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Project Summary

Evaluation of Biological Treatment of Pharmaceutical Wastewater with PAC Addition

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A lack of information on applicable removal technologies for total chemical oxygen demand (TCOD) prevented promulgation of best available technology economically achievable (BAT) limitations and new source performance standards (NSPS) for TCOD for pharmaceutical manufacturing plants in 1983 (EPA/440/1-83/084). Therefore, in 1984 U.S. EPA conducted a pilot plant study of activated carbon treatment technologies utilizing pharmaceutical wastewaters from a manufacturing plant that produces fermentation products (Subcategory A) and chemical synthesis products (Subcategory C). One technology that was evaluated was powdered activated carbon (PAC) addition to an activated sludge system (PACT*). A viscous floating mass of mixed liquor solids (VFMLS) developed in the PACT units and resulted in premature termination of the study. Therefore, in 1987 an additional pilot study was performed at the site of the 1984 study. The purposes of this study were to: 1) attempt to find the cause of the formation of the VFMLS, 2) generate additional research data for TCOD removal from pharmaceutical wastewater using the PACT process, 3) evaluate the efficiency of PACT in removing specific organics, 4) evaluate the effectiveness of PACT in reducing efflu-ent aquatic toxicity, and 5) evaluate the use of a selector to improve the settling characteristics of the mixed liquor. One control unit, two PACT units, and a unit equipped with a series of selector basins were operated.

This Project Summary was developed by EPA's Water Engineering Research Laboratory, Cincinnati, OH, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

In November 1982. the EPA proposed effluent limitations and standards for the pharmaceutical manufacturing point source category. The proposed BAT and NSPS limited the discharge of the nonconventional pollutant, TCOD, from existing and new sources. However, due to a lack of information on applicable TCOD removal technologies for subcategories A (fermentation products) and C (chemical synthesis products), limitations and standards for TCOD were not promulgated along with other regulations on October 27, 1983.

By way of responding to these additional information needs, from September to December 1984, EPA conducted biological and physical-chemical pilot plant evaluations of activated carbon treatment technologies utilizing actual pharmaceutical wastewaters. The biological treatment systems consisted of three activated sludge units operated in parallel. PAC

*Registered Patent Process. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

was added to two of the units in selected dosages. The third unit was used as a control. The PAC enhanced biological treatment systems operated during the 1984 test period reduced influent TCOD concentration to levels below those required by the 1976 best practicable control technology currently available (BPT) regulation. This regulation required an average 74% reduction from raw waste levels. PAC also improved the settling rate of the mixed liquor suspended solids (MLSS) when compared to the control unit. A VFMLS developed in the PACT units near the end of the acclimation period for the second set of PAC dosages and resulted in premature termination of the study. The VFMLS did not appear in the control.

Due to the formation of the VFMLS, it was decided that additional information was required before a PACT system could be considered as a viable treatment process for subcategories A and C pharmaceutical wastewaters. Therefore, from March to July 1987 additional biological pilot plant studies were performed at the site of the original 1984 pilot tests. Four biological pilot units were used in the 1987 study. One unit was operated as a control, two units were operated with PAC addition to the aeration basins, and one unit was operated with a series of selector basins ahead of the aeration basin.

The purposes of this study were to: 1) follow up on the previous work and attempt to find the cause of the formation of the VFMLS, 2) generate additional research data for TCOD removal from pharmaceutical wastewater using the PACT process, 3) evaluate the efficiency of PACT in removing specific organics, 4) evaluate the effectiveness of PACT in reducing effluent aquatic toxicity as measured by bioassay tests, and 5) evaluate the use of a selector to improve the settling characteristics of the mixed liquor.

Quality Assurance/Quality Control Methods

Sample analyses were conducted on-site in a field laboratory and in a base laboratory in another state. All samples were collected by the pilot plant operators using prelabeled containers. Analyses for pH, TCOD, soluble chemical oxygen demand (SCOD), nitrogen inhibited biochemical oxygen demand (NIBOD), nitrogen inhibited soluble biochemical oxygen demand (NISBOD), chloride, total suspended solids (TSS), and volatile suspended solids (VSS) were performed at the on-site laboratory. All other analyses were performed at the base laboratory. A portable computer was also located in the laboratory trailer during the study to perform engineering and analytical calculations and to store and transmit data between the field site and home office.

