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EVALUATION OF FILTER MEDIA FOR CLARIFICATION OF PARTIALLY DISSOLVED RESIDUES CONTAINING PLUTONIUM - "U"

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INTRODUCTION

A common process in the chemical industry employs the leaching of a desirable component from an insoluble substrate, followed by filtration to produce a clarified solution of the desirable component and a discardable residue. The work described here involved evaluating sintered metal filter media for separating dissolved plutonium from undissolved residues generated at various locations owned by the Department of Energy throughout the United States. The work was performed during a six-week assignment at the Savannah River Laboratory as part of a high school science enrichment program conducted in the summer of 1989.

The leach step used included dissolving the plutonium-containing solids in a solution of nitric-hydrofluoric acid. The undissolved plutonium-containing residues can be removed from a dissolver output slurry for discard or recycle using the sintered metal filter. In the proper application, the sintered metal filter offers the size and low maintenance desired for the types of glovebox processing often used at the Savannah River Site. A glovebox is an enclosed containment box in which all operations are performed through gloved port holes.

To simulate the partial solubility of the actual plutonium-containing residues, a non-radioactive power plant flyash was used. Lab scale tests have shown this surrogate ash to have a chemical composition similar to that of the

actual ash except for the plutonium. Compared to the actual ash, experiments revealed that the surrogate dissolves in the leach solution to similar percentages and also forms the same post-precipitate.

Therefore, results obtained with this simulant are believed to be valid indicators of filtration performance expected with the actual plutonium processing residues. They have been incorporated into the design and test plan for an engineering scale demonstration unit now in operation at Savannah River.

BACKGROUND

During the operation of a plutonium processing plant, some materials unavoidably come into direct contact with plutonium. These materials, such as rubber gloves, air filters, tapes, and equipment cleaning pads, are then said to be contaminated. The plutonium must be reclaimed from the materials and they must be disposed of in a way that is both environmentally safe and economically acceptable. A major step in a process which accomplishes this is the incineration of the materials to reduce their bulk volume.

Incineration converts the material to a fine granular or powdery material, much of it insoluble. The plutonium in the material is converted to what is called a high-fired oxide, which is difficult to dissolve. As a result, the challenge is to develop a process which dissolves the plutonium oxide into solution away from the remaining material, and separate that solution from the solid residue.

Over the past 30 years, the sites owned by the Department of Energy have built up a backlog of scraps and residues for processing. In view of a growing recognition of the the need to eliminate the backlog, converting the scraps to a discardable form has become an important mission at Savannah River and other Department of Energy facilities.

One technology used for processing the incinerator ash employs a hot nitric-hydrofluoric acid solution to leach the plutonium from the solid residue. When the ash is cooked in this acid mixture, the plutonium goes into solution so it can be recovered by other process steps.

Only about half of the plutonium dissolves during the first pass, compared with a goal of complete dissolution. Therefore, the filtered undissolved solids, or heels, must be dried and reprocessed. This is further complicated by the formation of a gelatinous post-precipitate in the solution. Because of its gelatinous nature, the silica-hydrate creates many filtration problems.

The sintered metal filter and a centrifuge were both considered as the means for separation. At this stage, the centrifuge has been considered inappropriate for glovebox use. Its large size and high rotational speeds make it undesirable for the cramped space of a glovebox. Although initial tests with a lab centrifuge are promising, the sintered metal filter was selected as the method of choice.

The filter is constructed from porous stainless steel. As a slurry passes through the filter, solids build up on

one side of the filter. When the solids significantly restrict filtration, the inlet and outlet streams are closed. Pressurized air is applied to the downstream side of the filter, backwashing the filtered solids from the filter. The backwash regenerates the filter for additional filtration.

When used with a compatible slurry, the metal filter can be regenerated 30 to 100 times (in the lab, 3 or 10 runs are needed for a successful test because of the weak backpulse available in the lab relative to the plant). This saves plant operators 30 to 100 manual filter changes when compared to filters which can only be used once. As a result, employing a sintered metal filter can considerably reduce overall radiation exposure to operators.

Under incompatible filtering conditions, solids get embedded in the pores and are unable to be backwashed. As a result, the effective filtering surface decreases rapidly across multiple runs and the filter must be replaced after two or three backwashes. This offsets the filter's primary advantage.

The rate that a slurry filters is directly proportional to the pressure drop generated by the method used for slurry transfer (such as pumping or vacuum transfer). Pressure difference between two points represents the driving force for moving a slurry from one point to the other. Resistance between the two points impedes the transfer. During filtration, the types of resistance evolve from the filter,

cake and process lines; in most cases, the filter resistance is assumed constant and line resistance negligible (Figure 1).

Therefore, the loss of filtration rate with time arises from increases in cake resistance as solids build up on the filter. If the solids are hard, they can form their own porous filtering matrix and cake resistance will build up slowly. For deformable solids, the filtering matrix is less pronounced and cake resistance builds up rapidly to stifle filtration.

The primary methods for transferring the slurry include pumping and vacuum transfer. Vacuum transfer is the preferred method because it pulls the liquid toward the target vessel. A pump pushes the liquid from behind. If break in a line occurred, the vacuum would limit or prevent leakage by drawing air into the system while the pump would augment the failure, allowing contamination of the surroundings. In addition, a vacuum can be generated by vacuum jets which have no moving parts (Figure 2). This reduces the level of maintenance inherent in a system using pumps instead.

The disadvantage to using a vacuum is the limited amount of driving force available for transfer. In an ideal case, the target tank can be evacuated to an absolute vacuum, or zero psi of pressure. Since the feed tank is at atmospheric pressure, or 14.7 psi, the maximum available driving force is 14.7 psi. Most common pumps generate 30 to

60 psi. This means that both the amount of filtration possible through the filter and rate at which it occurs are significantly higher when using a pump instead of vacuum transfer (Figure 3).

DISCUSSION OF EXPERIMENT

The first set of experiments aimed at 1) determining if the sintered metal filter held any promise for filtering the dissolver slurry and at 2) finding the filter pore size which performs best. These experiments were performed using small-scale filters with one-eighth the surface area of the full-scale filters. The second phase of this experiment will include full-scale testing to measure the release of the filter cake from the filter during the backwash step and to determine if vacuum transfer provides enough driving force to adequately transfer the slurry.

The slurry used is generated by dissolving a surrogate flyash in boiling nitric-hydrofluoric acid and precipitating silica hydrates with aluminum nitrate. The slurry results from 40 to 60 percent undissolved solids.

All filtration work was performed on the setup presented in Figure 4. The arrangement allows a vacuum to be pulled on the filter. Filtrate collects in the graduated cylinder below the filter. Filtrate volume is measured against time to determine the filtration rate. The pressure gauge permits pressure drop readings across the filter when [illegible] present upstream of the filter.

Tests were conducted on many filters including sintered metal filters rated at 40, 20, 10, 5, 2, 0.5 and 0.2 microns, and a Teflon fiber filter rated at 1.0 micron. Filtration runs were performed in following manner:

Runs 1 - 3
18-Hour Break
Runs 4 - 6
2-Hour Break
Runs 7 - 9

For each filter, the tests measured resistance to filtration and filter regeneration. We also examined the influence of slurry temperature on filtration.

RESULTS

Measuring the pressure drop across virgin filters yielded interesting results. Low pressure drops were experienced for the 40-, 20-, 10- and 5-micron filters with a substantial increase occurring for the other filters (Figure 5). As a result, the filters below 5 microns present a large initial resistance to filtration leading to a lower initial driving force. This translates into lower sustained filtration rates.

We conducted filtration tests on the 0.2, 0.5, 2.0, 10, 20 and 40 micron filters and the Teflon fiber filter using slurries cooled to room temperature. The 2-micron filter showed a good initial flow rate, but the flow rate decayed gradually across all runs (Figure 6). The 10-micron filter yielded comparable results (Figure 7). In both cases, the 18-hour break did not affect filtration. The 20- and 40-micron filters exhibited characteristics different from the

2- and 10-micron filters. With the 20- and 40-micron sizes, the 18-hour break clearly altered filtration. In both cases there was a significant increase in filtration rate after the break. After the break, the filtration rates decayed similarly (Figures 8 and 9).

