.

Most near-at-hand sanitary landfill sites have been exhausted. Also, land and labor costs are rising.

Recently quoted disposal costs are [2] Elmira, N.Y., \$5.85 per ton, [3] Cambridge, Mass., \$12.10 per ton. Incineration costs are falling, and recently announced "Melt-Zit Destructor" [3] has a projected disposal cost of \$3 per ton. Composting has received some attention, and a private corporation [4] in St. Petersburg, Florida, is currently composting that city's refuse at a cost of \$3.24 per ton to the city.

So far, no process has genuinely made a profit on refuse disposal. With this as an objective, several alternatives are analyzed, the state of the art reviewed, and the most promising method selected and analyzed in depth.

# DESIRABLE FEATURES FOR REFUSE DISPOSAL PROCESSES

1) Grit, odor, fume, and bacteria free with no risk of rodents and flies.

2) The revenue from the by-product sale should realize a net profit on the plant operation, or at least ensure that the overall disposal costs are less than the alternative method available to the community.

 Capable of central location at, for example, a light industrial site in order to minimize transportation costs.

4) The maximum capital investment per ton of disposal capacity to be of the same order as that for incineration, as it is felt the onus of municipal

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fund raising would deter the investment in a costlier project.

5) The process to be as omnivorous as possible.

6) Plant capacity to be adequate for a twentyyear life.

7) By-product quality either easily maintained or unimportant to avoid high labor costs.

# CHARACTERISTICS AND BY-PRODUCT POTENTIAL OF MUNICIPAL REFUSE

The character of municipal refuse is changing, the trend being to more paper and less garbage, ashes, and glass. A typical present-day municipal refuse [5] will be considered as:

60 percent paper

- 8 percent garbage
- 10 percent metal
- 12 percent ashes and glass
- 10 percent miscellaneous, by weight

Considering the minor groups first, there is virtually no market for glass cullets now. Furthermore, the laborious handpicking required would mitigate against salvage of this material. Garbage could be collected separately and used for hog-feeding; however, this feudal practice is soon to be abolished by the Public Health Service. The ferrous scrap content can be readily extracted magnetically; there is a market for scrap steel which is *hot* tinned or lead jointed. This will exclude the greater portion of the ferrous scrap collected which is in the form of tin cans. The remainder of the steel scrap could be baled and sold, and the operation of a scrap baler, in conjunction with a process which requires screening of the materials, may be marginally profitable.

The paper content is very interesting, not from the salvage point of view, but as a source of raw material, viz. cellulose which is also present in garbage. Most paper is virtually cellulose plus whatever filler is used to give it the desired characteristics. Any refuse destruction or reforming process could have cellulose as its main ingredient, so a closer look at its properties is indicated.

Cellulose [6, 7] is a carbohydrate polymer composed of anhydroglucose units and has the empirical formula  $C_6 H_{10}O_5$ . It is the chief structural element and major constituent of the cell walls of trees and other higher plants. Many important derivatives stem from it, such as cellulose nitrate, cellulose acetate, and so forth. For the purposes of this study, the most important processes cellulose can undergo are destructive distillation, bacterial and animal digestive decomposition, and hydrolysis.

Destructive distillation results in the following products and yields [6]:

Product	Percent by Weight
Charcoal	20-42
Water of pyrolysis	34
Acetone	0.2
Acetic acid	3.1
Tar and Oils	10-15
Combustible gases	6-8
CO <sub>2</sub>	10-12

Tars begin to come off at 200 C and a slightly exothermic reaction takes place about 270 C due to the occurrence of secondary and tertiary reactions. Further elevation of the temperature results in a slow formation of products, and no further volatilization occurs at 470 C.

Bacteriological decomposition can be carried out by mesophilic or thermophilic bacteria. The former take literally aeons of time; the latter will decompose the bulk of the cellulose in under two weeks given the proper conditions. From the foregoing, it is inferred that relatively rapid composting is possible. Cellulose is also capable of degestion by cattle and other ruminants if treated with a mild alkali and then washed. This method was used in the Scandinavian countries during the first world war when wood and straw pulp was fed to sheep succesfully. The potential of animal feeds from refuse is noted, but due to lack of nutritional data and the difficulty of separating non-digestibles such as glass shards from the feed, this alternative is not considered further.

Hydrolysis may be defined as a process in which a double decomposition reaction is carried out with water as one of the reactants. The hydrolysis of starch is the basis of the corn syrup industry. The hydrolysis of cellulose is accomplished with the aid of an acid as the hydrolysis reagent, e.g.

$$C_{6}H_{10}O_{5} + H_{2}O \xrightarrow{\text{HC1}} C_{6}H_{12}O_{6}$$
  
Cellulose Sugars

The acid acts as a catalyst and for fast industrial reactions and high yields, the process is carried out at high temperature. Having obtained the sugars, a variety of operations can be carried out, the principal one being fermentation to yield an aqueous solution of alcohol which is subsequently rectified, the reaction being:

yeast  $C_6 H_{12}O_6 \longrightarrow 2C_2 H_5 OH + 2 CO_2$ Sugars Ethanol

The kinetics of cellulose hydrolysis are studied in the Appendix.

# ALTERNATIVE PROCESSES FOR REFUSE DISPOSAL

Four processes are analyzed at various levels. The basis for operating cost estimates will be a present-day population of 120,000. The operating load based on a population of 120,000 is 249 tons of refuse per day. Plant life is taken as 20 years with bond interest at 4 percent, resulting in fixed charges of 7.35 percent on the total capital cost of the installation. For subsequent disposal of refuse, the plant will be charged \$4.50 per ton.

The four processes, their products and services are:

Process	Product or Service
Incineration	Refuse disposal
Destructive	Refuse disposal, acetic
distillation	acid acetone
Composting	Refuse disposal, compost
Hydrolysis	Refuse disposal, ethanol

#### INCINERATION

Incineration is a method which has been used for many years; it is now grit and odor free and a very competitive alternative to sanitary landfill or for reducing the volume required prior to landfilling. A flow diagram for a combined incinerator steamraising plant is shown in Fig. 1. A scheme utilizing incineration with waste heat recovery, power generation, and desalting has been built and commissioned satisfactorily for the township of Hempstead, Long Island in late 1965 [8-10]. This plant can be said to represent the latest incineration practice for which published costs are available. It is of considerable interest to analyze it in order to have a performance or cost index which any new process must better.

