

## MICROORGANISMS, FILTERS, AND WATER CLARITY AT TMI-2

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### ABSTRACT

Since the beginning of 1986, hydraulic fluid has been intermittently (inadvertently) added to the Three Mile Island Unit 2 (TMI-2) reactor coolant system. This hydraulic fluid has been a source of food for various biota. The RCS growth rate of these microbial colonies is often phenomenal. Controlling this biota has been relatively easy with the addition of hydrogen peroxide to the water in the reactor vessel. However, the microbial debris coupled with minute particles, clogged the reactor vessel water filters and seriously impaired water clarity. Removing these materials has been a tedious and cumbersome task. Various filter schemes have been used to improve water clarity. Changing RCS conditions make repetition of test results almost impossible. At present, the addition of coagulating agents coupled with a body feed system seems to be the most promising solution to TMI-2's water clarity dilemma.

### INTRODUCTION

In late December 1985 and early January 1986, the water clarity over the TMI-2 core decreased in quality to the point that defueling operations were slowed by the lack of visibility.

The installed filtration - defueling water cleanup system (DWCS) was operated at close to the designed flowrate. The filter-packs clogged with less than 200,000 gallons throughput. The design life of the filters was 6 million gallons throughput minimum.

#### Early Biota Discoveries

During the defueling process, video tapes are made to record core alterations. In late December 1985, verbal reports were made to the Director of Site Operations that an algae-like substance was growing in the core region. Subsequent examinations of the video tapes showed that a finger-like growth was attached to stainless steel surfaces but did not directly influence water clarity. A video tape search was made to determine the quantity and extent of this growth. The annulus region showed floating loose masses of "felt-like" growth as well as the attached algae-like growth to walls and other surfaces.

It was felt that lighting in the core area (plus the heat from the lights) had triggered this growth. Initially, little thought was given to the source of the bacteria or their source of the food. It was also thought that the bacteria were solely responsible for poor filter performance. In other words, the bacteria were sliming the filter surface, effectively reducing the surface area for filtration to nil.

An entry into the DWCS system was made to perform a boroscopic exam of a spent filter cartridge. Parts of the filter surfaces showed nothing (in either particulates or living organisms) while other surfaces showed a preponderance of both.

Attempts were made to collect samples of the microbial growth for identification. The first attempt was to pump and filter a small amount of water from the annulus region and the second was to scrape the surface of the region (suction hose attached to the scraper). Neither method collected enough material for analysis. Attempts at sampling for seeding imitation vessel water were abandoned. The emphasis was quickly shifted from identification to biocides, since if the biota could be destroyed, the need for identification would be unnecessary.

## Microbiocides

The search for a microbiocide was started. The restrictions to this problem-solution were that the microbiocide had to be very effective and it had to be chemically compatible with all phases of defueling and subsequent fuel shipments. In more direct terms, it could not alter the reactor coolant chemistry in terms of pH or halogen content to any noticeable degree plus it had to be compatible with the platinum-based hydrogen-oxygen recombiner catalyst contained in any of the various types of fuel shipping containers. Hydrogen peroxide was chosen to be TMI-2's microbiocide as it had little long range effects on vessel water or fuel shipments.

## Source of Microbes

The first full-scale use of hydrogen peroxide as a microbiocide at TMI-2 was on the spent fuel pool water. Another investigation was undertaken to determine both the extent and the source of the microbes. It was fairly quickly determined that almost all stored water at TMI-2 had the potential for microbial 'blooms' inherent with the water. The conclusion reached was that the source was either the Susquehanna River or airborne dust. In either case, the control had greater importance than the source of spores, germs, etc.

## Microbial Food Source

After the first treatment with hydrogen peroxide, an almost 100% kill was observed. But with time, a resurgence of growth was noticed. The hydraulic fluid used for defueling tool operations was identified as the prime food source for the microbes. This fluid, UCON, was borated and miscible to allow little chemical disturbance of water that came into contact with the spent fuel. It had good temperature resistance as well as high lubricating qualities. However, since the dissolved total organic carbon (TOC) was directly dependent upon the duration and frequency of accidental hydraulic fluid spills in the vessel area, a search was undertaken to replace it. Quintolubric was chosen as it displays many of the same positive attributes as UCON without being as significant a food source.