Looking at the data collectively, the 2-, 10- and 40-micron filters all demonstrated similar filtration rates for the first three runs while the 20-micron rate was notably higher (Figure 10). After the 18-hour break, the 20 and 40 rates increased to the same level and displayed the similar filtering characteristics and filtration rate decay Figure 11). Under the same conditions, the 2- and 10-micron filters experienced a gradual filtration rate decay typical of sintered metal filters and were unaffected by the break (Figure 12).

The 0.2 and 0.5 micron filters did not completely filter the first run. The Teflon filter performed poorly across two runs and experienced a large loss of filtration rate between runs (Figure 13). These results may be attributable to the filter resistance depicted in Figure 5.

We ran the same test on a 20-micron filter using a hot slurry. The filter performed poorly and the backpulse was basically unsuccessful (Figure 14). It is suspected that in heated solutions the gelatinous precipitate is pliable and readily conforms to the shape of the filter's pores. Subsequently, no filtering matrix is established for filtration through solids built up on the filter.

CONCLUSIONS

The 20- and 40-micron undergo a conditioning effect during the 18-hour break. Because of the larger pore size, particles occupy pore sites and dry. In doing this, the particles alter the filter pore size. With the 2- and 10-micron filters the slurry particles are too large to become embedded in the pore leaving the filter virtually unchanged. This conditioning effect is further supported by the nature of filtration rate improvement for the 20- and 40-micron filters. Although both filters experienced different initial flow rates, they both exhibited the same filtering characteristics after the conditioning.

Since the 20-micron filter displayed a noticeably larger initial filtration rate and a lower level of improvement than the 40 (which was not expected), this suggests that an ideal filter pore size exists which will combine the highest initial filtration rate with no conditioning effects. It is also expected that this pore size will be approximately that of the average particle size. Because of this, a slurry sample was submitted for pore size distribution tests. Lab results confirmed an average particle size of about 15.9 microns (Table 1).

With an average particle size of about 15.9 microns, the 10-micron filter presents itself as the most effective filter for separating a cooled solution of flyash dissolved in nitric-hydrofluoric acid. The filter is not an acceptable method for filtering a hot slurry. The Teflon

fiber filter cover may be a useful alternative at greater pore sizes, but it shows little promise at a pore size of 1 micron. From this point, extensive tests can begin with a full-scale 30-micron filter.

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Filtration rate (u) is directly proportional to the pressure drop and inversely proportional to fluid viscosity and the hydraulic resistance of the cake and filter medium.

The initial filter resistance is often assumed to be a constant Filter cake resistance is the resistance to filtrate flow per unit area of filtration; filter cake resistance increases with increasing cake thickness.

$$u = \frac{\Delta P}{\mu (R_c + R_f)}$$

μ = VISCOSITY
R_c = FILTER CAKE RESISTANCE
R_f = INITIAL FILTER RESISTANCE
ΔP = PRESSURE DROP

Figure 1

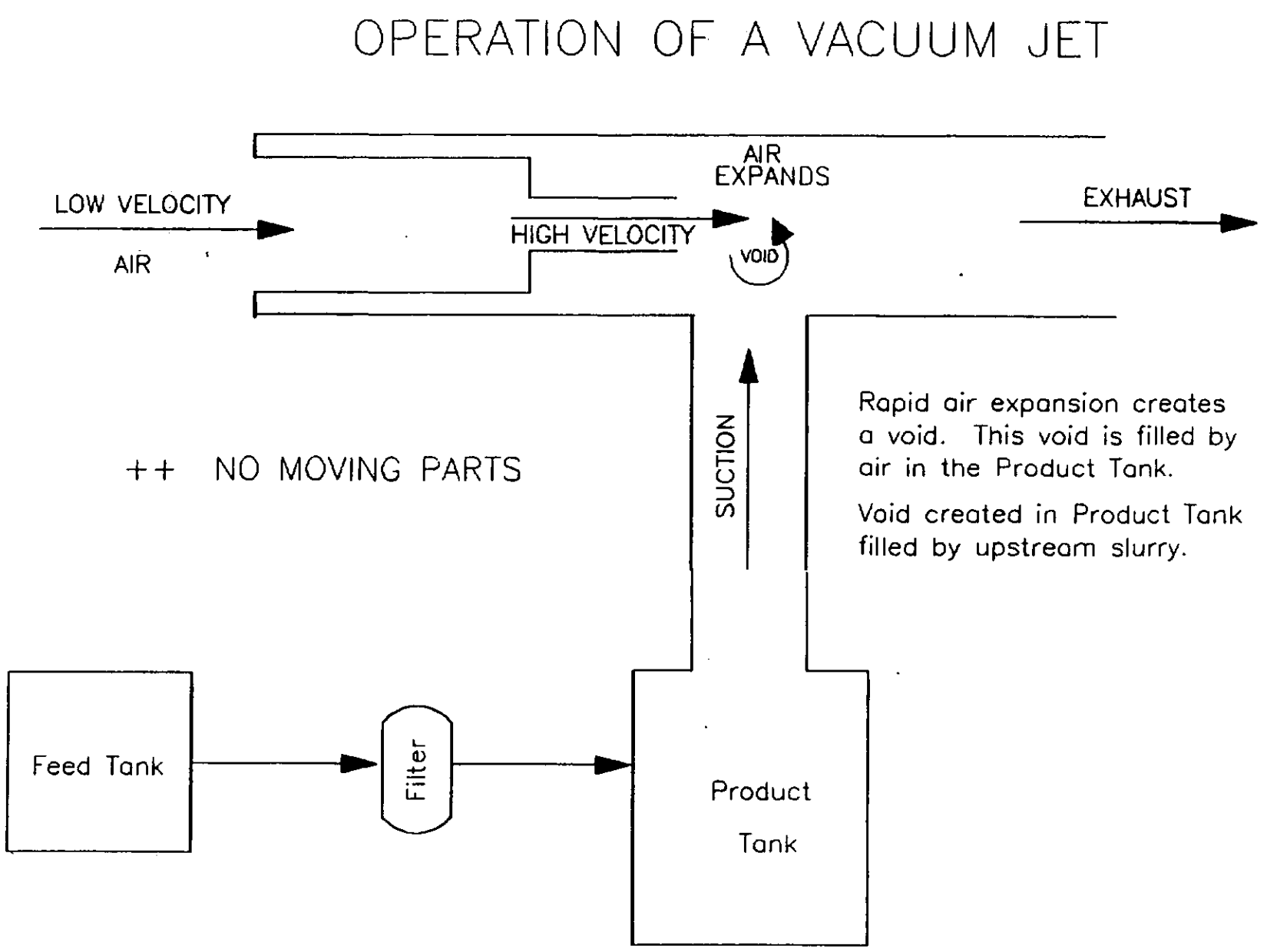
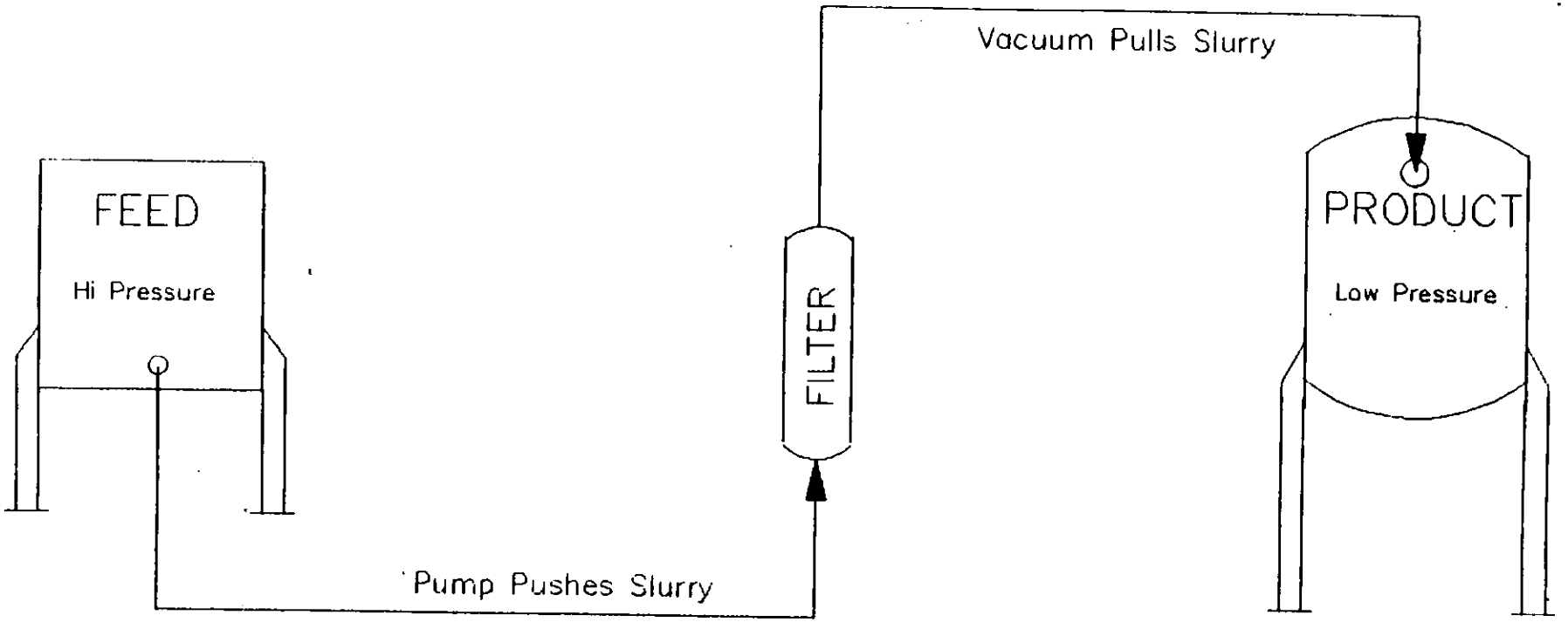