All samples were iced during collection, refrigerated after collection, and iced during shipment to the base laboratory (samples that were analyzed on-site were usually analyzed the same day they were collected but were preserved for reanalysis if necessary). Samples that were sent to the base laboratory for soluble phosphate (SP), nitrate + nitrite ($NO_3 + NO_2$), and ammonia (NH_3) analysis and samples analyzed on-site for TCOD and SCOD were preserved with sulfuric acid addition to pH 2. All samples for soluble parameters were filtered through 0.45 μm filter paper prior to preservation. Samples for chloride analysis were shipped to the home laboratory unpreserved.

TCOD and SCOD analyses were performed by the Hach colorimetric method (*Hach Water Analysis Handbook*, No. 11-01-84-SED, Hach Chemical Company, Loveland, Colorado, 1984). All other analyses for the conventional parameters were performed according to the procedures described in *Methods for Chemical Analysis of Water and Wastes* (EPA/600/4-79-020, U.S. Environmental Protection Agency, Cincinnati, Ohio, 1979) or *Standard Methods for the Examination of Water and Wastewater*, 15th Ed. (1980). At least 20% of all samples were analyzed in replicate for all conventional parameters except NIBOD and NISBOD. One sample each of NIBOD and NISBOD was duplicated each day that analyses were performed. At least 10% of all samples were spiked with appropriate reagents for analysis of all conventional parameters except TSS, VSS, NIBOD, and NISBOD. Blanks and/or standards were analyzed with each analytical run or sample lot as appropriate for the analysis.

QA/QC objectives for precision were to have a maximum of 20% difference between replicates for NIBOD, NISBOD, and VSS. A maximum of 15% difference between replicates was sought for all other conventional parameters. Accuracy objectives were ± 20% spiked sample recovery for NIBOD and NISBOD, and a ± 15% spiked sample recovery for other parameters.

Of 2,131 replicate samples, 2,093 (98.2%) were within the precision percent difference criteria. Of 190 spiked samples, 186 (97.9%) were within the percent recovery objectives. In all, 3,300 analyses were performed, with 3.6% of the analytical results deleted for QA/QC reasons. The project had a 95.8% completion rate for planned analyses.

During the two intensive test periods, samples were analyzed for specific organic compounds, for aquatic toxicity, and by the toxicity characteristic leaching procedure (TCLP) at various contract laboratories. EPA approved methods were used for all analyses.

Pilot Study Procedures

This study consisted of three separate parts: 1) the PACT study to evaluate the PACT process, 2) the selector study to evaluate the use of selector technology for improved mixed liquor settling, and 3) bench tests to try and reproduce the VFMLS.

PACT Study

From March to June 1987, a 101-day study was conducted using one biological pilot plant as the control and two biological pilot plants as the experimental (PACT) systems. The three activated sludge units were operated in parallel.

System Operation--

Raw pharmaceutical wastewater was collected just before it entered the full scale aeration basins and was used as the feed to the pilot units. This flow consisted of equalized pharmaceutical wastewater with a nominal amount (about 2%) of macerated, chlorinated sanitary waste.

The three activated sludge units were identified as Unit 1 (control), Unit 2 (PACT unit), and Unit 3 (PACT unit). Each system consisted of a rectangular aeration tank (189 L) and a conical clarifier (47.3 L). Each aeration tank was operated as a completely mixed basin. Peristaltic pumps were used to supply wastewater and return sludge to the aeration basins. PAC slurry was manually added twice per day to the front end of the aeration basin. A peristaltic pump controlled by a timer wasted sludge from the discharge end of each aeration basin.

The targeted aeration basin hydraulic retention times (HRT's) and solids retention times (SRT's) were 2.2 and 1 days, respectively, for all systems. During the first test period, 198 and 69 mg/L of PAC were added to the influent of Unit 2 and Unit 3, respectively, and

the second test period, 396 and 1,190 mg/L of PAC were added to the influents of Unit 2 and Unit 3, respectively. Daily composite samples were taken from the raw feed wastewater and from the three clarifier effluents using automatic sampling equipment, and from the wasted mixed liquor obtained from the waste solids holding tanks.

