WASTE TREATMENT STRATEGY— A GUIDE THROUGH THE MAZE

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Abstract

This paper discusses the key elements of the current EPA guidelines for the leather tanning and finishing industry for subcategory 1 tanneries. Suggestions are made for in-plant and end-of-pipe waste treatment, stream separation and water conservation and reuse.

Introduction

The recently promulgated EPA effluent guidelines for the leather tanning and finishing industry (November 1982) are less demanding than those proposed in the recent past. The changes most responsible for the new leniency are recognition of the unreliability of technology transfer, and the realization that clean water objectives had outrun the concept of cost reasonableness. The new limitations are therefore more focused on the costs and demonstrated capabilities of technology now available within the industry. While the resulting standards may come as some relief to the industry, it would be wise to look beyond the current round of standards. Compliance with the standards may not meet requirements of individual states nor EPA's long-range clean water objectives. The purpose of this paper is to discuss key elements of the regulations and through that discussion propose a long-term treatment strategy. To provide a frame of reference, a hypothetical existing tanning process will be described. Thereafter, attention will focus upon in-plant and end-of-pipe waste treatment, stream separation, and water conservation and reuse.

Waste Treatment Technology Bases

With the cooperation of the Tanners Council of America, the EPA conducted data gathering efforts from 1974 to 1980 (1). In this paper our discussion will be limited to subcategory 1 tanneries (hair pulp, chrome tan, retan-wet finish). However, the information presented has direct application to other hair pulp tanneries as well. A partial summary of these data is shown in Table I. The third and fourth columns provide the EPA's best estimate of the long-term average final effluent concentrations achievable using treatment appropriate under PSES (Pretreatment Standards for Existing Sources) and BPT (Best Practicable Control Technology currently available) standards for the subcategory 1 indirect and direct discharger, respectively. These data form the basis for the EPA's effluent guidelines and limitations promulgated in 1982 (2) (see Table II). The compliance date for the PSES regulations is set as November 25, 1985.

For the subcategory 1 indirect discharger, EPA's technology basis consists of 1) segregation of the beamhouse waste for catalytic oxidation of sulfide, 2) equalization and coagulation/sedimentation of the tanyard waste, and 3) neutralization of the combined waste (Figure 1). Sulfide oxidation is carried out as a batch operation with $MnSO_4$ as the catalyst. The pur-

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TABLE I
WASTEWATER CHARACTERISTICS[1] SUBCATEGORY ONE

Parameter		Raw wastewater Median	Raw wastewater Mean	BPT effluent[2] Direct discharge	BPT effluent[2] Indirect discharge
BOD_5	(mg/l)	1,184	1,078	40	–
	(kg/kkg)	53.72	63.47		
COD	(mg/l)	3,078	3,569		
	(kg/kkg)	186.70	187.58		
Total suspended solids	(mg/l)	1,582	1,913	60	–
	(kg/kkg)	102.12	104.00		
Oil & grease	(mg/l)	493	990	20	–
	(kg/kkg)	27.54	57.80		
Total Kjeldahl nitrogen	(mg/l)	200	204	–	–
	(kg/kkg)	12.06	12.05		
Ammonia	(mg/l)	69	63		
	(kg/kkg)	3.86	3.85		
Sulfide	(mg/l)	59	64	–	9
	(kg/kkg)	2.86	3.35		
Total chromium	(mg/l)	47	79	1	5
	(kg/kkg)	2.75	4.76		
Float ratio	(gal/lb)	6.5	6.83	–	–
	(l/kg)	54	57		
pH					7-10

[1]EPA Development document for effluent guidelines and standards for the leather tanning and finishing industry (1982).
[2]EPA Technology basis for long-term average concentrations.
Concentrations are provided only for pollutants scheduled for regulation.

pose of equalization is to dampen the wide fluctuations in effluent flow rate and composition typical of the tanning process. No treatment is achieved in equalization itself, however, the uniformity of effluent produced by this process improves the consistency of performance of subsequent treatment. The degree of uniformity required is dependent upon the nature of the subsequent treatment—biological treatment being far more demanding in most instances than physical processes. According to the EPA analysis, holding volumes required for adequate equalization are 24 hr for pretreatment and 48 hr when followed by biological treatment. From the design standpoint, equalization time is the ratio of treatment basin volume to the flow rate. Thus, the smaller the volume of water used in-plant, the smaller the volume (and cost) of this end-of-pipe treatment step.