OPERATION OF A VACUUM JET
LOW VELOCITY
AIR
AIR
EXPANDS
HIGH VELOCITY
VOID
EXHAUST
++ NO MOVING PARTS
SUCTION
Rapid air expansion creates a void. This void is filled by air in the Product Tank.
Void created in Product Tank filled by upstream slurry.
Feed Tank
Filter
Product
Tank

Figure 2



* PRESSURE DIFFERENCE IS DRIVING FORCE FOR TRANSFER

DRIVING FORCE = INITIAL PRESSURE DIFFERENCE –

PRESSURE DROP (FILTER + SOLIDS + PROCESS LINES)

VACUUM DRIVING FORCE LIMITED TO 30" Hg (14.7 psi)

(Actual Plant Process About 20" Hg)

PUMP DRIVING FORCES CAN EXCEED 100 psi

Figure 3

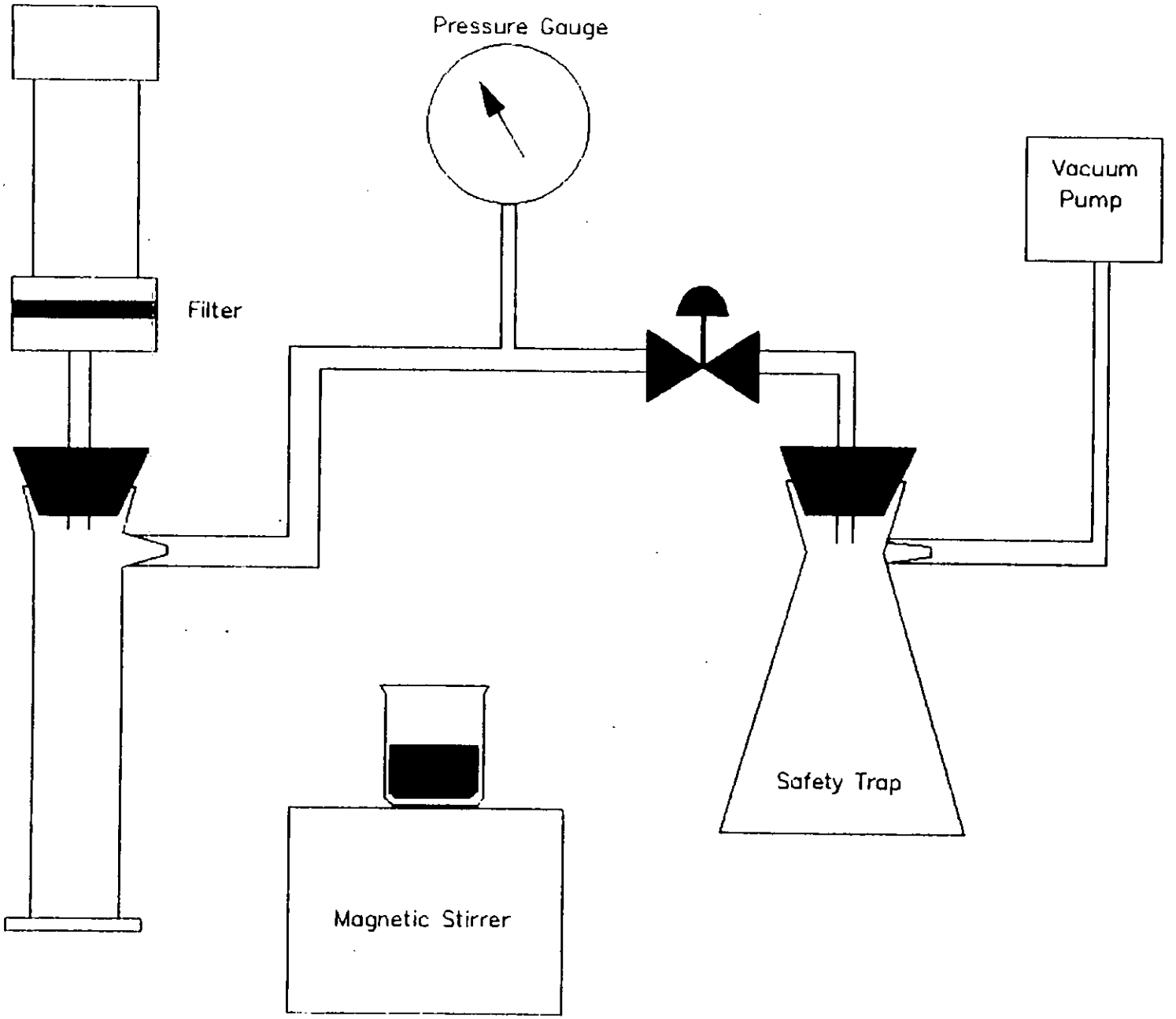

Pressure Gauge
Vacuum Pump
Filter
Safety Trap
Magnetic Stirrer

Figure 4

PRESSURE DROP ACROSS FILTER
(Units of inches Hg)