The Hempstead plant disposes of 750 tons of refuse per day and operates 7 days per week. Two 300-ton rocking grate continuous feed refuse furnaces



#### FIG. 1 FLOW DIAGRAM FOR AN INCINERATOR PLANT, WITH POWER GENERATION AND DESALTING

plus a 150-ton large trash furnace are used. The hot gases from the refuse furnaces are discharged through waste heat boilers which generate up to 160,000 lb of steam per hour at 460 psig, which is sent to one of two 1250-kw turbo-generator sets. The excess steam is reduced in pressure, and this plus the turbine exhaust is used to evaporate of the order of 400,000 gpd of fresh water. Both the power generated and the water obtained are consumed internally for auxiliaries and boiler feed. The plant is self sufficient, but sells no power, and the surplus water is not pumped to the town supply. The plant cost is \$6,000,000 to build [9], of which \$3,000,000 was for mechanical equipment [10].

An order of magnitude economic analysis is shown in Table 1.

TABLE 1				
Total Capital Cost	\$6,000,000			
Fixed charges at .0735 Maintenance costs at 2% of	440,000			
mechanical equipment costs	60,000			
Residue Disposal	123,000 123,000			
Total Cost/Year Disposal cost per ton \$3.82.	\$1,043,000			



FIG. 2 FLOW DIAGRAM, ACETIC ACID FROM REFUSE

It is concluded that incineration is a viable alternative to sanitary landfilling, but since profitability is the criterion here, it will be retained as a performance norm for capital and disposal costs. These costs will be taken respectively as \$8000 per ton of installed daily capacity and \$3.82 per ton.

### DESTRUCTIVE DISTILLATION

As far as we know, this process has not been applied to the disposal of refuse. It was used in the early 1900's for the manufacture of acetic acid and methanol from hardwoods. References [11-12] give details of the processes used, and the (then) state of the art.

An elementary flow diagram for the manufacture of acetic acid is given in Fig. 2.

On the recommendation of a leading manufacturer in the field, the process is designed around vacuum retorts 3 ft 6-in. dia and 30 ft long. The refuse is baled into 3 ft dia by 5-ft long slugs with a dry density of 30 lb/cu ft. The slugs are predried, inserted in the retorts, and the destructive distillation carried out to near completion with the evolution of noncondensable gases such as  $CH_4$  and  $CO_2$ , condensed liquors and char. The noncondensables are burned for process heating, the char is quenched, briquetted, and hopefully, marketed.

The condensate is allowed to settle crudely into heavy and light fractions. The heavy fraction is sent to a tar still to yield creosote and a phenolic agglomeration, or is burned for process heat. The lighter fraction is also an agglomeration which may be extremely complicated because of refuse heterogeneity. The acetic acid is stripped from the oils in the vapor phase using a suitable high-boiling solvent, and is then subsequently separated from the solvent by vacuum distillation to yield a commercial grade acid which is highly marketable.

Much of the plant cost is tied up in the vacuum predriers and retorts, and as the governing transfer mechanism is heat conduction into the billet of compressed refuse, an order of magnitude transient conduction analysis was performed which arrived at a retort residence time of 23 hr. Using a retort residence time of 23 hr and a capacity of roughly 3 tons of refuse per charge, the processing of a refuse load of 249 tons per day requires a total of 84 separate retorts. A suitable retort for duty at 500 C was estimated to cost \$65,000. Thus the cost for the retorts alone is of the order of  $5.5 \times 10^6$ . When the building, materials handling, and process equipment costs are added to this, the estimated plant cost becomes \$8,000,000. The revenue from the distillation products is based on 150 tons of cellulose content in the operating load of 249 tons per day. The ensuing decomposition of the paper results in roughly 60 tons of saleable products per day, 40 tons of moisture and combustible gases, and 50 tons of refuse which requires disposal along with the remaining 99 tons of miscellaneous refuse. A revenue estimate is given in Table 2. The disposal cost is calculated in Table 3. It is concluded that destructive distillation will not be an acceptable method due to capital cost, too heavy reliance on the products maintaining their present market price and high disposal costs.

#### COMPOSTING

This process has been used in Europe for at least 50 years. The Craigmillar plant in Edinburgh [13] rapid composts 140 tons of crude refuse per day into stable compost in five days using a Dano rotary digester. Compost in the United States has to compete with a massive inorganic fertilizer supply and formerly has been mainly used to fill holes in the ground [14]. Commercial interests are now trial marketing composts as a soil conditioner [4]; so a quick look at this alternative is justified.

TABLE 2. REVENUE ESTIMATE					
Item	% Yield		Tons/Day	Price	Revenue \$/Day
Acetone	.3		.45	6.5¢ /lb.	57
Acetic Acid	3.1		4.65	9¢/lb.	835
Tars	7		10.5		
				1¢/lb.	360
Oils	5		7.5		
Charcoal	25		37.5	\$7/ton	260
					\$1,512

TABLE 3

Plant Cost	\$8,000,000
Fixed charges at .0735	590,000
Labor, 12 men at \$7,000	84,000
4 men at \$9,000	36,000
Maintenance (2% retort investment)	110,000
Disposal 149T residual refuse/day at \$4.50/ton	245,000
Total Cost	1,065,000
Revenue at \$1,512/day	550,000
Net annual cost	515,000
Disposal cost/ton	\$5.66

Composting is a controlled process of decay which is affected by the microorganisms in the wastes themselves, and which takes place spontaneously when there is a humid environment within a mass of organic wastes such as may be found in domestic refuse. Thermophilic organisms decompose cellulose, and these organisms (actinomysetes and fungi) miltiply enormously in the temperature range 115-140 deg F. The maintenance of these temperatures is vital to rapid composting, and agitation is required to prevent temperatures in excess of 160 F. The Dano [13] and the Naturiser [4] are two processes which agitate the compost while optimum temperature and moisture levels are maintained by air and water injection, respectively. A flow diagram for a composting plant is given in Fig. 3.