The next step in treatment is coagulation/sedimentation (and fat skimming). The purpose of this step is primarily to reduce total suspended solids, specifically chrome. Indeed, chrome removal is the overriding reason why coagulation/sedimentation is recommended rather than simple sedimentation. In both processes chrome is precipitated under alkaline conditions. The precipitate however, is finely suspended. In the coagulation/sedimentation process lime and anionic polyelectrolyte are added to the tanyard wastewater to produce a more easily separable floc. After the tanyard effluent is clarified, beamhouse and tanyard

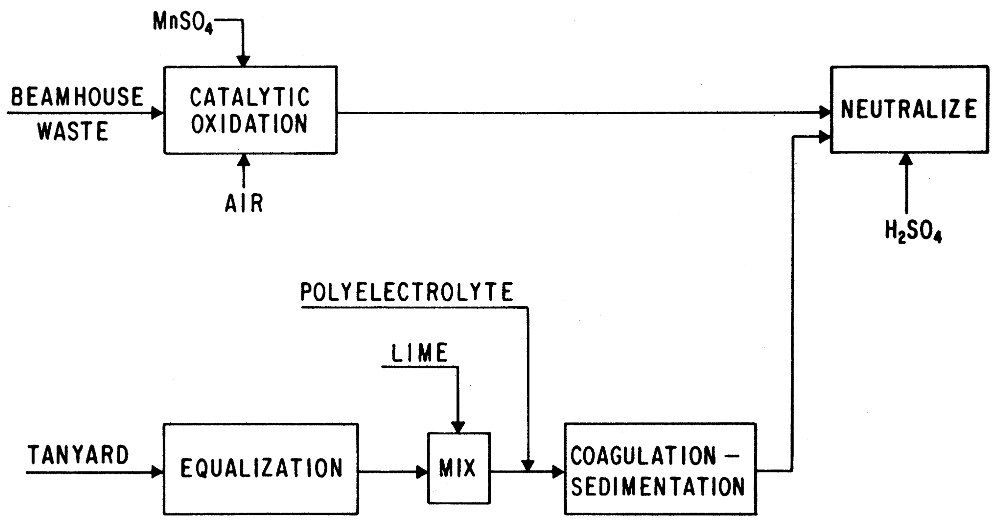

FIGURE 1. — Pretreatment Standards for Existing Sources (PSES)—technology basis.

wastewaters are combined, neutralized, and discharged. As indicated in the last column of Table I, the combined effluent is expected to contain a long-term average concentration of 9 mg/l sulfide and 5 mg/l total chrome (allowable effluent concentrations are 24 mg/l and 8 mg/l, respectively). By back-calculating from these figures, it is estimated that sulfide oxidation should reduce beamhouse waste to 12.8 mg/l sulfide, while coagulation/sedimentation is expected to reduce tanyard waste to 19 mg/l total chrome. The EPA did not speculate on or regulate TKN, COD, or BOD_5 removal. However, much of the suspended solids in tanning waste is proteinaceous, so that this process also removes a substantial amount of TKN and associated BOD_5 and COD.

Because direct discharges are under the NPDES (National Permit Discharge Elimination System) program, the EPA has not (as yet) found it necessary to issue a compliance date for BPT. Nonetheless, standards for four pollutants have been promulgated: BOD_5, TSS, grease and oil, and total chrome. EPA's BPT treatment for our hypothetical subcategory 1 tannery consists solely of end-of-pipe treatment (Figure 2) and includes equalization, coagulation/sedimentation, extended aeration, and clarification. As already mentioned coagulation/sedimentation has a distinct advantage in chrome removal, and even where chrome is of no real concern, use of coagulation/sedimentation ahead of biological treatment will pay dividends in volatile suspended solids removal. While it is true that biological treatment alone can remove much the same material, sedimentation operations, in general, provide greater reliability and consistency of performance than biological treatment at a lower cost per lb of solids removed. Thus, coagulation/sedimentation is recommended as a pretreatment. Attempts at eliminating physical/chemical pretreatment of waste have to date proved unsatisfactory. For example, direct application of waste to aerated and nonaerated lagoons produces poor quality effluent in almost every respect and requires more land area. Further, while direct anaerobic digestion followed by activated sludge treatment appears attractive, it must be questioned on the basis of the total mass of solid waste generated (including retained water), and on the basis that ammonia is generated as protein is digested.