## Microbial Induced Corrosion (MIC)

Contact with several other utilities was maintained throughout this initial investigative period and MIC became a popular discussion topic. Many items that had been in contact with the RCS waters were examined for MIC as well as video-camera searches for pitting or other types of corruptions were undertaken. Fortunately, no evidence of MIC has been found to date.

## Microbiocide in the RCS

After the proof of principle and effectiveness of 30% electronic grade (EG) hydrogen peroxide had been shown in the fuel pool, further evaluations were done to determine core compatibility. The biggest outstanding question was the "crud burst" effect. In other words, by the addition of hydrogen peroxide, would the activity of the dissolved and/or ionized

radioactive fission products increase, and if so, by how much? The radiological goal was to maintain the defueling area at 5 mrem/hr or less. In July, the first inoculation of the core was accomplished. It was very effective in controlling the microbes and the rise in area dose (i.e., specific activity) was within the established radiological goal. It was also noted that the life of the hydrogen peroxide was short. Heat, light, and radiation all contribute to its quick breakdown.

## Life Extension and Reclamation of Filter Canisters

After many months of microbial R&D programs, filters were being spent rather quickly. Some filters were lasting only a few thousand gallons before being "spent" on high differential pressure (55 psid max.). The total physical loading (from weight perspective) of each filter was too small to detect. At the rate TMI-2 was using filters, one of two things was inevitable - all filters would be used before defueling was completed or we would not be able to see to defuel. In fact, visibility was lost for long periods of time. This impacted the whole defueling schedule.

To avoid this problem, programs were started to recover or reclaim these slightly used filter canisters.

The simplest technique that could be used with minor system modifications was a back-pulse. This did not extend the life of the filters any great extent.

A small reverse-flow scheme was next attempted with similar lack of success.

Flowrates were lowered from design (170 gpm per 500 sq ft) to about 50 gpm per 250 sq ft. This extended the filter life by various unpredictable amounts. Unpredictability became the norm. Analysis upon analysis was performed to try to control or at least correlate changing RCS parameters. These also had little success. Water clarity began to receive more attention, even by the news media.

Another program was started. A chemical/physical flushing of spent filter canisters was attempted. A high-temperature boric acid flush was used to reclaim the filter canisters. This proved fairly successful (after several false starts). However, it must be noted that returning a filter canister to its "original" state for a throughput volume of a few thousand gallons is somewhat less than a booming success. Other methods of canister reclamation were also investigated including insertion of an ultrasonic probe into the filter canister to agitate the solids lodged in the metallic pores. However, this was abandoned due to the physical limitations on the probes.

Below is a graph of the pressure drop vs. volume of a recovered filter, (Fig. 1) and for comparison, a graph of two new (or previously unused) filter canisters (Fig. 2).

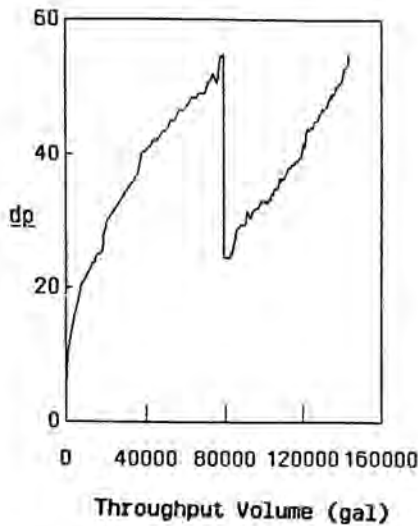


Fig. 1. Performance Test of DWCS Liner F-426 (Reclaimed).

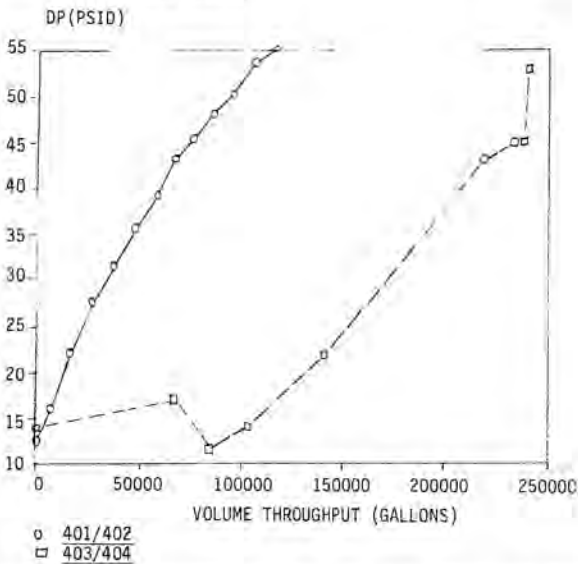


Fig. 2. DWCS Operations Summary for First Two Pair of Canisters.