The process is extremely simple but extensive materials-handling is called for, especially if the digester is dispensed with and a bull-dozer and concrete slab used as proposed by Stovroff [15].

The state of the art and an economic analysis can be dealt with by considering the St. Petersburg plant built by Westinghouse [4]. The plant handles roughly 140 tons of refuse per day per six-day week. Of this 140 tons per day input, 100 tons of saleable compost are produced, roughly 10 tons of salvageable materials are recovered (no revenue estimate was available for these materials), 10 tons of noncompostable trash are disposed of by dumping, and 20 tons are lost in the composting process by various forms of gaseous emission. The compost is sold at \$4 per ton f.o.b. plant for the agricultural grade (carbon: nitrogen ratio 35:1) and \$10 per ton f.o.b. plant for the horticultural grade (carbon: nitrogen ratio 25:1). The city currently pays \$3.24 per ton to the plant for disposal.

An order of magnitude cost analysis is given in Table 4.

On the basis of the estimate for the St. Petersburg plant, composting might be a competitive solution to



# (MANY OF THE ABOVE STEPS ARE OPTIONAL) FIG. 3 FLOW DIAGRAM, COMPOST FROM REFUSE

refuse disposal problems in extremely specialized circumstances. There exists in the St. Petersburg area a need for organic fertilizers for the citrus groves, and this need is filled admirably by compost. Composting has been tried repeatedly in the United States and has always failed for lack of a market; thus, while the St. Petersburg plant does make a profit, it is in an exceptional geographic locality where a ready market does exist. As existing methods of refuse disposal are also considered in this paper, composting has been included for completeness.

# HYDROLYSIS

So far as is known, this process has not been applied to refuse disposal, although the Chemical Abstracts list many applications of it to cellulosic materials such as cotton hulls, hardwoods, and so forth. The hydrolysis of wood was a war-time industry which had a swift demise in the post-war years in spite of the extensive efforts made to keep it alive [16]. A war-baby plant is recorded as surviving until 1947 which used the "Madison Process" [17], viz. 180 C temperature and 0.5 percent  $H_2SO_4$  to effect hydrolysis times of the order of  $3\frac{1}{2}$  hr, and yields of 50-55 gal per ton of wood.

The hydrolysis process is conceptually quite simple. Fig. 4 shows a rudimentary flow diagram which illustrates the principal steps in the hydrolysis of the cellulosic content of the refuse with subsequent fermentation followed by rectification to yield

TABLE 4		
Plant Cost (4)	ter e com	\$1,500,000
Fixed charges @ .0735		110,000
Labor 12 men @ \$6,000 p.a.		72,000
Maintenance at 1% of investment		15,000
Disposal of 10T/day residual refuse @ \$4.50/T		14,000
	TOTAL COSTS	211,000
Projected Revenue:		
43,680 T @ \$3.24/T		142,000
9,000 T @ \$10/T		90,000
21,000 T @ \$4/T		84,000
	TOTAL	316,000
Net Income		105,000
Profit Per Ton of Refuse (to private contractor)		\$2.4



FIG. 4 FLOW DIAGRAM, ETHANOL FROM REFUSE

ethanol. Briefly, the refuse is pulverized and discharged into a wash tank for flotation separation which allows segregation of the refuse into dense and light fractions. The lighter fraction, mainly cellulosic materials, floats out with the exiting water stream. This portion is shredded, fed into a reactor, and hydrolyzed at 230 C and 0.4 percent H<sub>2</sub>SO<sub>4</sub> with an optimum residence time of 1.2 min (as determined in the Appendix), to obtain maximum conversion. Flash cooling using the process feed water as coolant, neutralization, and filtering follow. Fermentation is then carried out for roughly 20 hr at 90 - 100 deg F, and the resulting ~ 1.6 percent aqueous ethyl-alcohol solution is rectified to yield 95 percent pure ethanol.

The ethanol yield is calculated for two values of paper content in the refuse, a median value of 60 percent by weight, and a minimum value of 40 percent by weight, respectively, are used to determine the effect of this variable. Of the 249 tons of refuse processed daily in the plant, there will be 150 tons of paper on the 60 percent paper-content basis. The paper cellulose content [18] will vary from roughly 60-65 percent in newsprint to over 95 percent in most other forms of paper manufactured. For estimation purposes, the cellulose content is taken as 75 percent. The ethanol yield calculations are detailed in the Appendix (Table 10) and arrive at a daily 95 percent ethanol production of 10,780 gal on the 60 percent basis, and 6850 gal on the 40 percent basis.

#### **REVENUE ASSESSMENT**

The current commercial grade ethanol price in tank car lots is  $52\phi$  per gal. For calculation purposes,  $40\phi$  per gal will be used to allow for large volume movement and conservative revenue estimates. The daily gross revenue from the plant is then of the order \$4300 and \$2740 on the 60 and 40 percent bases, respectively. These gross revenues are more than can be realized by any of the other alternatives and this, coupled with the simplicity of the plant equipment, prompted a more detailed cost analysis. This is summarized in the Appendix and arrives at a total cost of \$2,262,000 of which \$1,062,000 is for equipment.

In assessing the disposal costs, the 40 percent paper content costs for fixed charges, maintenance, labor and power are assumed set by the 60 percent paper content requirements and will thus be taken as equal. The material solid refuse disposal and biochemical oxygen demand (BOD) reduction costs are dependent on the paper content in the refuse (though not all quantities are directly proportional). The detailed material cost break-down is given in Table 11 in the Appendix. There is also solid refuse to be disposed from the process, namely 176 T/D (dry weight) and 201 T/D (dry weight) for the 60 and 40 percent bases, respectively. A detailed refuse mass balance is given in the Appendix in Table 12. This refuse will be wet to some extent and may require centrifuging prior to disposal by sanitary landfill. However, a major portion of it will be Ca SO4 and cellulose which may have a nominal value as a soil conditioner in certain areas. The refuse will be in very compact forms, as filter cake from the pressure filters and segregated dense refuse such as glass and ashes from the flotation tank. Disposal should, therefore, be more readily accomplished than that of crude refuse which requires compaction. A cost of \$4.50 per ton is used for disposal, based on the dry refuse weight, as the moisture content does not lend any bulk. The liquid wastes from the process will contain from 57 to 87 T/D of dissolved decomposed sugars, depending on the paper content at the time. This waste liquor will have a high BOD, and its treatment is also discussed in the Appendix where yearly BOD

