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FIELD TESTING OF NEAR-REAL-TIME MATERIALS ACCOUNTANCY AT THE PNC-TOKAI REPROCESSING PLANT

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POWER REACTOR AND NUCLEAR FUEL DEVELOPMENT CORPORATION

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要旨

期間　１９８０年4月～１９８５年１２月

目的　再処理工場において行ったニア・リアル・タイム計量管理の実証試験の成果を報告する。

要旨

再処理工場の保障措置の効果を高めることを目的として構築されたニア・リアル・タイム計量管理（ＮＲＴＡ）の実証試験を動燃東海再処理工場で実施した。

実証試験は１９８０年のＣ－１キャンペーンから開始し、その後データの蓄積のため継続して実施してきている。特に、１９８５年の後半の８５－２キャンペーンでは、より実際的な形で実証試験を行うために査察行為を模擬した形で取り入れ、ＩＡＥＡへの支援プログラムの一つとして実施した。

本報告書は、Ｃ－１キャンペーンから８５－２キャンペーンまでに実施した実証試験により得られたデータおよびＮＲＴＡの効果などに関する知見をとりまとめたものである。

なお、本報告書はＩＡＥＡと平行して、２機関が同時に発行するものである。

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* 再処理工場処理部化学処理第一課、**再処理部管理課　*** 安全部安全研究課
****再処理部、***** ＩＡＥＡ

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1. Introduction

The investigation of near-real-time materials accountancy in Japan began in 1978, with the initial definition and discussion of what is generally referred to as the "ten day detection time" model. In very general terms that model suggested that it should be possible to take in-process physical inventories in the process MBA of the PNC-Tokai reprocessing plant on a nominally weekly basis, with PNC completing at least a provisional analysis of all samples within three days after the inventory taking itself. (This model is described in greater detail in Chapter 2.)

Subsequent to the initial discussions, investigations proceeded along two parallel lines. JAERI concentrated on statistical calculations based on model data, and in general showed that the ten day detection time model would be effective in terms of an assumed detection goal of 8 kgs Pu in ten days (for abrupt diversion) or 8 kgs Pu in twelve months (for protracted diversion). PNC concentrated on the practicalities of taking in-process physical inventories without disturbing process operations, a universally agreed boundary constraint if near-real-time accountancy was to work at all.

In 1980 PNC undertook, on an internal basis, to study the practicality of near-real-time accountancy by collecting the necessary weekly in-process inventory data for two process campaigns, identified as C-1 and C-2. This data was jointly evaluated by PNC, JAERI and the IAEA in early 1981. A summary presentation was published by Ikawa in the Summer of 1981 [1], and the complete data was later published in a joint PNC-JAERI technical report [2].

Data collection, still on an internal basis, continued during 1981 and 1982, although there was an extended period during the 81-1 campaign when no in-process inventories were taken. Very little spent fuel was processed during 1983, and although a few isolated in-process inventories were taken, the data has little significance in terms of near-real-time accountancy.

During 1983 the IAEA, noting that there was considerable theoretical data to support near-real-time accountancy but very little practical field data, inquired concerning the possibility of conducting a field test or demonstration in the PNC-Tokai facility. After an extended plant maintenance shutdown during most of 1983 and all of 1984, operations were resumed in the Spring of 1985. At the same time further discussions between PNC and the IAEA were held, leading to a decision, in the Summer of 1985, to conduct a joint PNC-IAEA field test under the JASPAS support programme during the 85-2 campaign.

The purpose of the field test, as with the earlier internal field test work performed by PNC, was to demonstrate the practicality of near-real-time accountancy using the ten day detection time model and to collect meaningful real data (as contrasted with model data) to evaluate the potential effectiveness of near-real-time accountancy in IAEA safeguards.

The 85-2 reprocessing campaign began on 12 September 1985 and ended on 27 November 1985. The final cleanout physical inventory was taken on 10 December. During this period a total of 12 weekly material balances were prepared.

This report summarizes, discusses and evaluates the data collected, not only during the 85-2 joint field test, but also during all previous campaigns. Chapter 2 gives a description of near-real-time accountancy as it has developed at PNC-Tokai, although the reader is referred to [3] for a more complete description. Chapter 3 reviews the procedures followed in the collection of the data, in particular during the 85-2 campaign joint field test.

Chapter 4 presents the "raw" data collected. Chapter 5 then presents what is here termed an "Engineering Evaluation" of the data. The discussion in Chapter 5 is directed at a technical understanding of the data and at the resolution of a few identified problems which otherwise limit the usefulness of the data, not at a mathematical evaluation of possible diversion detection. The discussion accordingly is largely qualitative and subjective. Chapter 5 also presents a brief analysis of the data in terms of the "running book inventory" concept which some have suggested would be simpler and at least adequately effective.