Test Phases--

The planned PACT pilot operation was scheduled for a series of five phases as described below:

Phase 1 - Start-up--The three biological pilot plants were operated for 3 days after being seeded with mixed liquor from the full-scale aeration basins. The objective of this phase was to determine whether the effluents for each of the three pilot plants were comparable with respect to SCOD removal.

Phase 2 - Acclimation--Following Phase 1, PAC addition to the influents was initiated for Unit 2 and Unit 3 at rates of 198 mg/L and 698 mg/L, respectively. Single doses of PAC (as a slurry) were added to Unit 2 and Unit 3 each day for the first 4 days to reach the calculated equilibrium PAC concentration, and the operation of all three units was continued for an additional 35 days.

Phase 3 - First Intensive Test Period--Pilot plant operation continued for 14 days with Phase 2 operational conditions. Intensive sampling was initiated that included the collection of aquatic bioassay and organic samples. Four samples for carbon isotherm testing were also collected from the control unit effluent. Daily settling and oxygen uptake tests were performed on each of the pilot plant mixed liquors.

Phase 4 - Second Acclimation Period--This phase lasted 31 days and was used to increase the PAC concentrations in the aeration basins for Unit 2 and Unit 3. The original PAC doses were increased to 396 mg/L and 1,190 mg/L of raw feed, respectively. Over the first four days, additional PAC was added in slurry form to each aeration basin so as to achieve an equilibrium PAC concentration by the ourth day.

Phase 5 - Second Intensive Test Period--Pilot plant operation continued or 14 days with Phase 4 operational conditions. Intensive testing was performed similar to that described previously for the first intensive test period.

Selector Study

The selector pilot unit was operated over the same time period as the PACT systems. Unit 1 was used as the control while Unit 4 was the experimental selector system.

System Operation--

The selector system was identical to the control system with the exception that a series of selector tanks preceded the aeration basin. Initially, the selector system consisted of one 7.6 L (2 gal) anoxic selector tank and two aerated 7.6 L (2 gal) selector tanks operated in series. The return sludge and feed wastewater were mixed in the first anoxic tank and flowed by gravity through the two aerated tanks and into the 189 L (50 gal) aeration tank. The aerobic selectors were equipped with an air supply and metering valves. During the second phase of the selector study, a third 7.6 L (2 gal) aerated selector tank was added. The HRT of each tank was approximately 2 hr based on the raw waste flow.

Test Phases--

Due to the long time required to reach equilibrium, only two test conditions were studied.

Phase I - The selector system was seeded with mixed liquor from the full-scale plant. The system operation with respect to sludge settling was monitored by sludge settling tests. The stirred sludge volume index (SSVI) was calculated for the settling tests. Operation in this mode was continued for 75 days.

Phase II - When the data indicated the mixed liquor for the selector unit was not settling much better than the control unit, a third aerated selector basin, for a total of four selector basins, was added. The system was operated with four basins for the remainder of the study (26 days).

VFMLS Study

It was postulated that the cause of the VFMLS during the 1984 study was due to the presence of a material in the wastewater that was adsorbable and not readily biodegradable. This material might have accumulated on the carbon until it reached a concentration where it began to act like a high molecular weight polyelectrolyte and caused the MLSS and carbon to flocculate and form the VFMLS.

To test this hypothesis, preliminary bench tests were conducted in January at the home laboratory and more tests were conducted on-site during the pilot plant study. The basic procedure was to add spike materials to mixed liquor from the PACT pilot systems (or mixed liquor from a conventional activated sludge system with PAC added for the preliminary tests), aerate the mixture, allow the mixed liquor to settle, and observe the results. This sequence was then repeated once every 24 hr for the on-site tests. A total of eight spike materials were tested at various concentrations.

Feedwater Quality

Table 1 summarizes the raw waste characteristics. The raw waste TCOD was quite variable, ranging from 3,070 to 8,080 mg/L, and it averaged 5,230 mg/L for 64 samples. This average TCOD was significantly less than the average TCOD of the raw wastewater during the 1984 study (7,030 mg/L). Nitrogen and phosphorous were present in adequate amounts for biological treatment based on the effluent concentrations of phosphorous, ammonia, and nitrate + nitrite.