Biological treatment processes considered by the EPA in their analysis included aerated and nonaerated lagoons, activated sludge systems, rotating biological contractors (RBC's),

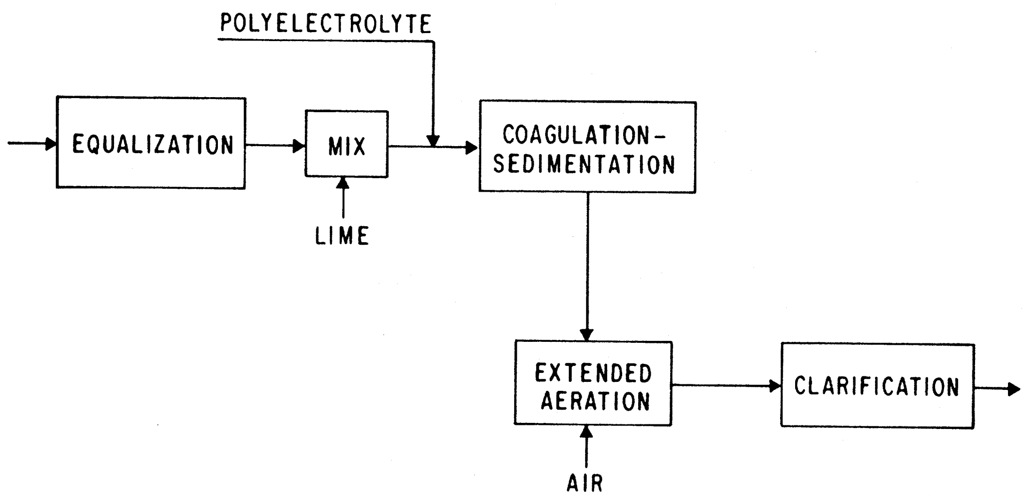

FIGURE 2. — Best Practicable Treatment (BPT)—technology basis.

TABLE II
PRETREATMENT STANDARDS FOR EXISTING SOURCES
SUBCATEGORY 1 TANNERY EFFLUENT GUIDELINES

Pollutant or property	PSES		BPT	
	Maximum for any 1 day	Maximum for monthly average	Maximum for any 1 day	Maximum for monthly average
	Milligrams per liter (mg/l)		kg/kkg (or lb/1000 lb) of raw material	
Sulfide	24	—	—	—
Total chromium	12	8	0.23	0.09
pH	Within the range 7.0 to 10.0		Within the range 6.0 to 9.0	
BOD_5	—		9.1	4.1
TSS	—		13.2	6.0
Oil & grease	—		3.8	1.7

and trickling filters. All are in use to some degree in the tanning and finishing industry. Of these, only properly designed activated sludge treatment has demonstrated long-term consistent performance within the industry (too little data are available on RBC's). Of the activated sludge treatment options available, the EPA chose extended aeration treatment as the technology basis because of its well-documented consistency, reliability, and thoroughness year round in treating high-strength waste. Anticipated performance of coagulation/sedimentation followed by extended aeration is shown in Table III.

Although the emphasis is placed on biological treatment per se, care must be taken not to underestimate the importance of final clarification. This is particularly true of the slow settling sludge typical of extended aeration processes, where up to 50 percent of the effluent BOD_5, for example, may be attributed to suspended solids loss.

Behind The Technology Basis

EPA's PSES and BPT cost and technology basis presents a number of apparent anomalies. Although the promulgated EPA effluent guidelines for the leather tanning and finishing industry are less demanding than those proposed in the recent past, EPA concessions to cost

TABLE III
BPT TREATMENT CAPABILITIES

	Coagulation/sedimentation[1] (% removed)	Effluent concentration from biological treatment, mg/l
BOD_5	50	40
TSS	95	60
Grease & oil	80	20
Total chrome	(90) 8 mg/l residual	1
Sulfide	—	≤6
COD	60	500
TKN	15	95
NH_3	—	90
Phenol	—	<.2

[1]End-of-pipe treatment.

reasonableness and demonstrated technology may well provide only temporary sanctuary. EPA's clean water objectives remain essentially intact.

First note that although sulfide is regulated for the indirect discharger, it is not regulated for the direct discharger. Clearly, if one were to make a choice solely on the basis of environmental impact, one would choose the reverse. It is true that direct discharge BOD_5 requirements will, in part, account for oxygen demand due to sulfide. Furthermore, sulfide oxidation was considered by the EPA in the next more expensive BPT treatment option. Therefore, the absence of BPT regulation must be regarded solely as a concession to cost reasonableness. With or without regulation, sulfide removal must be considered.

Second, note that implementation of EPA's cost and technology basis for BPT treatment is expected to result in substantial removals of pollutants not presently under regulation. This suggests that the absence of these pollutants from the BPT (or BAT) guidelines is not a function of cost, but rather the lack of demonstrated removal capability within the industry. It would follow then, that industry-wide compliance with guidelines by use of BPT type technology may result in the "demonstrated capability" necessary to regulate pollutants now absent from the guidelines, thus opening the door to further regulation. Of particular concern are COD, TKN, and ammonia, all of which were proposed for regulation in prior guideline drafts (3, 4). It would be patently unwise to ignore these unregulated pollutants.