Laboratory scale tests (called multi-element testing) were also attempted.

**Multi-Element Tests and Results (Chem Cleaning)**

Ten spent element filters (from the DWCS single element precoat and body-feed test) were used for reclamation testing in an effort to determine effective chemical cleaning methods. Each filter had reached 55 psid P at 0.5 gpm.

The test procedure for each filter was:

1. Rinse each element with demineralized water,
2. Soak each element in the solutions described below,
3. Retest each element for pressure (at a flow rate of 0.5 gpm),
4. The solutions and the final pressure after cleaning were:

Solutions	psid
a. 10% HNO <sub>3</sub> at ambient temp	1.0
b. air sparge in methanol	1.0
c. 3% oxalic acid	2.0
d. methanol	4.0
e. 1% Triton X-100	0.5
f. 1% Triton X-100 with ultrasonic cleaner	3.5
g. 6000 ppm boron at 180°F	void*
h. 20% NaOH at 160°F	1.0
i. 0.4 M ammonium oxalate 0.34 M H <sub>2</sub> O <sub>2</sub> 0.16 M citric acid	0.5
j. 0.4 M dibasic ammonium Citrate 4.5 gm/l phenylthiourea 0.4 gm/l EDTA	0.5
k. a new filter (for control)	0.5

\*results indicate a ruptured filter (test attempted twice)

In general, all of the solutions using surface active agents seemed fairly successful in cleaning the filters. Methanol was the least effective. All other solutions were less effective than the surfactant-laden solutions. Using the letter coding from the above list, a ranking can be developed (most to least effective):

- First: solutions e, i, and j
- Second: solutions a, b, and h
- Third: solution c
- Last: solutions d and f

Additional observations: 1) Solution "i" was the most active (foaming and effervescing were observed). 2) The ultrasonic cleaner used with solution "f" was horizontal thus allowing crud to resettle into filter.

These tests are planned to be repeated, as duplication of results ensures the accuracy of scientific experimentation. As RCS parameters are ever changing, to date, duplication of results has been a seldom obtained goal.

**Precoat/Body Feed Testing**

The multi-element test rig was also utilized for precoat and body feed testing. The precoat test portion of the DWCS multi-element testing was recently completed. The results are presented below.

The Fig. 3 shows the differential pressure plotted versus the total throughput volume in gallons for each of the seven conditions listed.

Some general conclusions can be drawn from this figure:

- 1) Celite 503 performed better than Celite 501 (Celite 501 performed poorly for every case in which it was used).
- 2) Heavy coatings of Celite 503 (0.1 lbs/sq. ft) performed a little better than Celite 501.
- 3) The most stable performance was Celite 503 at 0.05 lb/sq ft (the thickest coating).
- 4) In every case shown, little or no flow caused the cessation of the test.

Figure 4 shows the total throughput volume for each test versus the time elapsed in each test. Again, the same general conclusions can be drawn from this figure.

Some cautionary warnings may also be of use.

- 1) The actual difference between the curves for Celite 503 at 0.5 lbs/sq ft and 0.08 lbs/sq ft is negligible due to assumptions made to run the test. However, the 0.05 lbs/ft<sup>2</sup> case is a) easier to apply and b) it is conservative, as a body feed will also be added to filter elements and a lighter initial coating should provide for a longer filter life (less differential pressure to start with after the initial precoat).
- 2) Certain anomalies exist as to effluent turbidities versus the differential pressure (there seems to be no correlation between the two in some cases. As yet, this is not understood).

The body feed test portion of the multi-element testing was completed on July 24, 1986. The results are presented below.

Figure 5 shows the differential pressure plotted versus the total throughput volume for each of the five conditions listed.