Table 1. *Summary of Raw Wastewater Characteristics*

Parameter	Average	Range
TSS, mg/L	396 (92)*	90-722
TCOD, mg/L	5230 (64)	3070-8080
SCOD, mg/L	3820 (66)	1550-5850
NIBOD, mg/L	2700 (21)	1700-3800
NISBOD, mg/L	2200 (20)	1200-3300
pH, units	-	5.9-12.8 (159)
Temperature, °C	25.5 (112)	18.0-31.0

**Parentheses indicate number of observations.*

PACT Study

The operational and analytical data from the first and second intensive test phases are summarized in Table 2. During Phase 3, the average HRT and average SRT values were very close to the target values and ranged from 2.18 to 2.20 days and 10.0 to 10.1 days, respectively. The average temperature in the aeration basins ranged from 35.0 to 36.5°C while the pH ranged from 6.4 to 8.0. The effluent TCOD concentrations averaged 654, 413, and 344 mg/L for Units 1, 2, and 3, respectively, while the effluent SCOD concentrations averaged 498, 364, and 220 mg/L, respectively.

The average HRT and SRT values ranged from 2.18 to 2.20 days and 10.0 to 10.4 days, respectively, during Phase 5. The pH ranged from 7.3 to 8.1 and the

Table 2. Average Operational Parameters and Effluent Quality During the Intensive Test Periods

	Phase 3 Day 43-56			Phase 5 Day 88-101		
	Unit 1	Unit 2	Unit 3	Unit 1	Unit 2	Unit 3
Operation Data						
Feed Flow, mL/min	60.0	60.3	59.9	59.8	60.2	60.3
PAC Dosage, mg/L Feed	0	198	698	0	396	1190
Return Sludge Flow, mL/min	60.1	63.5	61.2	176	203	59.5
Waste Mixed Liquor Flow, L/day	17.7	18.6	15.6	17.7	18.2	18.7
F/M, g TCOD/day-g MLSS*	0.58	-	-	0.54	-	-
HRT, days	2.19	2.18	2.20	2.20	2.18	2.18
SRT, days	10.0	10.0	10.1	10.4	10.1	10.0
Mixed Liquor Data						
Temp., °C	36.5	35.0	35.0	34.5	35.0	36.0
pH (range)	6.5-8.0	6.4-7.8	6.5-7.7	7.4-8.1	7.3-8.0	7.3-8.0
Dissolved Oxygen, mg/L	2.7	2.4	2.0	2.5	2.2	1.7
TSS, mg/L	4030	5620	8050	4870	7780	11600
SSVI, mL/g**	229	144	58.3	194	107	53.0
Effluent Data						
TCOD, mg/L	654	413	344	532	244	164
SCOD, mg/L	498	364	220	449	210	127
NIBOD, mg/L	11	6	9	13	7	<4
NISBOD, mg/L	6	<4	6	4	<3	<3
TSS, mg/L	77	20	128	39	23	20
VSS, mg/L	63	17	123	34	22	18
Removals						
Average Feed TCOD, mg/L	5030	5030	5030	5690	5690	5690
Average Feed SCOD, mg/L	3480	3480	3480	4210	4210	4210
TCOD Removal, %	86.7	91.6	92.9	90.5	95.6	97.1
SCOD Removal, %	85.5	89.4	93.6	89.1	94.9	96.9

*Food to microorganism ratio.
**Stirred sludge volume index.
-Not determined.

average temperature ranged from 34.5 to 36.0°C in the aeration basins over the Phase 5 test period. Average effluent TCOD concentrations for Phase 5 were 532, 244, and 164 mg/L, for Units 1, 2, and 3, respectively, and average effluent SCOD concentrations were 449, 210, and 127 mg/L, respectively.

The addition of PAC resulted in some improvement in MLSS settling rate as the SSVI data in Table 2 indicates. However, problems with floating sludge in the clarifiers, especially Unit 2, were encountered. The large PAC dosages added to Unit 3 resulted in a consistently good settling sludge.