Third, note that in Tables I and III, the quantities of pollutants are tabulated in terms of concentration. This is not mere convenience; the amounts of these pollutants that can be removed is a direct function of pollutant concentration. It is axiomatic in waste treatment that without resorting to exotic means, thorough primary sedimentation, biological and final clarification will leave predictable residual levels of conventional pollutants in the effluent (i.e., 20-25 mg/l BOD_5, 20-25 mg/l TSS, and 15-20 mg/l grease and oil). The result is that each unnecessary liter of water used in processing results in a corresponding amount of unnecessary pollution. Saving water and treating waste before it becomes unnecessarily dilute makes good economic sense. These approaches are discussed below.

In-Plant Strategies

It cannot be overstated that water conservation is the foundation of any successful approach to waste reduction. It is not to be looked at as an attempt to whittle away at pollution at the expense of product quality. In-plant waste reduction can best be accomplished through a combination of stream segregation treatment and water use reduction/conservation. Both approaches are based on the premise of improved treatment efficiency and cost by treating small volumes of highly concentrated waste. At the end of the pipe we have already seen that the hold-up volume required for equalization prior to biological treatment is a direct function of effluent flow. Similarily, the sizes of primary and final sedimentation basins are likewise a function of effluent flow rate.

The "mean" subcategory 1 tannery has been shown to consume 57 kg of water for every kg of hide processed. Based on their survey, the EPA concluded that present tanneries could, at minimal expense, reduce their water usage to 45 kg/kg. USDA scientists (5) and Aloy *et al.* (6) have found that water usage can be decreased even more by resorting to batch washing exclusively. The distribution of waste produced by the USDA process which utilizes batch washing exclusively is shown in Table IV. To further examine these figures, a water use table (Table V) based on the work of Thorstenson (7) has been constructed which compares USDA batch washing to continuous rinsing. In addition, a hypothetical tanning process is outlined to allow us to further subdivide the process steps.

The single biggest culprit appears to be the "final continuous washes." As reported by Aloy *et al.* (6) there appears to be little leather quality justification favoring continuous washing over more water conservative batch processing. Note also that about the same amount of fresh water is used in the dirty (beamhouse) operations as is used in the clean (tanhouse) operations. At ERRC we have conducted research on three process schemes to address these issues. The first of these process schemes utilizes countercurrent processing techniques (see Figure 3) (8). There are three basic elements in this approach. In the first element continuous washes (such as that used for reliming) are replaced with a suitable series of batch washes. In any series of washes, less "waste" is removed in each wash so that each succeeding wash is cleaner than the one before. That being the case, effluent from the last wash in reliming, for example, should be suitable for use as the relime float for the next batch of hides.

The second element in countercurrent processing concerns process step crossover. Where reliming ends and bating begins is largely a matter of definition. The last wash in reliming might just as well be considered a first step in preparation for bating. Under this latter definition, there would appear to be no harm in using a wash containing a small amount of $(NH_4)_2SO_4$. Thus, there would also appear to be no harm in using a "cleaned up" bate solution as a preparatory relime wash.

TABLE IV

SOURCE OF TANNERY POLLUTANTS—USDA STANDARD PROCESS[1]

	Pollutant, % of total								
Waste stream	TKN	NH_3 - N	Non NH_3 - N	TSS	COD	BOD	Oil and grease	Sulfide	Chrome
Soak	8	7	8	10	10	18	13	—	—
Unhair	42	6.5	56	49	35	59	21	66	—
Relime and 2 washes	23	7	29	25	31	12	36	30	—
Bate and 1 wash	22	67	3	6	6	1	4	2	—
Pickle	3	6.5	2	2	4		2	.5	20
Tan and 1 wash	2	4	2	2	8	5	2	1	63
Retan, color, fat liquor, and 1 wash	—	—	—	2	2	2	3	—	17
Finish	—	—	—	4	4	4	18	—	—

[1] From Taylor (5).

TABLE V
WATER USAGE IN HIDE PROCESSING

USDA Standard			Thorstensen
Subprocess	Float ratio	Total	Total
Soak	1.0	1.0	6.9
Unhair	1.0	1.0	6.9
Relime	2.0	4.5	13.8
Wash	1.25		
Wash	1.25		
Bate	1.25	2.5	8.3
Wash	1.25		
Pickle	0.5	2.2	2.1
Tan	0.8		
Wash	0.9		
Retan, color, fat liquor	4.5 (2.0[1])	13	13.8
Wash	8.5 (3.0[1])		
Finish	6.9	6.9	6.9
Total		31.1	58.7

[1] Aloy *et al.* (6).