Some general conclusions can be drawn from this figure:

- 1) The slopes of all the curves are similar.
- 2) The accelerated precoat test was decidedly the best performer on a throughput volume basis.
- 3) The 5 ppm and the 15 ppm tests do not relate to "successful" tests. That is, the data as collected points to test failure (such as flow bypassing the element).
- 4) Inconsistancies in differential pressure occur somewhat randomly within a small band.
- 5) The 20 ppm body feed test (at the end of the run) is inconsistent with other data.

Figure 6 shows that the slope (rate of throughput volume) was fairly constant for all of the conditions tested. The difference in run time is evident by this figure.

Overall, the accelerated precoat condition was perceived as the best improvement in both portions of this test.

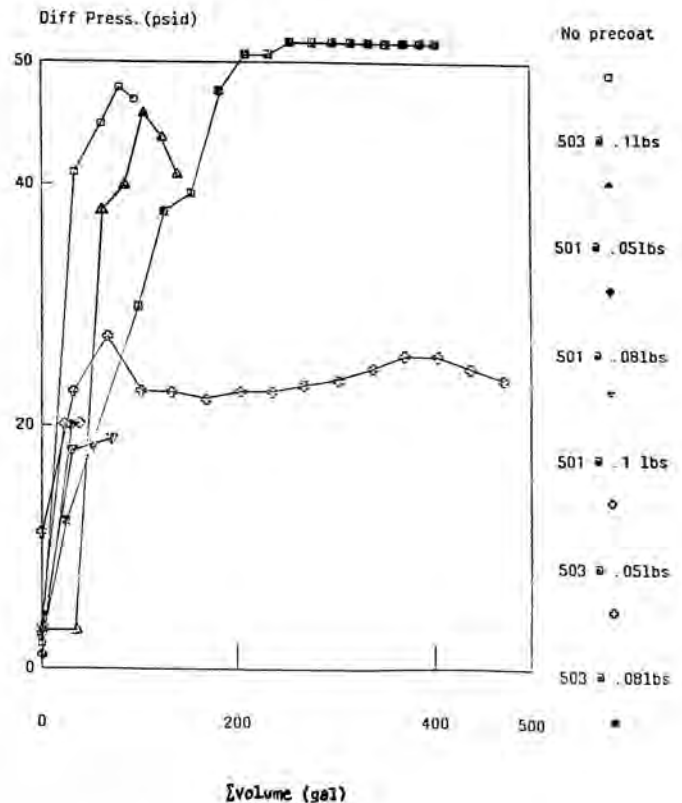


Fig. 3. DWCS Precoat Tests.

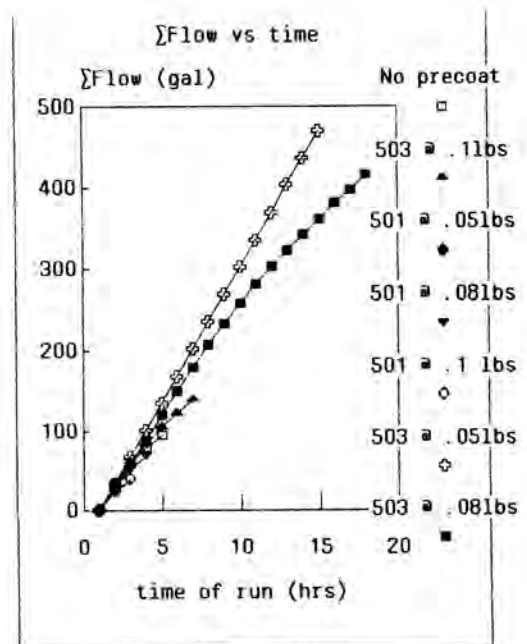


Fig. 4. DWCS Multi-element Test.