TCOD and SCOD Removal

The addition of PAC to the activated sludge process improved the removal of both TCOD and SCOD. The comparison of the two intensive test phases was complicated by the different raw wastewater characteristics for the two phases. The influent TCOD and SCOD values were greater for Phase 5 than Phase 3 but the control unit effluent TCOD and SCOD concentrations were less during Phase 5 than Phase 3. Thus, the Phase 3 698 mg/L PACT unit average effluent SCOD concentration (220 mg/L) was slightly greater than the Phase 5 396 mg/L PACT effluent SCOD concentration (210 mg/L). The average SCOD removal percentage of the 396 mg/L PACT unit (94.9%) was also slightly greater than the 698 mg/L PACT unit (93.6%).

Carbon adsorption isotherm tests were performed on four control unit effluent samples from each of the two intensive test phases. All eight of the tests were combined to produce a single isotherm plot. The line of best fit for the data is as follows:

$$\frac{X}{M} = (0.80)C_e^{1.14} \qquad (1)$$

where:

X = amount of SCOD removal attributed to the PAC, mg/L

M = PAC dosage, g/L

C_e = equilibrium effluent SC(concentration, mg/L

The SCOD data from the PA systems can also be presented on Freundlich plot. It was assumed that additional SCOD removal by the PA systems compared to the control wh was operated in parallel w accomplished by the PAC. This allowe calculation of the amount of SC removed per unit of carbon added in SCOD per gram of PAC. The data evaluated with the resulting line of b fit:

$$\frac{X}{M} = (4.7)C_e^{0.85} \qquad (2)$$

Figure 1 compares the isoth derived from the pilot plant results that derived from the jar tests. In case both isotherms are very sim which suggests that isotherms deriv from the jar tests would provide a g prediction of the PAC requirement