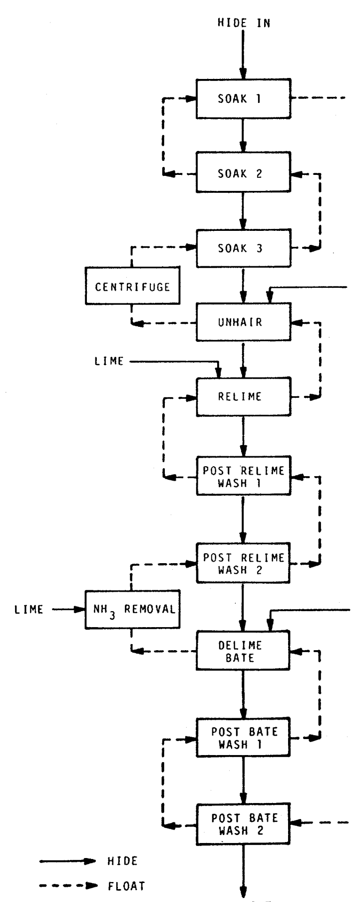

FIGURE 3. – USDA countercurrent hide processing system.

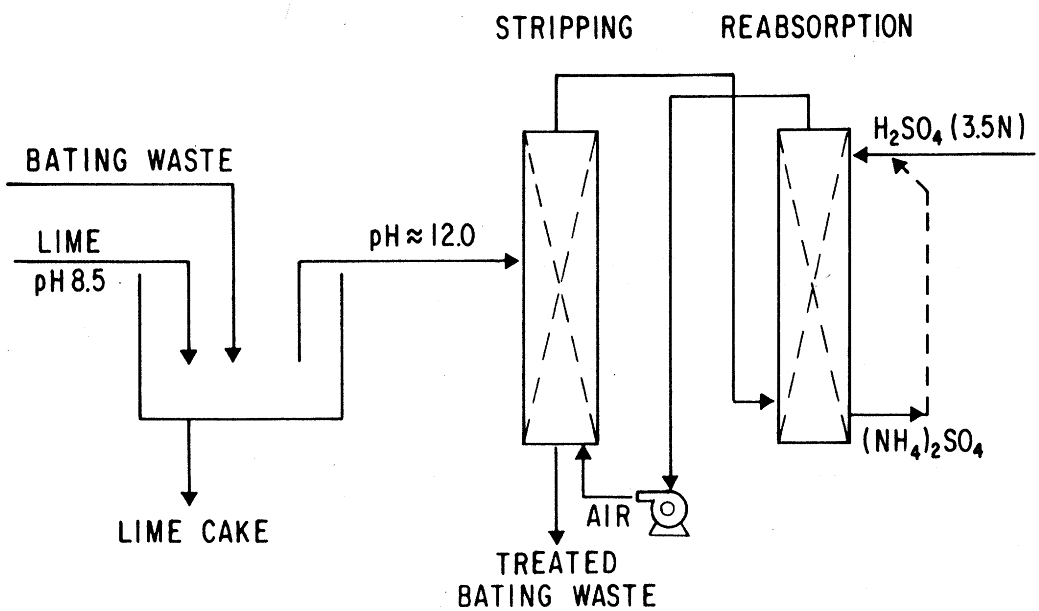

FIGURE 4. — Removal and recovery of ammonia.

The second process system now under investigation at ERRC for bate clean-up is shown in Figure 4. Ammonia stripping/reabsorption is best used in conjunction with the countercurrent process to assure maximum removal of ammonia and other pollutants from small volumes of waste. As we envision it, lime will be added to the bating stream in a sedimentation tank. Lime will serve the dual function of converting ammonia from a dissociated to an undissociated form (99 percent undissociated at pH 12) while at the same time enhancing precipitation of solids. The bate will then be pumped into an ammonia stripping and reabsorption unit. Here approximately 98 percent of the entering ammonia will be swept into a circulating air stream and subsequently reabsorbed into sulfuric acid, the resulting ammonium sulfate to be used in subsequent bate solutions. Using Tables IV and V as a basis for calculation it is estimated that by means of such a process a 21 percent reduction in effluent nitrogen can be achieved in a treatment handling less than 8 percent of the total process float.