Item	60% paper content yearly cost \$		40% paper content yearly cost \$
Plant Cost	\$2,262,000		\$2,262,000
Fixed charges at .0735	166,000		166,000
Maintenance at 2% of equipment cost Material, costs from Appendix	21,240		21,240
\$826/Day (includes power) Labor, for a total work force of 26 men	305,000	\$618/day	225,000
at \$6,000 Disposal of 176 tons/day residual refuse	156,000	201 T/D	156,000
at \$4.50/ton B.O.D. Reduction 3¢ /lb B.O.D.	280,000	at \$4.50/T	330,000
23,500 Ib/day	258,000	15,700 lb/day	172,000
Total Cost/Year	1,186,240		1,070,240
Yearly Revenue: $3.93 \times 10^6$ gallons at \$.40/gal.	1,570,000	$2.5 \times 10^6$ galls. at \$.4/gallon	1,000,000
Yearly Profit	383,760		70,240
Profit per ton of input refuse	\$4.21	-	\$ 0.772

TABLE 5. ECONOMIC ANALYSIS OF ETHANOL PRODUCTION FROM REFUSE

reduction costs of \$258,000 and \$172,000 are arrived at for the 60 and 40 percent paper contents, respectively.

An economic analysis is made in Table 5 for the two paper contents of 60 and 40 percent.

The prospects opened up by the figures in Table 5 are interesting. It should perhaps be emphasized that the 40 percent paper basis is an extremely low figure, and that 60 percent by weight paper content is an average value for urban municipal refuse. The process appears to have strong profit potential in areas where there are refuse paper contents in the region of 60 percent. Around 40 percent paper content, the disposal cost is not exorbitant, and the process is competitive with incineration if the present market price of ethanol holds steady. As the profitability is dependent on the market price of ethanol, the effect of price fluctuations is considered in the next section.

# MARKET PRICE FLUCTUATION

Considering the 60 percent paper content in refuse first, in the event of the market price falling,

the process does not lose money until the price falls below  $30.2\phi$  per gal. If incineration is the other alternative disposal method, then at \$3.82 per ton incineration costs, the plant can absorb a \$347,000 per year loss and can sell ethanol at  $21.4\phi$  per gal before incineration becomes competitive.

On the 40 percent paper basis, the plant already makes a loss of \$70,240 per year, and can absorb an extra loss of \$276,760 per year before incineration becomes competitive; i.e., the selling price can drop to 32.24 per gal.

As there is already a well-developed market for ethanol at 52¢ per gal (the 1965 consumption was  $710 \times 10^6$  gal), there would appear to be an incentive for the development of refuse hydrolysis to the commercial stage leading to its adoption in areas with 40-60 percent (or greater) paper contents in their refuse.

#### CONCLUSION

Table 6 summarizes the results obtained from the alternatives analyzed. Disposal costs are all given

TABLE 6				
Process	Product	Profit \$ Year	Capital Cost Per Ton of Daily Capacity \$	Remarks
1. Incineration	Disposal	- 347,000	8,000	Used as a perform- ance norm.
2. Destructive Distillation	+ Acetic Acid	- 515,000	32,000	Capital cost too high. Marketable Product
3. Composting	+ Compost	+214,000	10,700	Private venture (Cost to town \$3.24/ ton.) Rapid digestion process. Very poor market.
<ul> <li>Hydrolysis</li> <li>(60% paper)</li> <li>Hydrolysis</li> <li>(40% paper)</li> </ul>	<ul><li>+ Ethanol</li><li>+ Ethanol</li></ul>	+383,760 - 70,240	9,100	Based on a selling price of 40¢ /gal. Ready market.

on the basis of a 120,000 strong community. It is noted that the capital and disposal costs for incineration and composting have been deduced from existing plants with respective daily disposal capacities of 750 and 140 tons, and should be of the correct order of magnitude for a 249 ton per day plant. (Larger plants will have an economy of scale; therefore, their disposal costs will be lower than those in Table 11.)

On the basis of capital cost per ton, destructive distillation would probably be eliminated from any further consideration. When the yearly costs are considered, this process is rejected.

Incineration, composting, and hydrolysis are all competitive on installed capital cost. Incineration will continue to be used by many communities as it reduces the volume of refuse considerably in a hygienic manner, and is competitive with sanitary landfilling costs.

Composting may be viable in a few areas. However, the compost market has yet to be established in the United States, and this would be a strong determent against its adoption.

Ethanol production is very competitive with incineration and can make a profit at paper contents of order 60 percent. If the present price structure remains stable, its development should be vigorously pursued. The competitive capital cost, the ease of product handling, the established market and highpotential profitability, especially if the paper content of municipal refuse continues to rise, are persuasive factors for its adoption.

# **RECOMMENDATIONS FOR FURTHER WORK**

This paper grew from a study aimed at obtaining a viable means of refuse disposal. The ethanol yield predictions were based on the hydrolysis of wood, and obviously further work has to be done in the following areas:

1) Accurate kinetic data on the hydrolysis of paper. Preliminary tests at the Thayer School indicated that hydrolysis reaction rates were faster than those for wood under comparable conditions. If this is borne out by further work, either the allowable acid concentrations or the temperatures used may be lowered. This can reduce the materials costs. From accurate kinetic data, a pilot plant can be built and the predicted yields verified.

2) Accurate determination of the ethanol yield from paper hydrolysis followed by fermentation of the reducing sugars under varying fermentation conditions. This would be obtained from pilot plant tests.  Determination of the effluent liquod BOD and the most economical method of reduction. These would be obtained from pilot plant tests also.

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## HYDROLYSIS KINETICS

The kinetics of the hydrolysis of cellulose are most important and a brief review is now given. Saeman [19] has done extensive work on the kinetics of cellulose hydrolysis and the following material is summarized from his paper:

1) The sugars produced from cellulose are subject to decomposition on continued exposure to the hot dilute acid.