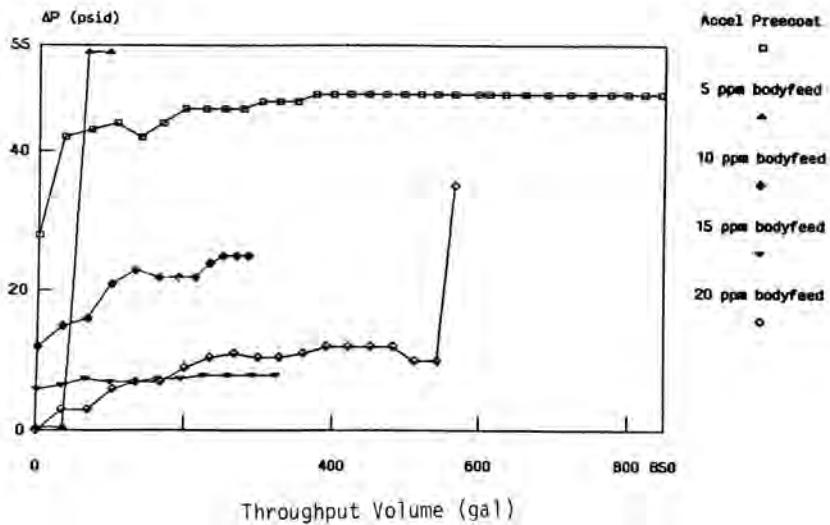


Fig. 5. DWCS Multi-element Tests.

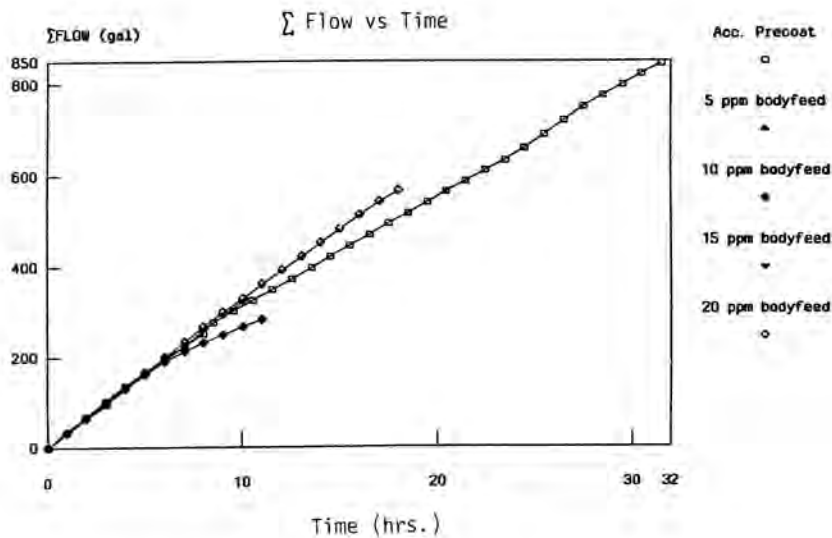


Fig. 6. DWCS Multi-element Test.

#### Total Organic Carbon Removal

As previously mentioned, microbial colonies in combination with hydraulic fluid spills lead to higher than desired total organic carbon (TOC) levels. Two techniques used for TOC removal are; ultraviolet (UV) light coupled with hydrogen peroxide, and UV light coupled with ozone. Fifty gallons of RCS (TOC level 80 ppm) was passed through two ultraviolet lights then seeded with 200 ppm hydrogen peroxide. Samples were drawn every half-hour and tested for H<sub>2</sub>O<sub>2</sub> and TOC levels. After a three volume throughput, no change in TOC level was recorded.

Similar experience occurred with ozone/UV but the use of ozone posed more difficulties than peroxide due to OSHA breathability/exposure limits. Excess ozone was absorbed in potassium iodide solutions.

Charcoal filters were also utilized in an attempt to remove TOC. This technique was shown to be effective (although not to levels below 70-80 ppm TOC). By pumping RCS through charcoal filter beds, TOC levels were reduced from 180 ppm to 80 ppm.

## Total Suspended Solids (TSS) Characterization

The total suspended solids in the RCS as determined by sample analyses has occasionally reached 100 ppm, especially during periods of aggressive defueling. To understand the source of filter media plugging, many samples have been sent to off-site laboratories for analytical characterization. Analytical techniques such as emission spectroscopy, spark source mass spectrometry, x-ray diffraction, cascade filtration followed by scouring and transmission electron spectroscopy with energy dispersive x-ray fluorescence of the filters have been used to characterize the suspended solids in quantity and nature. The results of these analyses have shown three interrelated sources of filter plugging i.e., 1. biological, 2. inorganic, and 3. organic. The biological growth in the RCS caused filters to plug due to their slime-forming character. Inorganic species (Fe, Ni, Si, Zr, U, Ag (In, Cd), and Al) were found in all of the samples and are presumed to be colloidal in nature. The organic introduced by the hydraulic fluids and metabolized by the biota have tended to stabilize the colloids (an organic coating was observed on the particles). The principal sources of the inorganic colloids are presumed to be 1) degraded core materials, Fe, Zr, U, Ag, etc., 2) zeolites used for reactor coolant demineralization, Si, Al, and diatomaceous earth used as a filter aid. The majority of the particulates were determined to be 0.5-1.0  $\mu\text{m}$ , a size optimum for plugging the 0.5  $\mu\text{m}$  porous metal media.