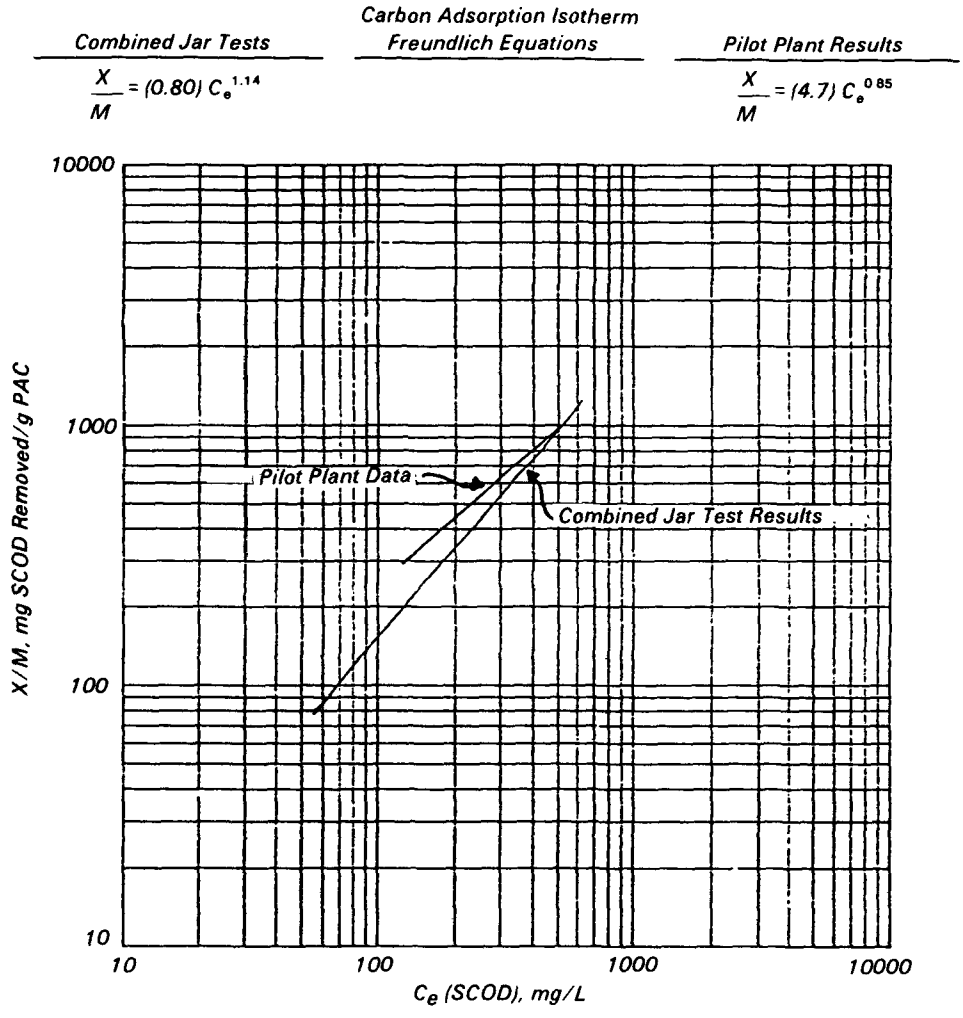

Figure 1. *Comparison of the isotherm derived from the pilot plant results with that derived from the combined jar tests.*

produce any desired effluent SCOD concentration (down to C_e - 127 mg/L). The similarity between the adsorption isotherms also suggests that there was no enhanced biodegradation of refractory compounds in the PACT systems.

Specific Organic Compounds Removal

Based on preliminary samples of raw wastewater and the plant raw product list, eight compounds were selected for quantification using GC/MS techniques for intensive test samples. In addition, computer searches to tentatively identify and quantify all detected peaks were completed. Of the six volatile compounds analyzed for (total xylenes, ethyl benzene, acetone, acrylonitrile, methylene chloride, and ethyl acetate), only two effluent samples and one waste mixed liquor sample contained quantifiable amounts of any of the six compounds. No differences between the control and PACT units with respect to removal of these compounds or concentration of these compounds in the waste sludges were found.

Two semivolatile compounds, alpha-picoline and dicyclohexylamine, were specifically quantified in all samples. Alpha-picoline was found more frequently in Unit 1 (7 of 8) waste mixed liquor samples than Unit 2 (6 of 8) or Unit 3 (4 of 8) and in higher concentrations. However, with the exception of one control sample with 1,600 µg/L and one control sample with 84 µg/L all samples were below 50 µg/L. Alpha-picoline was found in 7 of 8 control unit effluent samples but not in any effluent samples from either PACT unit. Thus alpha-picoline was slightly higher in the control effluent samples as well as the waste mixed liquor samples. All of the control effluent samples contained less than 30 µg/L of alpha-picoline, however.

Dicyclohexylamine was found in 7 of 12 waste mixed liquor samples from the second intensive test period but was not measured in any samples during the first period. The concentrations were somewhat higher (up to 700 µg/L) in the control samples than the PACT samples (up to 150 µg/L). Dicyclohexylamine was only found in one effluent sample (Unit 1 at 110 µg/L).

A wide variety of compounds were identified in the computer searches of the effluent and waste mixed liquor samples. However, there was no apparent relationship between PAC concentration and tentatively identified volatile organic compounds in either the effluents or waste mixed liquors.

The TCLP analysis of the waste mixed liquors determined that none of the samples analyzed would be classified as toxic as defined under proposed U.S EPA hazardous waste identification regulations (40 Code of Federal Regulations 261.24). No organic compounds were detected in any of the samples.

Aquatic Toxicity Removal

Bioassays of the samples from the first intensive test period did not show any significant difference in toxicity removal between the control and PACT units. The average acute (*C. dubia*) toxicity (medium lethal concentration, LC_{50}) of the effluent samples was 46%, 51%, and 47% for Unit 1, Unit 2 and Unit 3, respectively, while the average toxicity of the raw waste was 0.81%. There was also no obvious differences between the chronic toxicity of the various pilot plant effluent samples.

The average LC_{50} of the raw waste for Phase 5 was 1.0%, which was similar to the Phase 3 LC_{50}. The effluent samples from the second intensive test period, however, were more toxic than from the first test period. The average LC_{50} values from Unit 1, Unit 2, and Unit 3 were 14%, 5.8%, and 0.82%. The data for Phase 5 indicate that the toxicity appeared to increase with increasing PAC concentrations. Only limited chronic toxicity data were obtained due to problems experienced with the bioassay test procedure.