2) Two consecutive first order reactions occur: Cellulose  $k_{1-}$  reducing sugar  $k_{2-}$  sugar decomposition products, with the respective reaction rate constants  $k_1$  and  $k_2$ .

3) The rate of hydrolysis of cellulose and the decomposition of the sugar is a function of the acid concentration, temperature and time, and is independent of the digester size when the liquid-to-solid ratio is of the order 8:1 or greater. The greater the acid concentration and the temperature, the faster the reaction rates.

4) The energy of activation of the reaction is independent of the acid concentration, being 42,900 cal per (mole) for cellulose to sugar and 32,800 cal per (mole) for sugar to decomposition products, respectively.

5) Increasing the acid concentration or the temperature or both together, increasing efficiency of conversion of cellulose to sugar. In the temperature range 170-190 deg C, a 10 deg C rise in temperature gives 186 percent increase in the reaction rate for cellulose to sugar, and 125 percent increase in the sugar decomposition rate.

A plug flow continuous reactor is to be used, and can be analyzed in the same manner as a batch reactor. From Levenspiel [20] for two consecutive first order reactions in a batch or plug flow reactor, the following relationships can be derived.

For the reaction

$$k_1 \rightarrow R \xrightarrow{k_2} S$$

Commencing with a concentration  $C_{AO}$  of A, and no R or S present. (A represents cellulose, R – fermentable sugar, S – decomposed sugar.)

$$\frac{C_A}{C_{AO}} = e^{-k_1 t}$$
(1)

$$\frac{C_R}{C_{AO}} = \frac{k_1}{(k_1 - k_2)} \frac{(e^{-k_1 t} - e^{-k_2 t})}{(k_1 - k_2)}$$
(2)

$$C_{S} = C_{AO} - C_{A} - C_{R}$$
(3)

$$\frac{(C_R)}{(C_{AO})_{max}} = \frac{(k_1)}{(k_2)} \frac{k_2}{(k_2 - k_1)}$$
(4)

$$\max = \frac{1_{ge} (k_2/k_1)}{(k_2 - k_1)}$$
(5)

We now have the necessary kinetic picture to design a reactor using the rate constants from Saeman's work, but first the effects of higher temperature operation will be studied.

In order to illustrate the effects of acid concentration and temperature, the following table reproduced from Saeman's work [17] gives the ratio of rate constant,  $k_1$ , to rate constant,  $k_2$ , for various values of acid concentration and temperature.

Now, from equation (4), it is seen that the maximum yield of fermentable sugars increases as the ratio,  $k_1/k_2$ , increases. Thus, for a maximum sugar yield using the data of Table 7 *only*, the conditions of 1.6 percent H<sub>2</sub>SO<sub>4</sub> and 190 C would be chosen. However, it is known a posteriori that the acid costs can, perhaps, make the difference between profit and loss for the hydrolysis process. It is, therefore, desirable to obtain a high ratio of  $k_1/k_2$  for a lower acid concentration. This can be done by increasing the temperature of the process till the holding time t max from equation (5) is too short for good reaction control.

From Table 7, and an 0.4 percent  $H_2 SO_4$  concentration, it is seen that  $k_1/k_2$  is roughly doubled for a 20 C temperature rise from 170 to 190 deg C. As Saeman was limited by his apparatus to upper

	TABLE 7	
Temp., dec C	$H_2SO_4$ , percent	$k_1/k_2^a$
170	0.4	0.66
	0.8	0.84
	1.6	1.00
180	0.4	0.81
	0.8	1.07
	1.6	1.31
190	0.4	1.11
	0.8	1.36
	1.6	1.68

ak, = rate of hydrolysis of cellulose

k<sub>2</sub> = rate of decomposition of glucose

				and the second	
Temperature °C	Rate const. k <sub>1</sub>	Rate const. $k_2$	k <sub>1</sub> k <sub>2</sub>	Maximum sugar yield %	Residence time for max yield, min
170	.00355	.00534	.66479	29.58	228
180	.01045	.01219	.85719	33.99	88.5
190	.02936	.02685	1.0932	38.43	35.599
200	.07896	.05722	1.3799	42.8	14.8
210	.20385	.11816	1.7251	47.14	6.36
220	.50638	.23692	2.1373	51.28	2.818
230	1.21.32	.46209	2.6254	55.22	1.285

TABLE 8. KINETIC COMPUTATIONS FOR 0.4-PERCENT ACID CONCENTRATION IN TEMPERATURE RANGE 170-230 C

temperatures of 190 C, he did not obtain rate constants above this temperature. It is, however, possible to accurately predict these constants at higher temperatures, as Saeman obtained the activation energy of the reactions and accurate kinetic data at lower temperatures. The Arrenhius rate equation [20] can be used to predict the rate constants at various temperatures provided a verified rate constant at a known temperature and the reaction activation energy are available. The Arrenhius rate equation can be stated as:

$$k = k_0 e^{-E/RT}$$
(6)

where k is the rate constant to be determined at temperature T Kelvin;  $k_o$  is a reference constant determined from a given rate constant and temperature; E is the activation energy of the reaction in calories per (mole); R is the ideal gas low constant, 1.98 cal per gm mole, deg K.

The reference temperature for equation (6) is taken as 170 C and the reaction constants  $k_1$  and  $k_2$  for an 0.4 percent acid concentration are, from Saeman's work, 0.00355 min<sup>-1</sup> and 0.00534 min<sup>-1</sup>, respectively. The Arrenhius rate equation predictions can be verified, as the values of  $k_1/k_2$  at 180 and 190 C, and 0.4 percent acid concentration can now be predicted and checked against Saeman's values in Table 7. The unknown rate constants for higher temperatures are also calculated, and confidence can be placed in them if the agreement between predicted and measured values of  $k_1/k_2$  is close. A digital computer was used for all the calculations as extreme accuracy is required in evaluating the constants, maximum yield and residence times. The

rate constants,  $k_1$  and  $k_2$ , were calculated from 170 to 230 deg C in 10 C steps; the ratio,  $k_1/k_2$ , maximum yield of sugars and residence time for maximum yield were also computed. Table 8 gives the results.