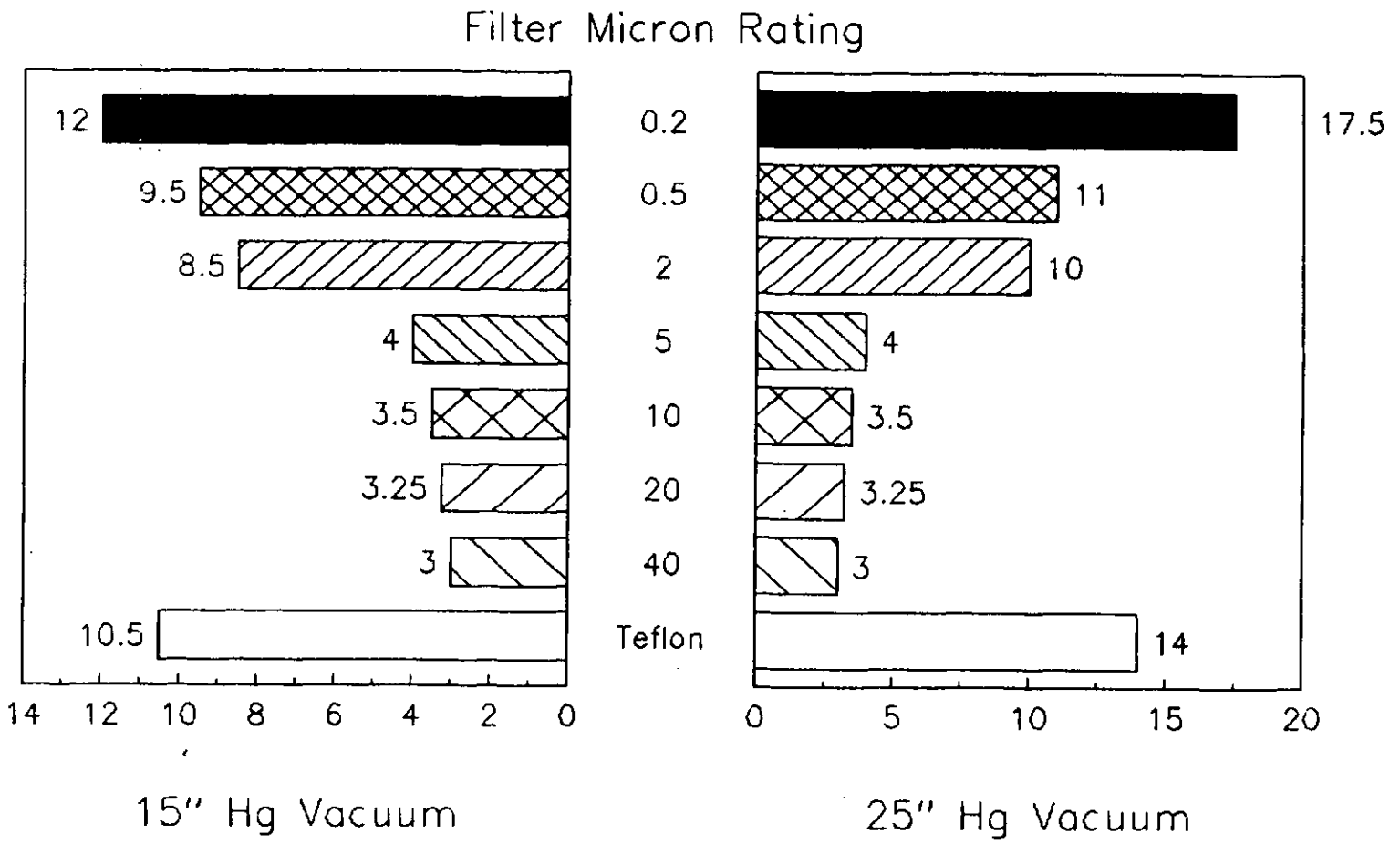


Figure 5

FILTRATE VOLUME vs. TIME

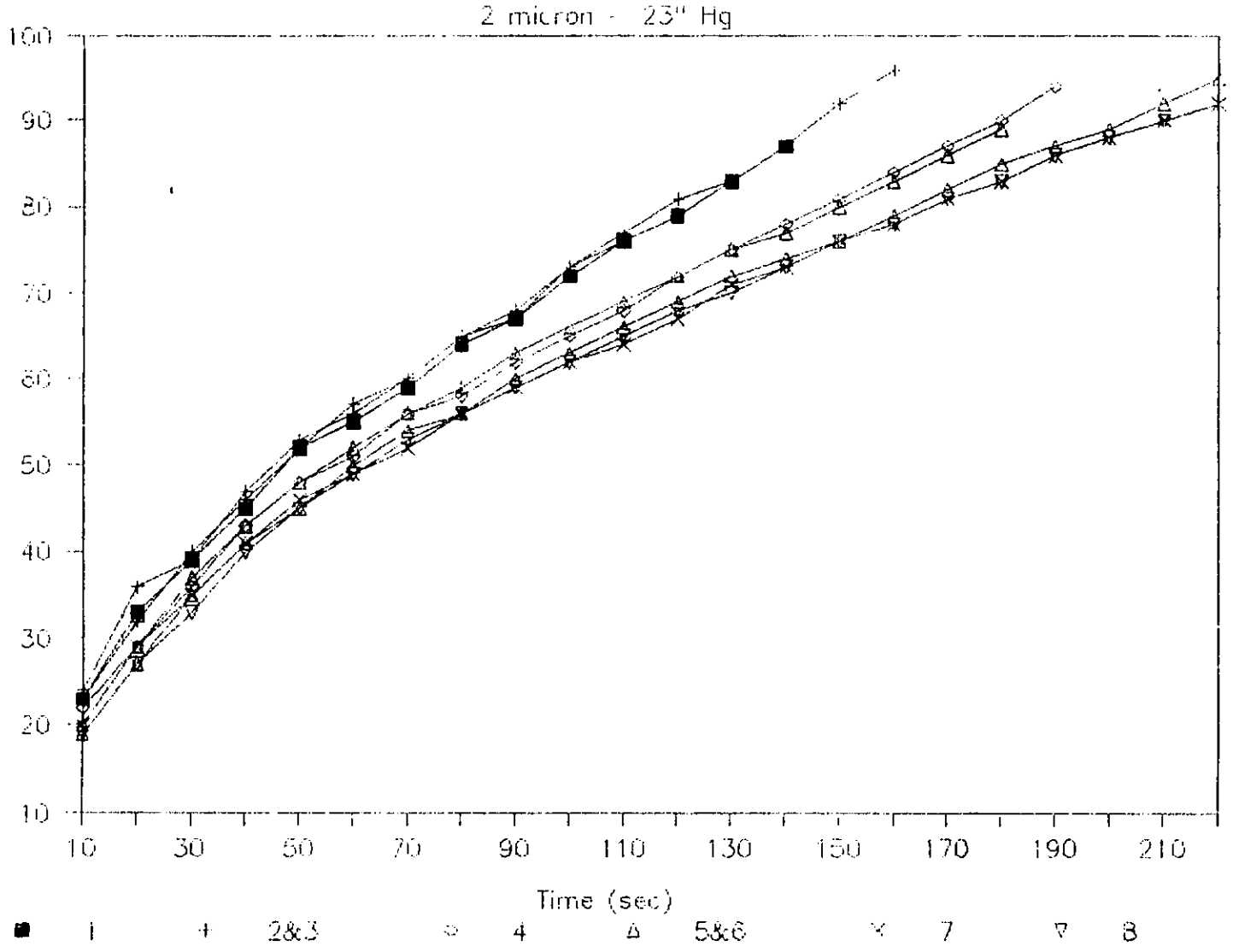


Figure 6

FILTRATE VOLUME vs. TIME

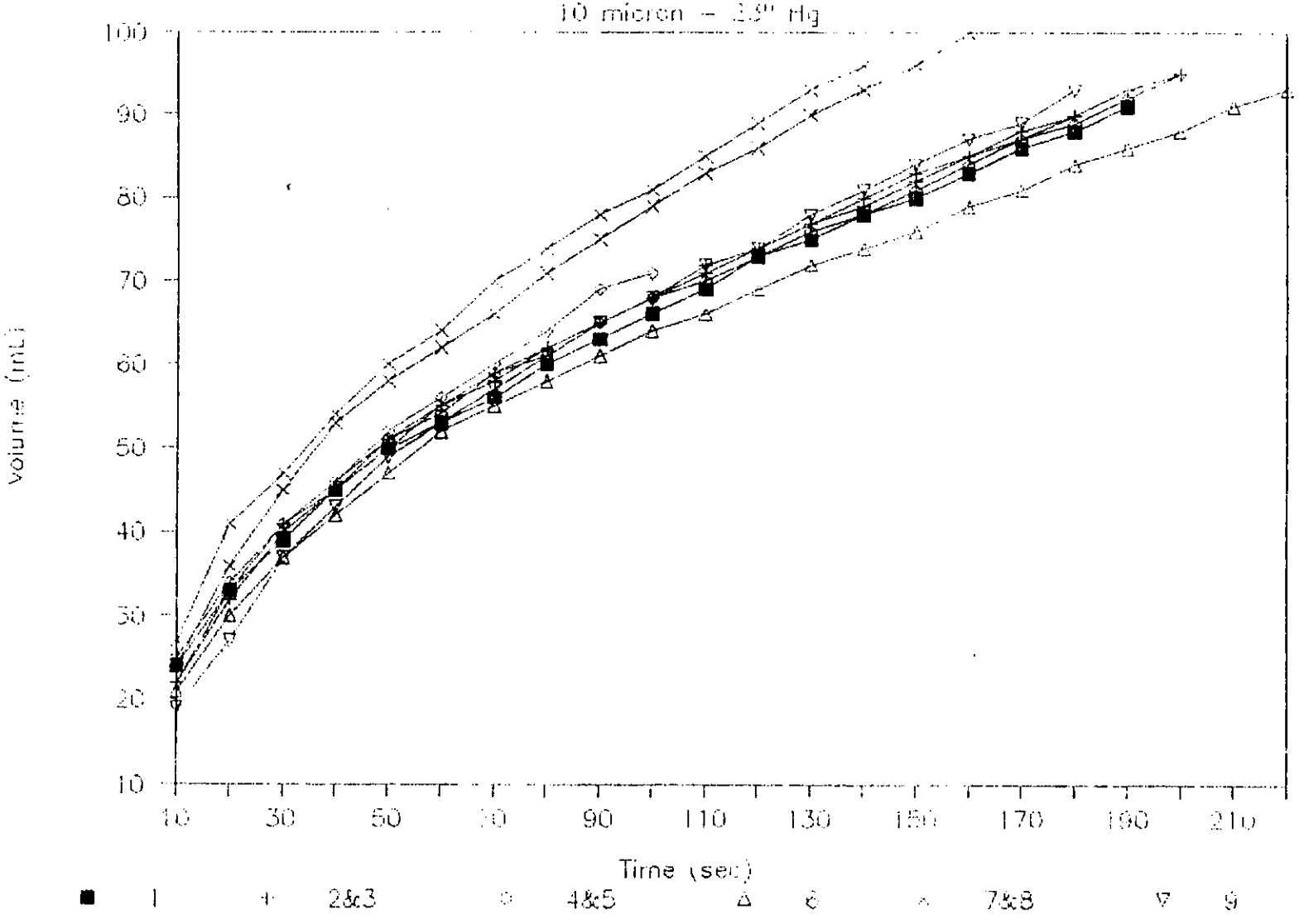