The EPA technology basis for in-plant treatment focuses largely on sulfide oxidation and coagulation/sedimentation of beamhouse waste and the treatment of tanning effluent for chrome. These processes have been previously described. Treatment of beamhouse waste and the effect of that treatment on the total effluent are summarized in Table VI. Results of an alternative scheme studied at ERRC (the ESL process) are also presented. Here only the unharing waste is treated. The ESL process (9) involves recovery of sulfide and other pollutants using adiabatic vacuum flashing. It too is best used in conjuction with countercurrent reuse. To obtain the results shown in Table VI, relime solution is fortified with sulfide and used for unhairing of the next batch of hides. The spent unhairing solution is then treated to remove TSS, sulfide, and proteins. The clear solution can then be used for soaking. The ESL process (Figure 5) consists of fat skimming, carbonation and precipitation for lime removal, acidification, degasification, and filtration for sulfide and protein removal (10). In the process, waste is acidified in-line and pumped to a vacuum degasification chamber where hydrogen sulfide is removed and readsorbed in sodium hydroxide. The degasified waste is then pumped to a filter press for protein removal. As can be seen,

TABLE VI
COMPARISON OF BPT VS ESL UNHAIRING WASTE TREATMENT

	Removal as % of total				
		Coagulation/sedimentation		ESL process	
	Catalytic oxidation	Beamhouse waste	Tannery waste	Beamhouse waste	Tannery waste
Sulfide	∿9 mg/l[1]		<6 mg/l[1]		<6 mg/l
BOD		50	45	86	61
TSS		90	80	<95	70
Grease & Oil		(∿90)	55	99	56
COD		65	40	85	56
TKN		50	45	86	56
NH_3		–	–	–	–
Total volume treated (kg/kg)			21.6		1.0

[1]Concentration of sulfide remaining after treatment.

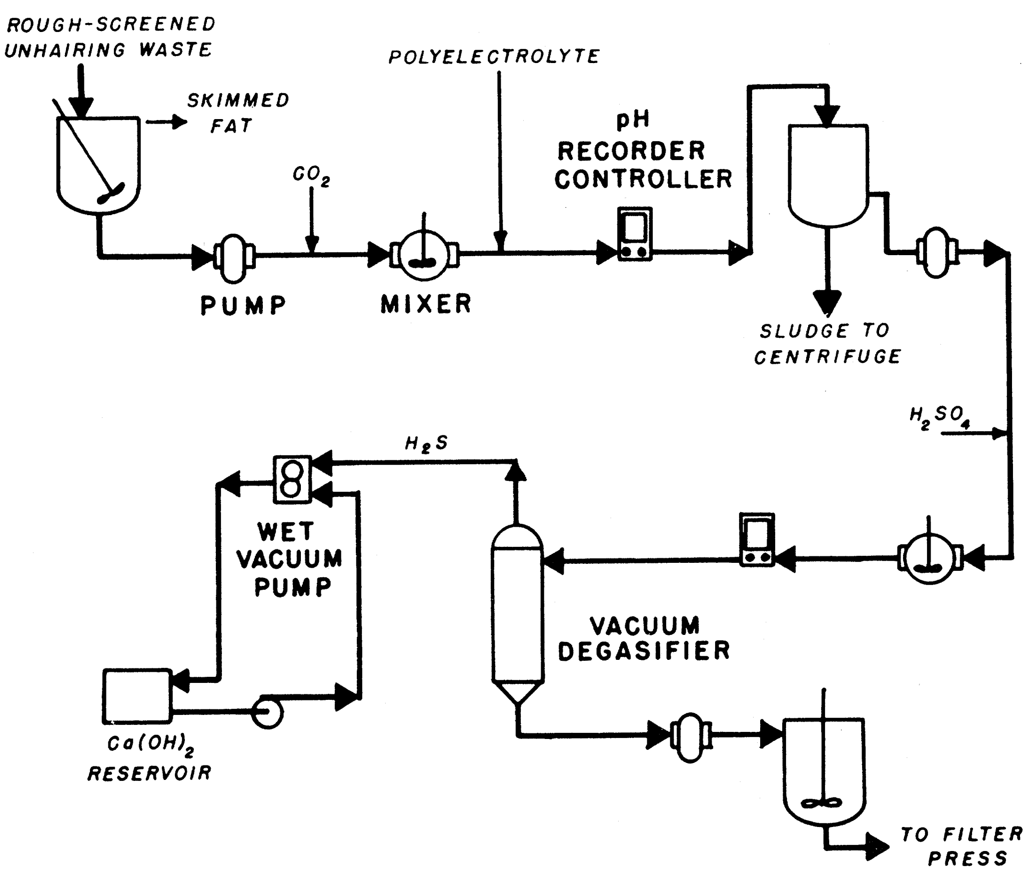

FIGURE 5. – Treatment of lime sulfide unhairing waste.

although the overall results are much the same in either treatment, there are, we feel, two important differences. The volume of effluent treated in the ERRC process is less than 5 percent of the EPA technology basis of 22.8 kg/kg of beamhouse waste (i.e., 40 percent of the total flow). Second, most of the sulfide is recovered for reuse. Also, catalytic oxidation of the sulfide in the beamhouse waste is avoided. Potentially salable by-products are obtained which may create savings in sludge disposal costs. These are advantages worthy of consideration.