The accuracy of the predictions of  $k_1/k_2$  in Table 8 with the known values in Table 7 is compared with Table 9.

Excellent agreement is obtained between the predicted and measured values; thus, one can have confidence in using the predicted values of  $k_1$  and  $k_2$  at higher temperatures. From Table 8, it is seen that going to higher temperatures pays off well, as the reaction proceeds much faster and the maximum sugar yield is greater. The hydrolyzing conditions are chosen as 230 C, and 0.4 percent acid concentration with a predicted maximum sugar yield of 0.5522 in a time of 1.285 min. A continuous plug flow reactor is to be used, and the aqueous sugar solution flash cooled to 100 C almost immediately on discharge to effectively quench the reaction and thus stabilize the yield at 0.55. (A batch reactor would have a range of

FABLE 9. COMPARISON OF k <sub>1</sub> /k <sub>2</sub> , RATIOS PREDICTED AT 180 C AND 190 C AND 0.490 ACID CON- CENTRATION WITH THOSE MEASURED BY SAEMAN					
Temperature deg C	$k_1/k_2$ Predicted	$k_1/k_2$ Measured	Percent Error		
180	0.857	0.84	+2		
190	1.093	1.11	-1.55		

residence times and, hence, maximum yield would not be obtained.)

As the hydrolysis kinetics of paper are unknown (although the kinetics of wood hydrolysis just used



FIG. 5 CONCENTRATION-TIME PROFILES FOR THE CONTINUOUS HYDROLYSIS OF CELLULOSE AT 230 C AND 0.4 PERCENT H<sub>2</sub> SO<sub>4</sub>

should be quite close if not identical), a measure of residence time control is required in the process design. This is provided for by having a control valve to regulate the volumetric flow rate of water through the constant volume reactor; thus, the optimum holding time can be found for maximum yield. (A kinetic study of paper hydrolysis is recommended for future work. A pilot plant reactor can then be designed to verify the yield predictions.) For reactor volume calculations, a residence time of 1.2 min is chosen as the reaction is still proceeding when the mix leaves the reactor. Fig. 5 shows the concentration time profile for cellulose, sugar, and decomposition products at the hydrolysis conditions of 230 C and 0.4 percent acid concentration. It is seen that a residence time of 1.2 min gives a yield of 54 percent at discharge conditions. Thus, there is effective leeway to quench the reaction and obtain maximum yield. Also, from Fig. 5, it is noted that 23 percent of the original cellulose will be intact, and this will presumably be recovered at the filters along with the Ca  $SO_4$  from the neutralization and the unhydrolyzables. The decomposed sugars (22 percent) will be dissolved in the liquid. The reactor volume can now be found.

## **REACTOR CAPACITY**

The continuous plug flow reactor capacity can now be obtained, based on 150 tons per day of paper from the 249 tons per day of refuse, plus 20 tons per day of nonpaper material such as garbage and plastics which are presumed to be nonhydrolyzable. A (nominal) liquid to paper ratio of 15 to 1 is used, as preliminary tests had indicated that for paper hydrolysis, liquid to solid ratios of 10:1 were barely adequate.

The hourly water-flow rate required, based on 150 T/D paper is:

$$\frac{150 \times 15}{24} \times \frac{2000}{8.34} = 22,500 \text{ gph}$$

The 170 tons per day of paper and nonhydrolyzables from the flotation treatment will be presumed to contain their own weight of water on entering the reactor. The volume equivalent of the input materials is taken as approximately:

$$\frac{170}{24} \times \frac{2000}{8.34} = 1500 \text{ gph}$$

Total volumetric flow rate = 24,000 gph. The required reactor volume to give 1.2 min residence time is then:

$$\frac{24,000 \times 1.2}{60} = 480 \text{ gal}$$

As the reactor volume is fixed at 480 gal, based on 150 tons per day of paper (60 percent paper content

in the refuse), it is necessary to find the residence time which will occur for 40 percent paper content in the refuse, i.e., a paper load of 100 T/D plus 20 T/D of nonhydrolyzables.

The hourly water-flow rate for paper hydrolysis is now:

$$\frac{100 \times 15 \times 200}{24 \times 8.34} = 15,000 \text{ gph}$$

and the approximate flow rate excess caused by the 100 T/D of paper plus 20 T/D nonhydrolyzables is:

$$\frac{120 \times 2000}{24 \times 8.34} = 1060 \text{ gph}$$

Total volumetric flow rate on a 40 percent paper basis is then 16,060 gph. The residence time in a 480-gal capacity reactor is then:

$$\frac{480 \times 60}{16,060} = 1.74 \text{ min}$$

From Table 8, the maximum yield for a residence time of 1.74 min is roughly 0.53 of the cellulose at a temperature of roughly 227 C. Thus, the yield from the 40 percent paper content will be less than that for 60 percent paper content.

The 95 percent ethanol yield calculations can now be carried out.

# ETHANOL YIELD CALCULATIONS

The ethanol vield is calculated for two values of paper content in the refuse, 60 and 40 percent by weight (dry basis), respectively. Most paper is essentially pure cellulose plus 2 - 3 percent of whatever filler is used to give it its desired characteristics, the only exception is newsprint which is modified groundwood and contains roughly 60-65 percent by weight of cellulose. As a major portion of the paper in refuse is newsprint, a median value of 75 percent is taken for the cellulose content of all papers in the refuse. The remaining 25 percent is assumed to be fillers plus residual unhydrolyzables from the newsprint, and will be removed from the process by filters downstream of the reactor. After the sugars are made, they must be fermented. The "fermentability" of the sugars is at present unknown, but must be in the same region as those from wood hydrolysis. Reference [17] gives the fermentability of wood sugars as 85 - 96 percent depending on the

type of bacteria used for fermentation. These values were obtained around 1945, and it is assumed that present-day fermentation technology can handle the same type of sugars from paper hydrolysis. The fermentability is, therefore, taken as 95 percent. (This is another area for future work.) Table 10 itemizes the stages in calculating the final 95 percent (190 proof) ethanol yield from 60 and 40 percent paper contents, respectively.