Figure 7

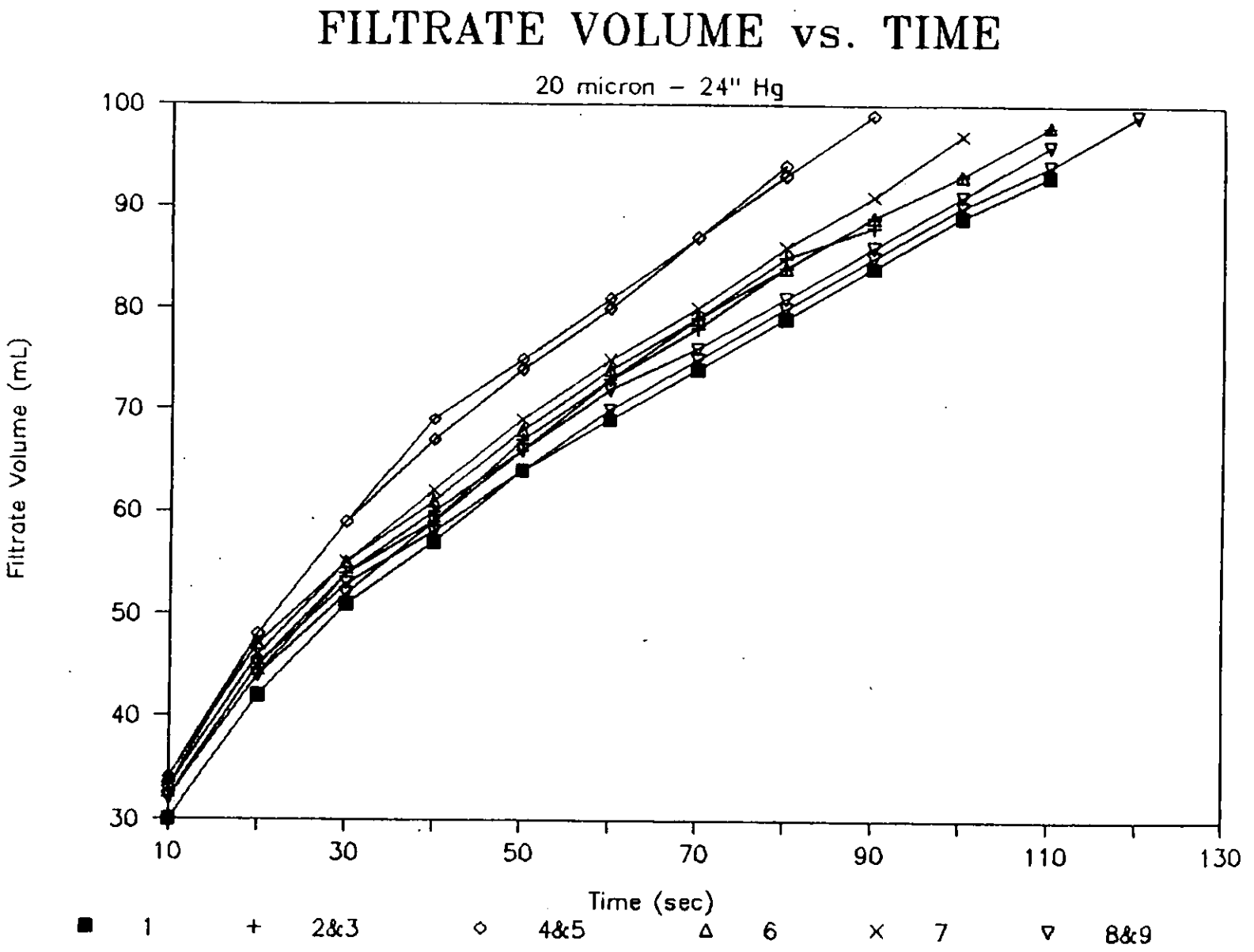

FILTRATE VOLUME vs. TIME
20 micron – 24" Hg
Filtrate Volume (mL)
100
90
80
70
60
50
40
30
10
30
50
70
90
110
130
Time (sec)
1
2&3
4&5
6
7
8&9