Pollutants Not Covered By Regulation

By use of BPT as much as 80 percent of the TKN entering biological treatment will pass through, mostly as ammonia. While a proposed means of reducing ammonia has been presented, nonetheless it is clear that TKN pass through can be a serious problem. Many states, in fact, limit effluent discharge on the basis of total oxygen demand (TOD) calculated as $TOD = BOD_5 + 4.5\ TKN$. Calculated in this manner, the total oxygen demand of the final effluent is, for all practical purposes, entirely TKN. TKN and ammonia were proposed for regulation in prior effluent guidelines. In 1979, it was anticipated that extended aeration could be designed to provide nitrification in addition to the removals described above (4). However, no system within the industry to date has demonstrated the capability of nitrifying on a consistent basis. At ERRC we conducted extensive laboratory scale research on extended aeration treatment (11, 12). Pretreated lime-sulfide unhairing waste diluted to 200 ppm TKN served as a model for composite tannery effluent.

In these studies we found that extended aeration could efficiently reduce effluent TKN from 200 ppm to below 10 ppm at 20°C and a sludge residence time of 20 days (the food-to-mass ratio was 0.05). However, the system was destabilized by high salt concentration (2 percent), ferric chloride (less than 500 ppm), or low residual alkalinity (less than 50 ppm). These results provide excellent testimony for the value of substantial equalization to minimize concentration spikes from individual process steps.

Treatment efficiency under low temperature also presented a problem. Figure 6 summarizes the results of an experiment demonstrating the inability to oxidize ammonia at low temperature. In this experiment diluted pretreated lime-sulfide unhairing waste was nitrified in an extended aeration basin at 15°C at a sludge residence time of 40 days until steady state was reached. Thereafter, the temperature of the basin was slowly decreased at approximately 0.5° per week. At the close of the experiment the jacket chiller was turned off and nitrification returned to the initial state. As shown in Figure 6, below approximately 10°C nitrofiers in the activated sludge were no longer able to keep up with the effluent TKN. The results indicate that for winter time treatment stability, enclosed treatment facilities may be required. Further, the results underscore the benefits of in-plant nitrogen control.

COD is a broad spectrum test that provides a useful measure of organic compounds resistant to biological oxidation. As a consequence, in 1981 the EPA proposed COD as an "indicator pollutant." That is, COD would be used as a measure of treatment efficiency, indicating the relative absence or presence of priority (toxic) pollutants.

Based on research conducted at ERRC it appears that although these earlier proposed limitations on COD can be met, the value of COD as an indicator pollutant is rather doubtful. At ERRC we have conducted research to determine the degree of treatment necessary to achieve BAT option II type effluent limitations. In general, we believe that the greatest opportunities for attaining such effluent levels lay in physical/chemical (P/C) treatment of the concentrated waste streams followed by biological treatment of streams not suitable for