## Image Enhancement

Decreased visibility in the reactor vessel has made it increasingly difficult for the operators to continue their defueling activities for the operators to continue their defueling activities. One alternative to increased visibility is to provide a computerized image enhancement system with a "near real time" image of core conditions to the operators. 2-D and 3-D Image Enhancement Systems are under review using various vendors equipment and existing video tapes of underwater operations in the TMI-2 reactor vessel. Other improvements under review include the use of sophisticated cameras and light sources in combination to provide a more refined image. Other enhancements under review include acoustic and sonar mapping techniques.

## Temporary Filter Systems

**Bag Filter** - Several attempts at filtering were made using bag-type filters. The filter media was much thicker than the installed system. The thickness of the media, in principle, gives this type of filter a greater life due to more sites for material to be trapped. This type of filter acted as a surface-type system. Being surface area related, the filter life was very short and showed little promise for continued testing. Different media were tried and basically, all were ineffective. This concept was quickly rejected.

**Deep Bed Filter** - Sand and diatomaceous earth (DE) were layed in various ways to test deep-bed type filters. Different grades of each medium coupled with different thicknesses of layers (plus different configurations such as SAND-DE-SAND, SAND-DE, DE-SAND, etc) were attempted. Channeling of the fluid through the filters was corrected.

The filter performed similar to a surface filter. This type of filtration was also abandoned.

**Centrifuge** - Another means of separating solids from liquids was by centrifuging the liquor. This technique was extensively tested at TMI-2 with the following results - high area dose rate, clogging of the centrifuge resulting in high maintenance activities, higher than acceptable effluent turbidity and high maintenance worker exposure rates during the cleaning phase. This was also abandoned.

**Swimming Pool-type Filter** - As water clarity was the desired goal, diatomaceous earth precoat-type swimming pool filters were used for RCS filtration. These filters improve water clarity but have only been shown to reduce turbidity to levels that are still unacceptable for TMI-2 defueling operations. Several other problems quickly arose while using this type of filtration. Among these are high-maintenance, fuel-contamination of the diatomaceous earth (DE) resulting in disposal problems, the probable discharge of DE fines back into the vessel resulting in further turbidity problems and waste that is extremely difficult to dewater. This filtration technique is still available for use.

## Full-Scale Precoat/Body Feed

Results of the Multi-Element Precoat/Body Feed Tests indicated that full-scale filter life would be extended with proper precoat/body-feed addition levels. This technique was employed on several canisters. A typical filter life graph is presented below for comparison to the reclaimed and new filter graphs shown previously. Precoat/body feed alone did not extend the filter life.

## Coagulant Testing

Laboratory tests - Small samples of RCS were taken and used for a spectrum of coagulant tests. By using 500 ml aliquots, a choice of coagulant and body feed types could be made. After the types are chosen, the relative concentration of each can be determined for optimal operation by varying one while holding the other constant. This type of testing was fairly extensive as the three graphs below demonstrate.

These results are very dependent on the quality of the RCS. New test programs are required whenever large changes in RCS occur.

Figures 7 and 8 show a typical test sequence.

## Full Scale Implementation

The results of the laboratory experiments were implemented on the water cleanup system filters. A flow diagram of this system is presented below as Fig. 9. Figure 10 is a graph showing the first filter canister performance utilizing this system.