Figure 8

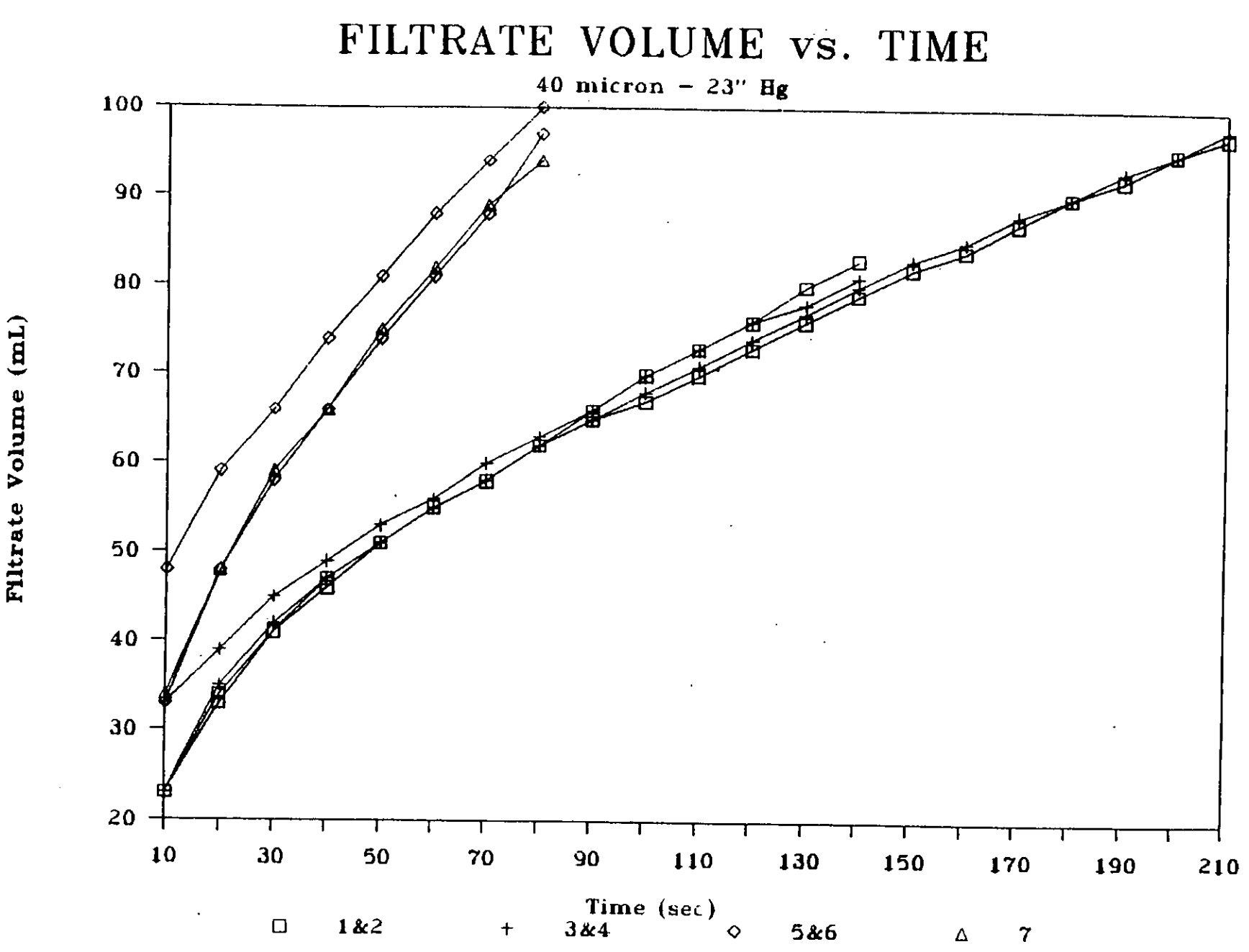

FILTRATE VOLUME vs. TIME
40 micron – 23" Hg
Filtrate Volume (mL)
100
90
80
70
60
50
40
30
20
10
30
50
70
90
110
130
150
170
190
210
Time (sec)
1&2
3&4
5&6
7

Figure 9

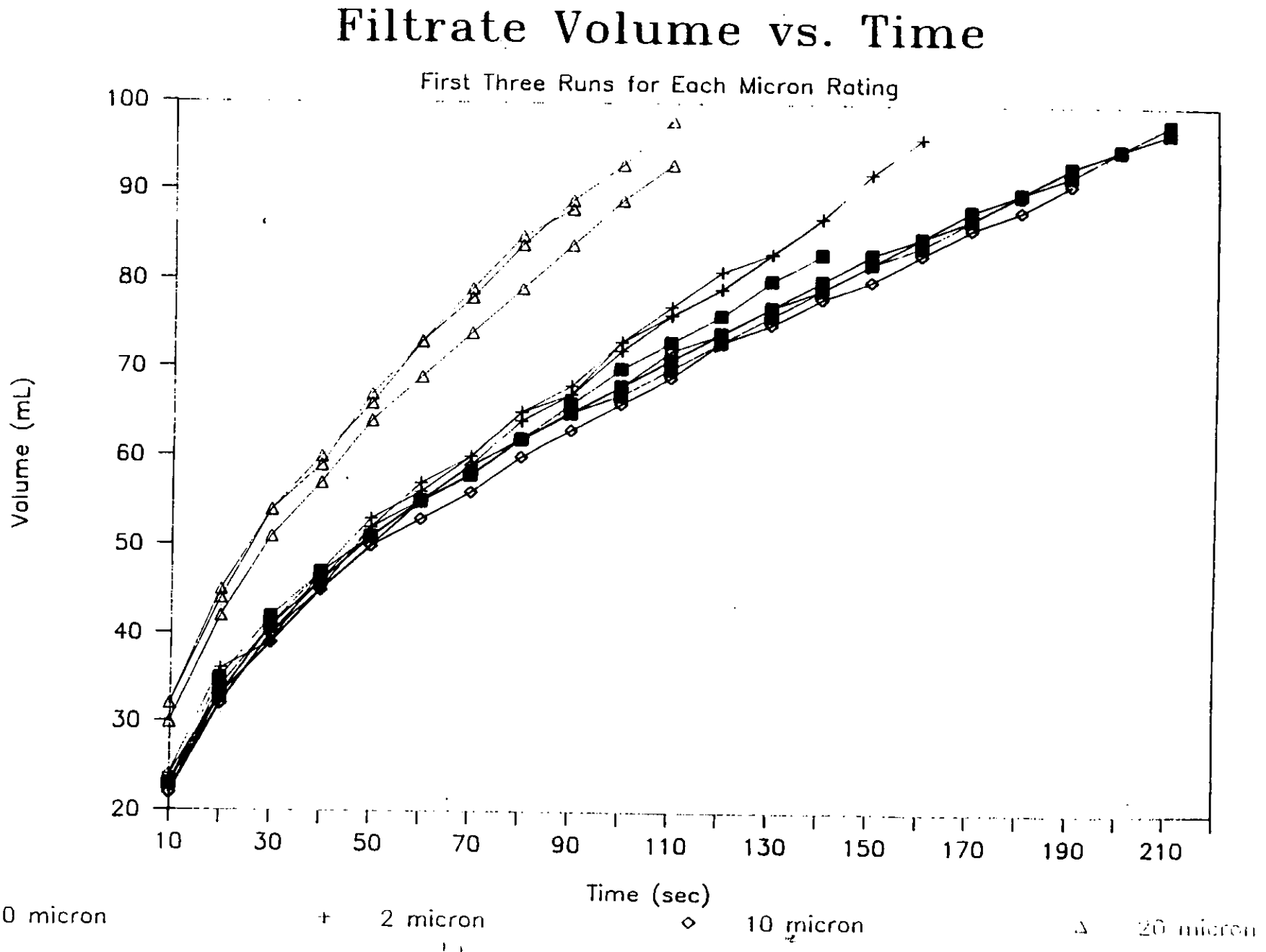

Filtrate Volume vs. Time
First Three Runs for Each Micron Rating
Volume (mL)
100
90
80
70
60
50
40
30
20
10
30
50
70
90
110
130
150
170
190
210
Time (sec)
40 micron
2 micron
10 micron
20 micron