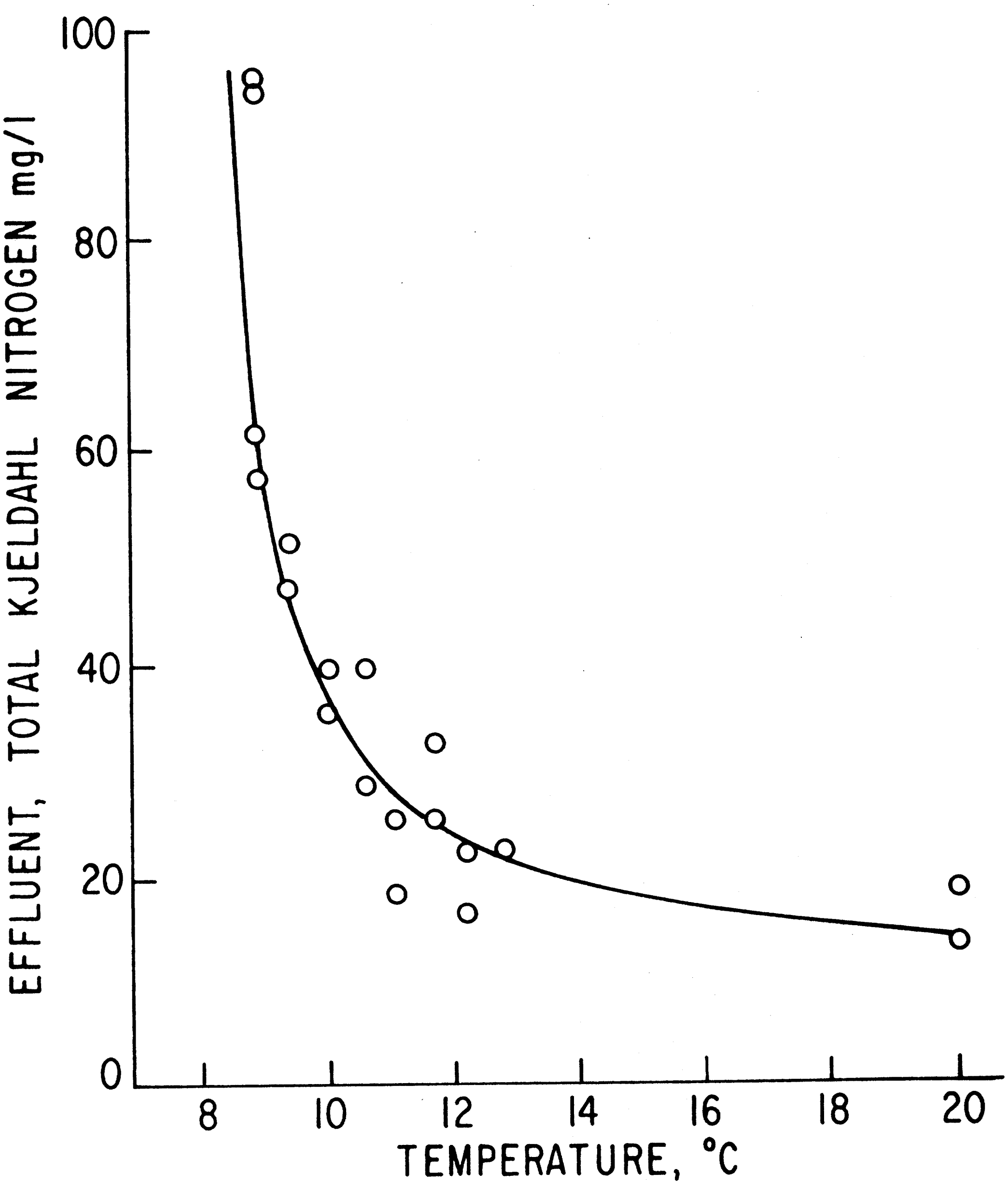

FIGURE 6. — Waste treatment nitrification as a function of temperature.

reuse. In particular, the research focused on P/C treatment of lime-sulfide unhairing waste and extended aeration treatment as a model system for biological treatment. Specifics of the processes are reported eleswhere (2, 8, 9). It was found that nearly 50 percent of a tannery's nonsulfide COD (and 80 percent of the hair protein COD) can be removed by P/C-treatment of this stream alone. If coupled with primary sedimentation of end-of-pipe waste and extended aeration, the resulting effluent should meet the 1979 proposed COD limitations.

Of equal importance to the above findings, we found that in the course of experimentation that over a wide range of P/C-treatment efficiencies and waste concentrations, subsequent extended aeration treatment removed an essentially constant 90 percent of the influent COD. Similarly, a constant (but somewhat larger) residual was obtained in treating essentially the same waste on an RBC (13). These results coupled with the chromatographic data of Barford and Kupec (14) on waste treated by the process of Cooper *et al.* (15), provide compelling evidence of a constant, soluble hair fraction resistant to further biodegradation. Even treatment with activated carbon will not remove such material (13). Consequently, every kg of hair remaining after pretreatment relegates an equivalent amount of recalcitrant chemical oxygen demand to the final effluent stream. This result renders COD ineffective as an "indicator pollutant."

Summary

In contrast to earlier EPA regulations, the present 1982 EPA guidelines for leather tanning and finishing appear to have a firm foundation in demonstrated technology and cost reasonableness. Tannery subcategory 1 BPT and PSES can be met by end-of-pipe treatment. Water use reduction is an essential ingredient in minimizing treatment costs. It is clear however, that this latest round of guidelines does not meet previously espoused long range clean water objectives. In-plant treatment will likely be required to provide a secure environmental future.

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