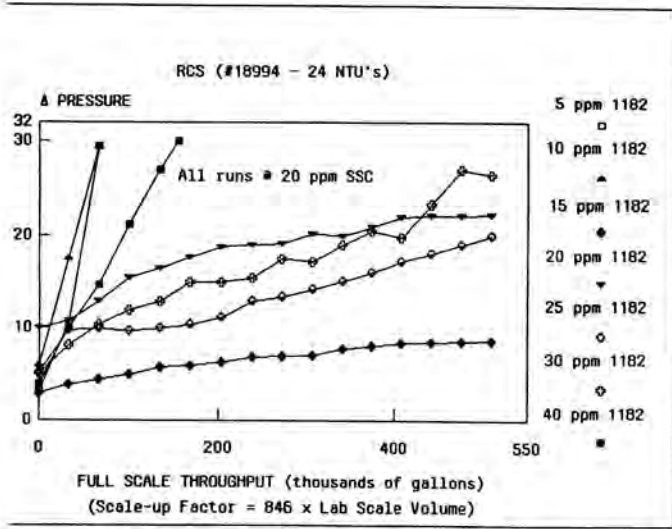


Fig. 7. Coagulant (1182) Comparison Test Series.

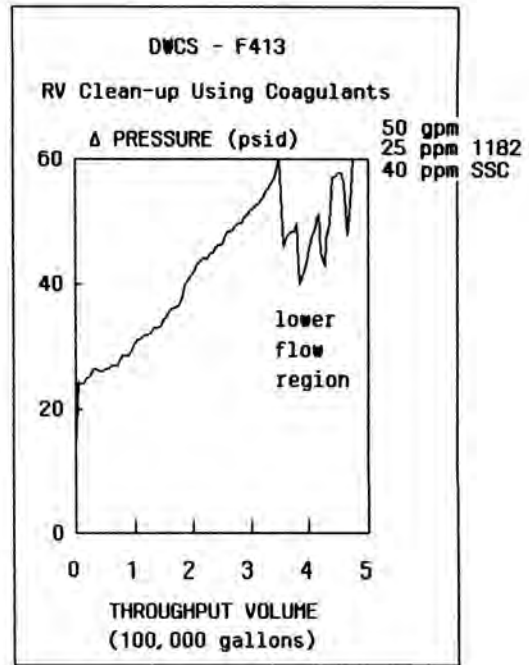


Fig. 10. DWCS-F413 RV Clean-up Using Coagulants.

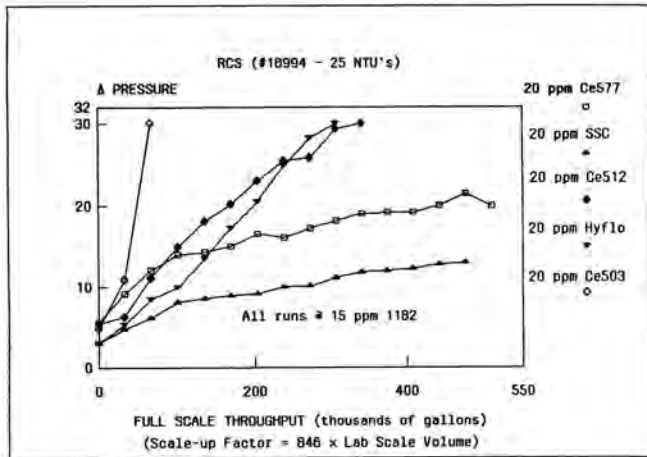


Fig. 8. Body Feed Test Series.

Although the filter life is extended, whether the desired goal of 2 million gallon throughput is achieved has yet to be determined. This type of system is the most promising to date, giving the longest run times and best effluent quality.

To summarize, an overall view of this project is presented as Fig. 11, briefly describing each of the work activities, described by subsection of this paper.

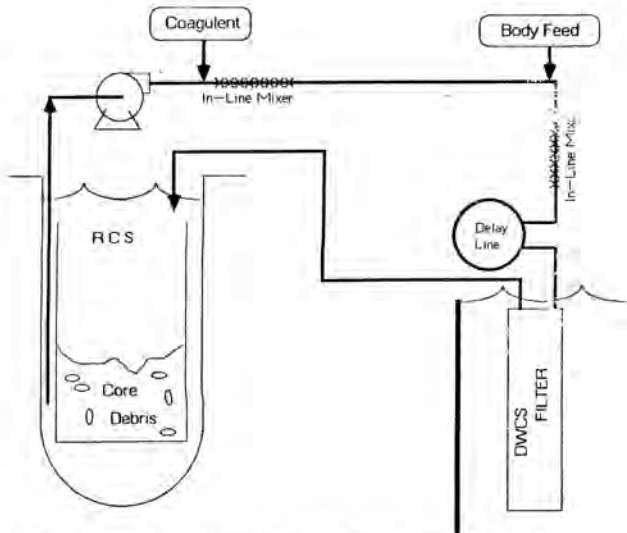


Fig. 9. DWCS Filtration Using Coagulant and Body-Feed.