Figure 10

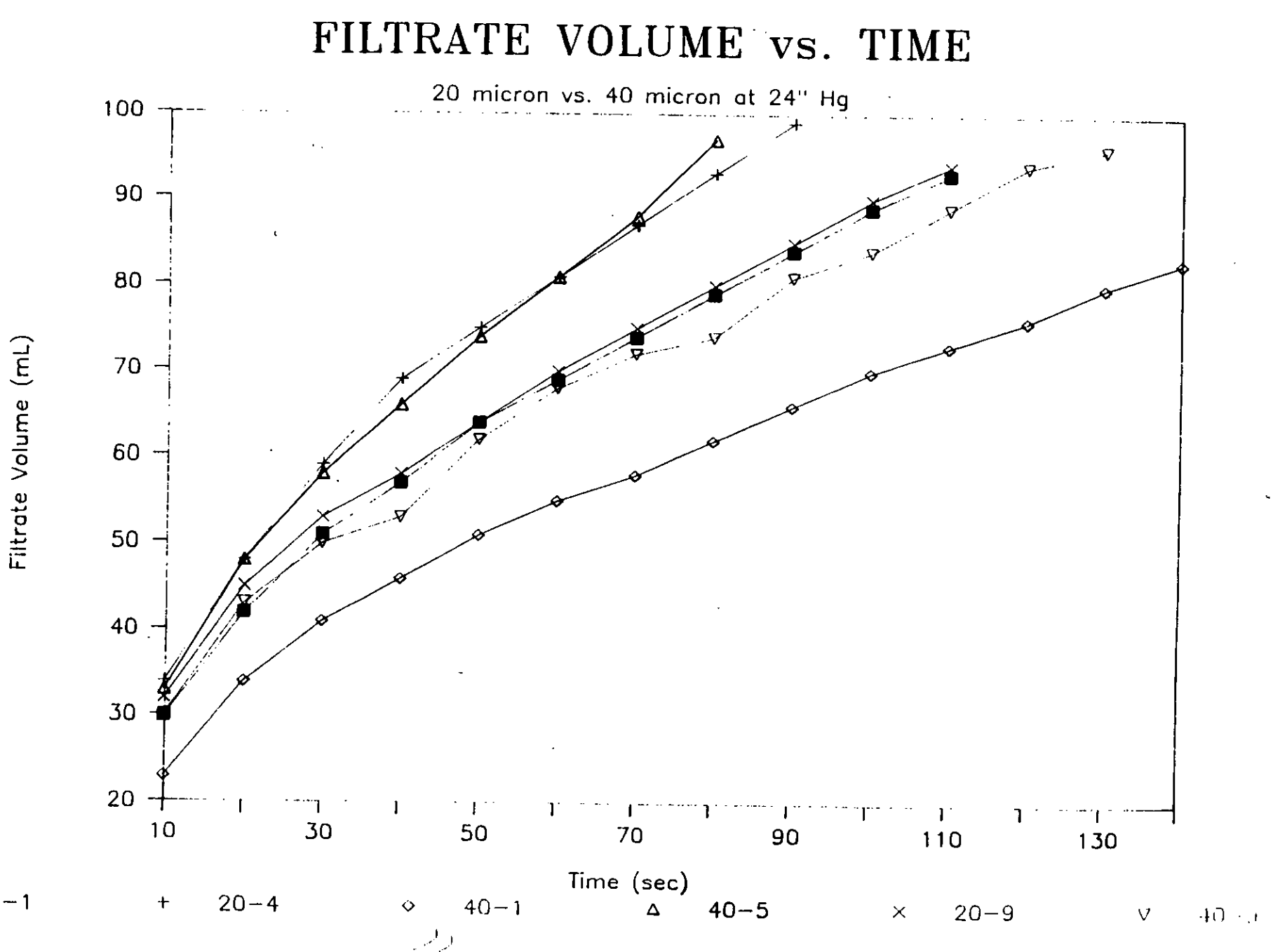

FILTRATE VOLUME vs. TIME
20 micron vs. 40 micron at 24" Hg
Filtrate Volume (mL)
100
90
80
70
60
50
40
30
20
10
30
50
70
90
110
130
Time (sec)
20-1
20-4
40-1
40-5
20-9

Figure 11

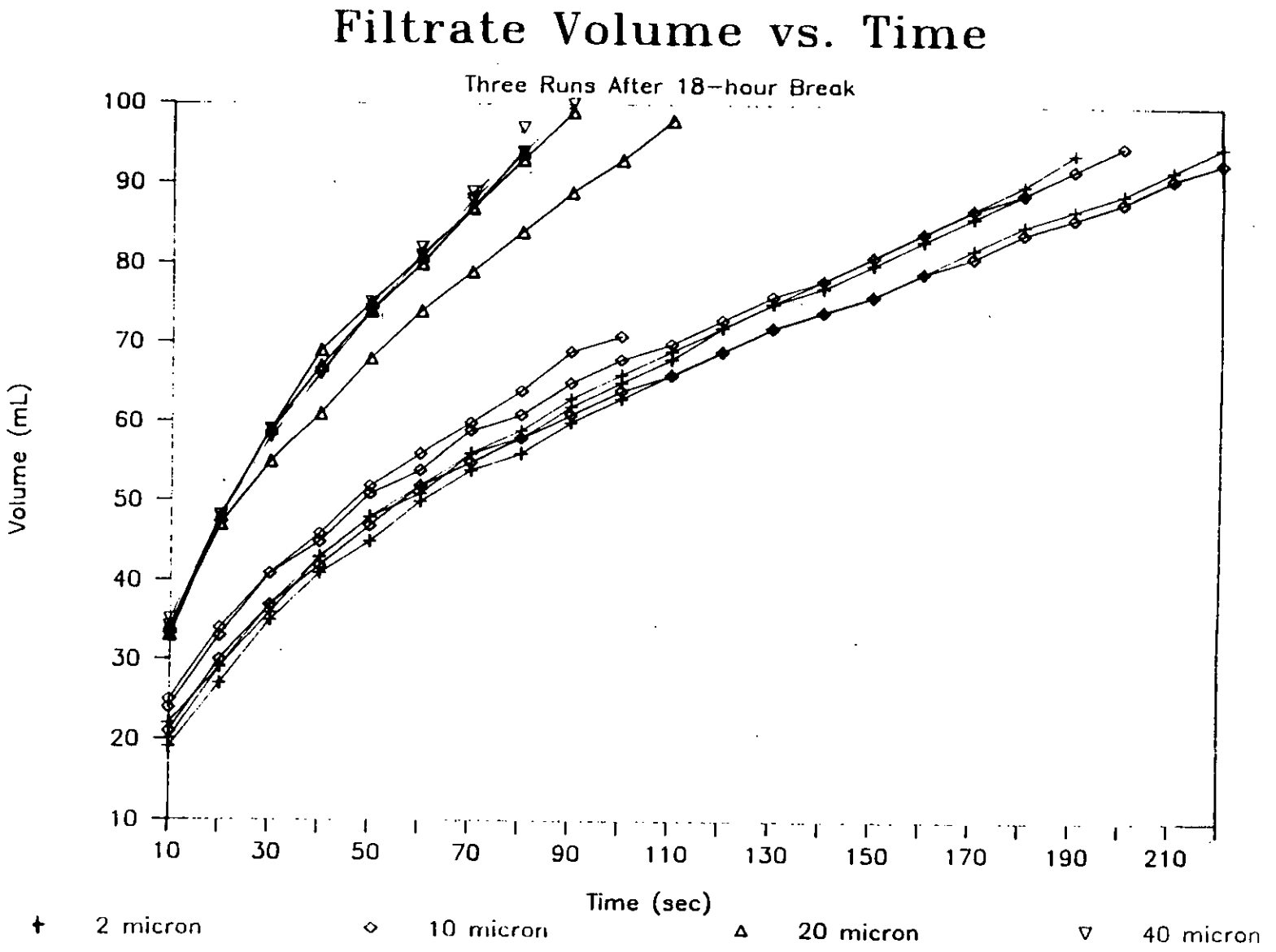

Filtrate Volume vs. Time
Three Runs After 18-hour Break
Volume (mL)
100
90
80
70
60
50
40
30
20
10
10
30
50
70
90
110
130
150
170
190
210
Time (sec)
2 micron
10 micron
20 micron
40 micron