The materials requirements are calculated next.

# MATERIALS REQUIREMENTS

The materials requirements are obtained as follows; (60 percent) and (40 percent) denote the 60 and 40 percent basis requirements, respectively.

#### SULFURIC ACID

0.4 percent of 24,000 gal of water per hr gives an acid requirement of 9.6 T/D (60 percent) and 0.4 percent of 16,000 gph gives 6.65 T/D (40 percent). Can be purchased in bulk at \$25/ton.

# Ca CO<sub>3</sub>

To effectively neutralize the  $H_2SO_4$ , equimolar quantities of CaCo<sub>3</sub> are rquired, i.e., 9.6 T/D (60 percent) and 6.65 T/D (40 percent). Can be purchased in bulk, crushed to 300 mesh size, at \$3.50 per ton.

#### WATER

For hydrolysis purposes, 576,000 gal per day (60 percent), 384,000 gal per day (40 percent).

For flotation tank make-up and ethanol condenser duty, roughly 50,000 gal per day is required for both 60 and 40 percent bases. Total water requirement 626,000 GPD (60 percent), 434,000 GPD (40 percent).

This is assumed to cost 10¢ per 1000 gal.

#### ELECTRICITY

The total connected horsepower is roughly 300. Also lighting requirements have to be met, irrespective of paper content, the total power cost is estimated at \$125 per day for both bases.

# FUEL OIL

Process heating requirements are evaluated as follows. An enthalpy balance is performed around the reactor complex. For the 60 percent basis, the inputs are 1500 gal water per hr at 20 C contained in the input refuse to the reactor, and 22,500 gal

TABLE 10. 95-PERCENT ETH	ANOL YIELD CALCULATIONS	
Paper content % in refuse Paper input T/D Cellulose (75% of paper) T/D	60 150 112.5	40 100 75
Maximum available sugar, evaluated from hy equation, = cellulose $\times \frac{180}{162}$ T/D	drolysis 124	83.5
Net sugar yield at 55% conversion for 60% pr (1.2 min res. time) 53% conversion for 40% pr content (1.74 min res. time) T/D	aper paper 69	44.4
100% pure ethanol yield from sugars, evaluate from fermentation equation = sugar $\times \frac{92}{180}$	35.3	22.6
Net ethanol yield after fermentability allowance = .95 × ethanol T/D	33.6	21.4
95% ethanol yield T/D	35.3	22.5
Gallons per day production based on specific gravity of 0.79	10,780	6,850

water pr hr at X C. The output is 24,000 gal water per hr at 230 C.

Thus,

 $1500 \times 20 + 22,500 \times X = 24,000 \times 230$ 

Thus,  $X = 244^{\circ}C$ 

Now, the feedwater heater receives 22,500 gal of water per hour at roughly 210 C from the heat exchangers in the flash cooling section. (A process heat loss of 6 percent has been used to determine the temperature of the water from the heaters.) Thus, the energy requirements for feed heating are:

$$(244 - 210) \times \frac{9}{5} \times 22,500 \times 8.34 = 11.5 \times 10^{6}$$

say

and

 $7.55 \times 10^{6}$ 

say

 $8 \times 10^6$  Btu/hr

The distillation column reboiler energy balance yielded  $15 \times 10^6$  Btu/hr (60 percent) and roughly  $11 \times 10^6$  Btu/hr (40 percent).

The total energy requirements are then:

 $27 \times 10^6$  Btu/hr (60 percent)

and

# $19 \times 10^6$ Btu/hr (40 percent)

Using fuel oil with a calorific value of 140,000 Btu/gal and allowing for combustion inefficiency, roughly 4900 (60 percent) and 3900 (40 percent) gal per day will be required at a cost of \$0.0683 per gal when purchased in bulk. Table 11 summarizes the materials costs for the two paper contents.

The next section deals with the refuse mass balances.

## Refuse Mass Balances

In order to assess the amount of solid refuse which will require disposal, balances are carried out in Table 12 on a dry basis. It is recognized that the residue will be wet and therefore heavier, but its volume will not be greater than that of comparable dry material. (In fact, it will be much less as the refuse will now be well compacted.)

TABLE 11	DAILY MATERIAL	COSTS
Sulfuric acid	\$240	\$161
CaCO <sub>3</sub>	33.6	22.6
Water -	62.6	43.4
Electricity	125	125
Fuel Oil	365	266
Daily Total Cost	\$826.2	\$618.0

## BIOCHEMICAL OXYGEN DEMAND REDUCTION (BOD)

The BOD demand of the waste liquor is an imponderable at present, and attempts can only be made to estimate this very important value. For a typical fermentation industry such as brewing, the BOD of the waste liquors can range from 420-1200 ppm [25]. On the other hand, in the roofing felt industry which uses salvaged paper as its raw material, the BOD of the waste liquors can run as high as 6000 ppm; the usual method of treatment in this industry is fermentation followed by activated sludge with mechanical aeration [26]. The unknown BOD value of the waste liquor from the hydrolysis process will be taken as 5000 ppm, and will be assumed to be in acceptable discharge condition from the waste treatment section at a value of 100 ppm.

An order of magnitude cost of treating these liquors can be extrapolated from the roofing felt industry costs which are of the order  $3\notin$  per lb/of BOD removed [26].

The daily amount of BOD removal from the roughly 576,000 GPD (60 percent) effluent, is 23,500 lb, and from the roughly 384,000 GPD (40 percent) effluent is 15,700 lb; the respective yearly treatment costs at 3¢ per lb are \$258,000 (60 per-

TABLE 12. REFUSE MASS BALANCES (DRY BASIS)				
	60%	paper	40% P	aper
	Solid,T/D	Liquid,T/D	Solid,T/D	Liquid T/D
Input refuse	249		249	
Separated out	- 79		129	
Nonpaper group carried				
through into reactor	20		20	
Non-cellulose in paper	37.5		25	
Unhydrolyzed cellulose				
(23% of gross cellulose)	26		18	
Cellulose converted to		(2)	0.10.0	20
Collulate converted to		62		39
decomposed sugars		24.5		18
decomposed sugars			1042	
Totals	162.5	86.5	192	57
INPUT CHEMICALS				
$H_2SO_4$		9.6		6.35
CaCO <sub>3</sub>	9.6		6.35	
OUTPUT CHEMICALS				
H <sub>2</sub> SO <sub>4</sub>	13.5		8.9	
Water and $CO_2$		5.7		3.8
2	S			
Total solids for disposal by				
sanitary landfill	176 T/D		200.9 T	/D

412





cent) and \$172,000 (40 percent). It is recognized that the above costs are approximate and the actual BOD demand and cost of treatment are two areas which will need thorough investigation.