# TMI-2 WATER CLARITY PROBLEM

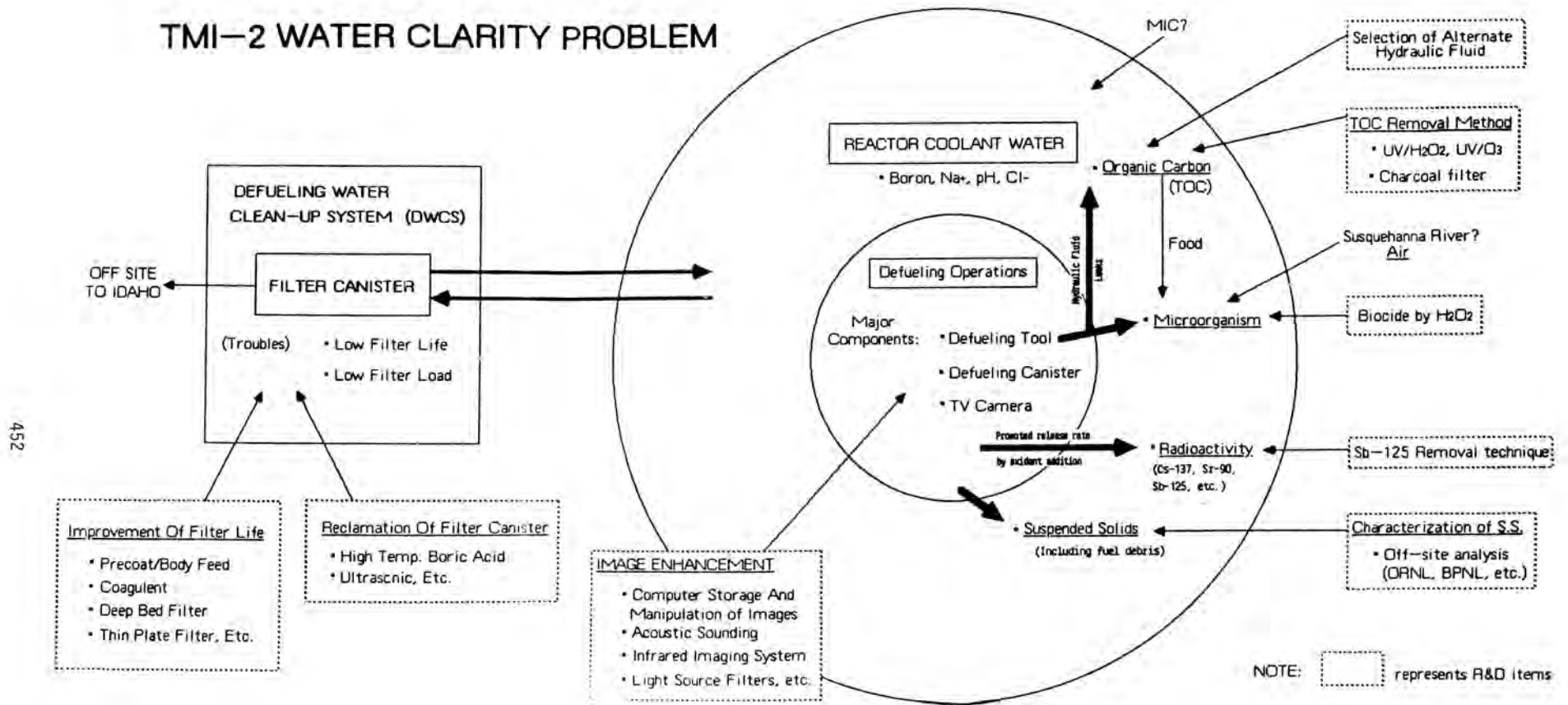


Fig. 11. TMI-2 Water Clarity Problem.