Figure 12

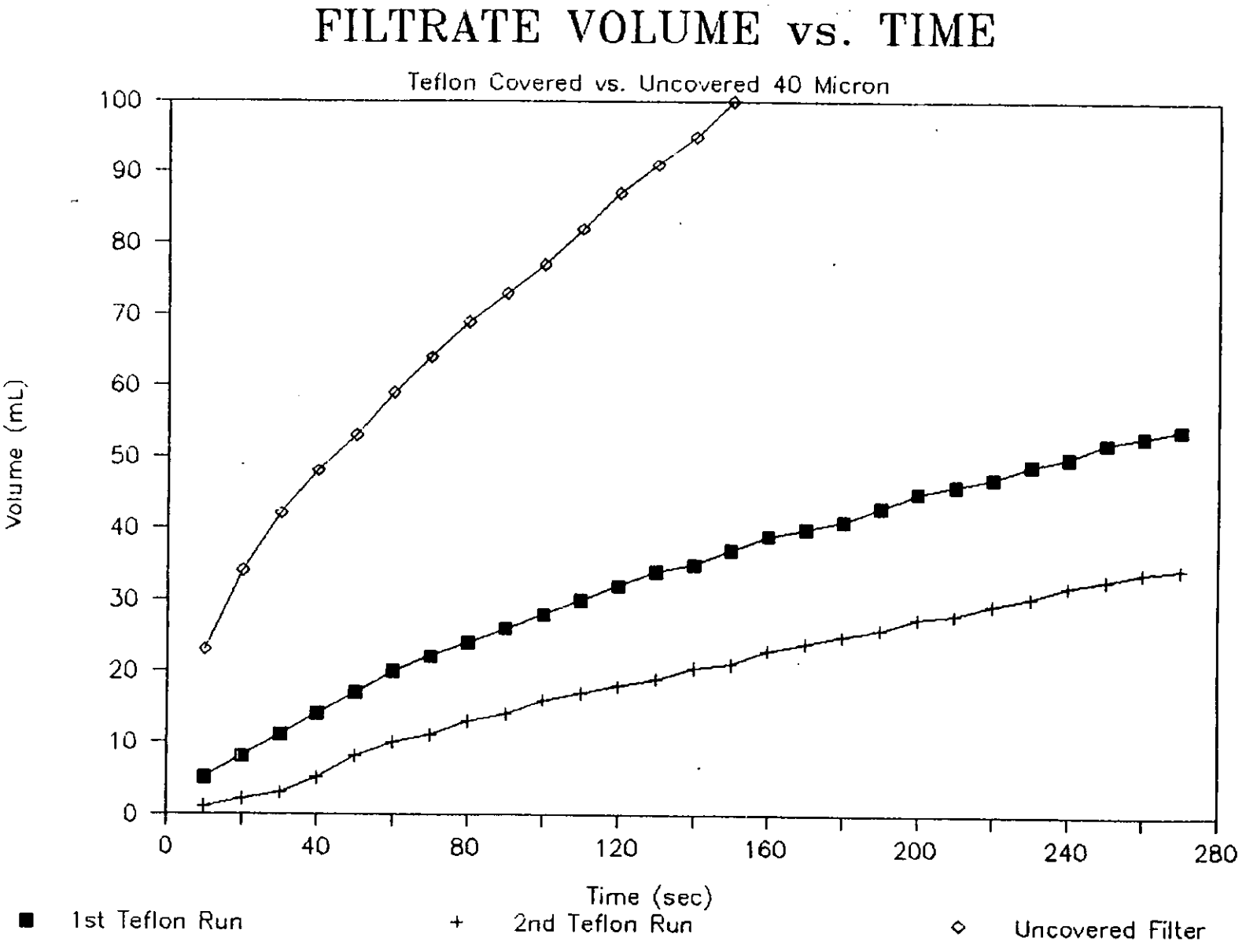

FILTRATE VOLUME vs. TIME
Teflon Covered vs. Uncovered 40 Micron
Volume (mL)
100
90
80
70
60
50
40
30
20
10
0
0
40
80
120
160
200
240
280
Time (sec)
1st Teflon Run
2nd Teflon Run
Uncovered Filter

Figure 13

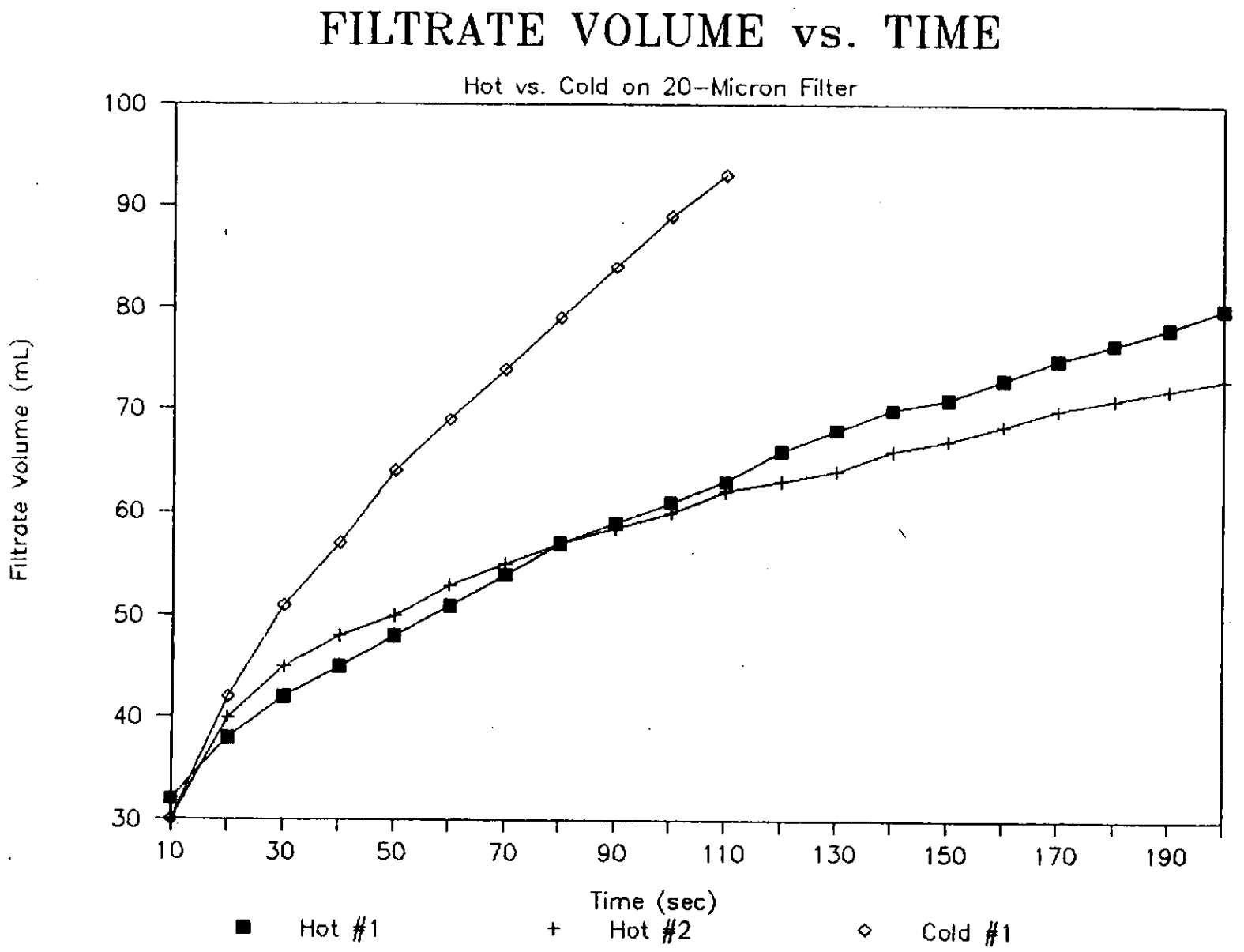

FILTRATE VOLUME vs. TIME
Hot vs. Cold on 20-Micron Filter
Filtrate Volume (mL)
100
90
80
70
60
50
40
30
10
30
50
70
90
110
130
150
170
190
Time (sec)
Hot #1
Hot #2
Cold #1

Figure 14

STATISTICS

	MEAN Diameter	S.D.
Number, Length :	2.17 µm	3.63 µm
Number, Area :	4.23 µm	4.18 µm
Number, Volume :	7.34 µm	6.32 µm
Length, Area :	8.25 µm	10.66 µm
Length, Volume :	13.48 µm	11.88 µm
Area, Volume :	22.03 µm	16.83 µm
Volume, Moment :	34.89 µm	15.77 µm

	MEDIAN Diameter	MODE	CONFIDENCE
Number :	1.02 µm	0.70 µm	99.70%
Area :	15.89 µm	46.03 µm	99.96%
Volume :	36.70 µm	46.03 µm	100.00%

Table 1