The installation of a treatment plant for the process employing fermentation followed by activated sludge is estimated to cost in the region \$250,000 [26].

# ORDER-OF-MAGNITUDE PLANT COST ESTIMATION

The principal items of capital expenditure and their capacities are detailed on the expanded flow diagram of Fig. 6. References [21-24] were used to estimate plant sizes and costs. When available, estimates from leading plant manufacturers were used.

#### Concrete Storage Hoppers

Three days refuse storage capacity is required to level out flunctuations in supply and allow continuous plant operation.

Design capacity		933 tons
Volume at 300 lb/cu yd	=	6200 cu yd
Estimated cost	=	\$100,000

Pulverizers and Shredders

The refuse is pulverized prior to separation, and the light fraction (mainly paper) shredded after separation. The capacity of each unit is to be 40 tons per hr to allow for the batch operation of the flotation separator.

Cost/pulverizer unit	\$35,000
Total cost	\$70,000

#### Separation Section

The design capacity will be 13 tons per hr, and when operating on a half hourly batch basis, 45 cu yd of refuse will be processed at any one time. Hence, an agitated bed-wash tank of 90 cu yd capacity plus a dense refuse conveyor and water-cycling equipment will be needed. The estimated cost of this section is \$50,000.

#### Reactor

This will be of 480 gal capacity fitted with an agitator. The construction is to be of stainless steel for duty at a pressure of 420 psia and a temperature of 230 C. The cost is estimated at \$50,000.

#### Neutralizer

Will be to the same specifications as the reactor. Estimated cost \$50,000.

## Feed Water Storage Vessel

A 5000-gal capacity carbon steel vessel is provided to store the feed water to the reactor and act as a reservoir for the process feed water which has seen duty as the coolant for the flash chambers. The vessel pressure is 550 psia. Estimated cost \$10,000.

## Paper Slurry and Feed Water Pumps

The finely shredded cellulose material and light unhydrolyzables are pumped to the reactor in the form of a slurry whose consistency can be varied by controlling a by-pass water bleed from the main line to the reactor. This bleed will help preheat the cellulose prior to insertion in the reactor where it encounters the remainder of the feed water and the acid. A positive displacement pump is envisaged for the slurry pump: g duty, possibly of the multiple chamber ram-type. Feed water condensate and recycling pumps are also required. No accurate cost estimate is set on the pumps, and their purchase cost is included in a \$200,000 appropriation under the heading miscellaneous.

## Flash Chambers

These come in two sections: the first set will cool the liquor to 100 C in 9 stages of 14.3 C temper-

ature drop per stage; the second set will cool the liquor from 100 to 40 deg C in four stages of 15 C temperature drop per stage. The feed-water flow of 22,500 GPH is used as the coolant for 24,000 GPH liquor flow with an inlet temperature of 20 C, and flows counter-current to the liquor flow. The energy loss from the liquor to the sorroundings is accounted for by an efficiency of 94 percent on the overall energy balance on the heat exchanger. This means that the coolant water increases its temperature by the same amount that the liquor decreases its temperature in each stage. Thus, the feed water gains 150 C in each of the stages in the first flash section, and 14.3 C in each of the stages of the second flash section. For calculation purposes, the effective log mean temperature difference is taken as 10.85 C for each stage. The heat-flux constant in each stage at  $5 \times 10^{6}$  Btu per hr. Using an overall heat-transfer coefficient of 300 Btu per hr sq ft, the required surface for each stage is of the order 1000 sq ft. The total estimated cost at \$12 per sq ft for the flash chambers and exchangers together is then \$108,000.

#### Filters

Diatomaceous earth pressure filters will be used with a flow rate of 8 gal per sq ft per hr. Three 1000 sq ft filters will be required at a cost of \$23,000 each. Total filter cost \$69,000.

TABLE 13				
Item		Cost		
Storage hoppers		\$100,000		
Pulverisers	70,000			
Screening Section	50,000			
Reactor	50,000			
Neutraliser	50,000			
Feed Storage	10,000			
Flash Chambers	108,000			
Filters		69,000		
Vats	35,000			
Bubble cap colum	20,000			
Heat exchangers	15,000			
Boiler	35,000			
B.O.D. reduction	250,000			
Erection & Miscel	laneous Plan	200,000		
Total Equipme	ent Cost	1,062,000		
Building		1,200,000		
	Total Capital Cost	2,262,000		

# Fermenting Vats

The normal fermenting time required is of the order of 20 hr; hence, the provision of 24-hr vat storage in seven 10,000 gal capacity redwood vats is estimated to cost \$35,000.

## Bubble Cap Column

Equilibrium stage calculations showed a suitable bubble cap column to be 3 ft dia  $\times$  30 ft high with 20 plates using a reflux ratio of 4 lb of reflux per pound of product. The estimated cost is \$20,000.

#### Heat Exchangers

Three heat exchangers will be required for feed heating, reboiler, and product cooling. The respective areas and costs are 600, 650, and 700 sq ft, and \$4500, \$5000, \$5,500, totaling \$15,000.

#### Steam Boiler

A package boiler capable of supplying 40,000 lb per hr process steam is estimated to cost \$35,000.

## Miscellaneous

Erection and miscellaneous equipment costs such as feed and slurry pumps are estimated at \$200,000.

#### Building

A 3-story building with a plan area of 20,000 sq ft is required. For an aluminum-on-steel process building complete with floors, heating, plumbing, and lighting an order of magnitude cost is \$20 per sq ft. The estimated building cost is \$1,200,000. A cost tabulation is given in Table 13.