Selector Study

The selector unit was operated to determine if improvements in the settling characteristics of the mixed liquor could be achieved. The selector unit mixed liquor showed some improvement in settling characteristics as measured by the SSVI test when compared to the control over the entire Phase I test period. The average SSVI values over Phase I were 222 and 163 mL/g for Unit 1 (control unit) and Unit 4 (selector unit), respectively. The selector unit mixed liquor SSVI, however, was equal to or greater than the control unit values for most of Phase II. During Phase II, Unit 1 and Unit 4 SSVI's averaged 194 and 208 mL/gm, respectively. The most obvious difference between the two phases (besides the additional selector basin added for Phase II) was that the DO concentrations in the selector basins were generally lower during Phase II as compared to Phase I. This observation, along with the presence of *Haliscomerobacter hydrossis* found in the microscopic examination, suggests that the lack of DO in the selector basins was the cause of the poor settleability of the selector mixed liquor. Pure oxygen was used for the last 6 days of operation of Phase II, and the SSVI tests performed during this period indicated some improvement in settling of the mixed liquor.

VFMLS Study

None of the bench tests completed in January, May, or June 1987 resulted in the formation of a VFMLS. Either the individual spike materials tested were not the cause of the original VFMLS or the proper conditions for the formation of the material were not simulated in the bench tests. The task of recreating the VFMLS was made difficult by the fact that the product mix at the plant changes, which results in variable wastewater characteristics. It is possible that a unique pharmaceutical wastewater matrix existed during the 1984 study that did not exist during this study.

Conclusions

The pilot study project conclusions are summarized below:

1. Efforts to identify the cause of the VFMLS that formed during the 1984 study were unsuccessful.
2. Effluent SCOD concentrations were significantly reduced by the addition of PAC to the activated sludge system when compared to the control unit. For the first intensive test phase, effluent SCOD concentrations were reduced by 26.9% and 55.8% compared to the control unit effluent SCOD when adding 198 mg/L and 698 mg/L of PAC, respectively. Effluent SCOD concentrations were 53.2% and 71.7% lower than the control unit value when 396 mg/L and 1,19(mg/L of PAC were added respectively.
3. Isotherms derived from this plant's wastewater treatment plant effluen are a good predictor of the PA(requirements to produce any desire(effluent SCOD concentration (at leas down to C_e = 127 mg/L) by th(following equation:

$$\frac{X}{M} = (0.80)C_e^{1.14}$$

4. PAC addition improved the settlin characteristics of the MLSS However, all three system experienced floating and bulkin sludge during the study.
5. There was no significant difference i the removals of acetone, acrylonitril methylene chloride, ethyl acetat ethyl benzene, total xylenes, alph picoline, and dicyclohexylamin between the control unit and th PACT units.
6. Effluent aquatic toxicity was n reduced by the PACT systems ar In some cases the PACT effluen were apparently more toxic than th control unit effluents as measured t the acute test. The effluents we substantially more toxic for th second intensive test period compared to the first as measur by the acute toxicity test.
7. The study did not show a correlation of COD with aqua toxicity, BOD, or specific identifiat organic compounds in th pharmaceutical wastewater.
8. The TCLP analyses of the cont and two PAC units' mixed liq samples did not identify a contaminants above the regulat limits.
9. The selector unit mixed liquor sett somewhat better than the control u mixed liquor at certain times but exhibited filamentous bulki characteristics. The presence of filamentous organism *Haliscome bacter hydrossis* in one selec mixed liquor sample collected dur the second intensive test per suggested there was a shortage dissolved oxygen in the selec basins. A shortage of dissol oxygen may have contributed to failure of the selector system control the growth of filament organisms during this period of tir

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The complete report consists of two volumes, entitled "Evaluation of Biological Treatment of Pharmaceutical Wastewater with PAC Addition:"
Volume I: (Order No. PB 88-212 527/AS; Cost: $25.95, subject to change)
Volume II: (Order No. PB 88-212 535/AS; Cost $32.95, subject to change
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