Since the emphasis to date has been on a technical understanding of the collected data, relatively few statistical analyses have been performed. Except for some linear regression work included in Chapter 5, no statistical analysis is included in this report. Extensive statistical studies are underway, and will be published in follow-up reports.

Chapter 6 discusses a number of important questions regarding work load, effect of unmeasured inventory fluctuation, data requirement, verification and authentication, and effectiveness of NRTA. Finally, Chapter 7 summarizes the conclusions which can be drawn from the field test work.

2. Summary Description of N.R.T. Accountancy at the PNC-Tokai Reprocessing Plant

Many versions of near-real-time materials accountancy (or dynamic materials accountancy, an earlier name) have been proposed. In the version studied at PNC-Tokai in-process physical inventories of the reprocessing plant process area are taken on an approximately weekly basis. The original proposal called for all material balance data to be complete, at least on a provisional basis, within three days after the inventory, satisfying a requirement expressed in the late 1970's for a ten day detection time. Although this requirement has gradually been relaxed over the years, an attempt was made during the 1985 joint field test to enforce a ten day time restriction. The earlier data was collected on an internal basis by PNC staff. Although no particular timeliness requirement was enforced, most material balances in fact were completed within a period of one to three weeks after the inventory date.

A joint PNC-JAERI-IAEA report [3] describes the "ten day detection time model" in detail. In summary, input and output (product) measurements are made in the input and product accountancy tanks, using exactly the same procedures and data as is used for conventional materials accountancy. These input and product accountancy tanks are calibrated once per year, during the Summer inter-campaign shutdown, in the presence of IAEA inspectors.

The process area inventory can be sub-divided into the following categories:

a. Four buffer tanks feeding the three cycles of plutonium solvent extraction purification and the plutonium product evaporator. (Three recycle tanks were added to the list in later work.)

b. The three cycles of plutonium solvent extraction purification (mixer-settlers in all cycles).

c. The plutonium product evaporator.

d. Miscellaneous piping, small vessels, oxidation towers, etc.

The locations of the major items of process equipment and in particular the tanks where in-process inventory measurements were made, are shown graphically in Figure 2.1. The total process flow diagram and associated measurement points used in conventional materials accountancy are shown in greater detail in Figure 2.2.

The four buffer tanks and three recycle tanks are equipped with level recorders, density recorders or indicators, and sample probes, so their content is in all cases directly measureable. (This assumes that the volume of solution in the tank is at least adequate to cover the bottom of the sampling probe, a minimum of about 10 - 15% of total tank capacity. In a few instances, primarily with the recycle tanks, this constraint was not satisfied and it was necessary to impute a concentration from the history of previous tank contents. Since the exact quantity in a tank which is almost empty is in any case small, this detail problem is not believed to be significant.)

Except for the buffer tank feeding the first extraction cycle, all tanks also either are equipped with air sparges for solution mixing or can be mixed by pump recirculation. Measurements are obtained by reading the level recorders and by conversion of the result into litres using calibration equations developed by PNC.

These calibration equations have not been verified by IAEA inspection. This is considered to be of only secondary importance, however, because any inaccuracy (or falsification) would affect only the first material balance in each sequence. Thereafter the inaccurate (or falsified) calibration equation is used for both the beginning and the ending inventory, and the inaccuracies cancel. Some residual inaccuracy could still be introduced as a result of a tank being full one week and nearly empty another, but these effects also quickly cancel.

In-process physical inventories were scheduled at a time when the product evaporator was empty, thus eliminating the problem of measuring the Pu concentration in the evaporator. The evaporator operates in a batch mode, and during full production is discharged about once every 24-36 hours, so this constraint on the scheduling of in-process inventories has little practical meaning except when the process area is flushing between fuel batches. (Flushing between fuel batches occurs during a campaign when it is desired to introduce fuel of a distinctly different composition. The flushing minimizes the extent to which the two materials are commingled during processing.)

Given that a series of material balances, and resulting MUF values, have been computed based on weekly in-process physical inventories, the next question is how best to evaluate the resulting data. The Japan Atomic Energy Research Institute (JAERI), under a research contract between PNC and JAERI, has developed a mini-computer system which has a capability of storing and evaluating the field test data. The hardware configuration of the system is shown in Fig. 2.3. One unique feature of the system is that the safeguards related organizations, i.e., IAEA, the Government of Japan, and the plant operator, can use the common data base through the JAERI's standard software or an idependent one which will be developed by each organization. Details are described in [4].

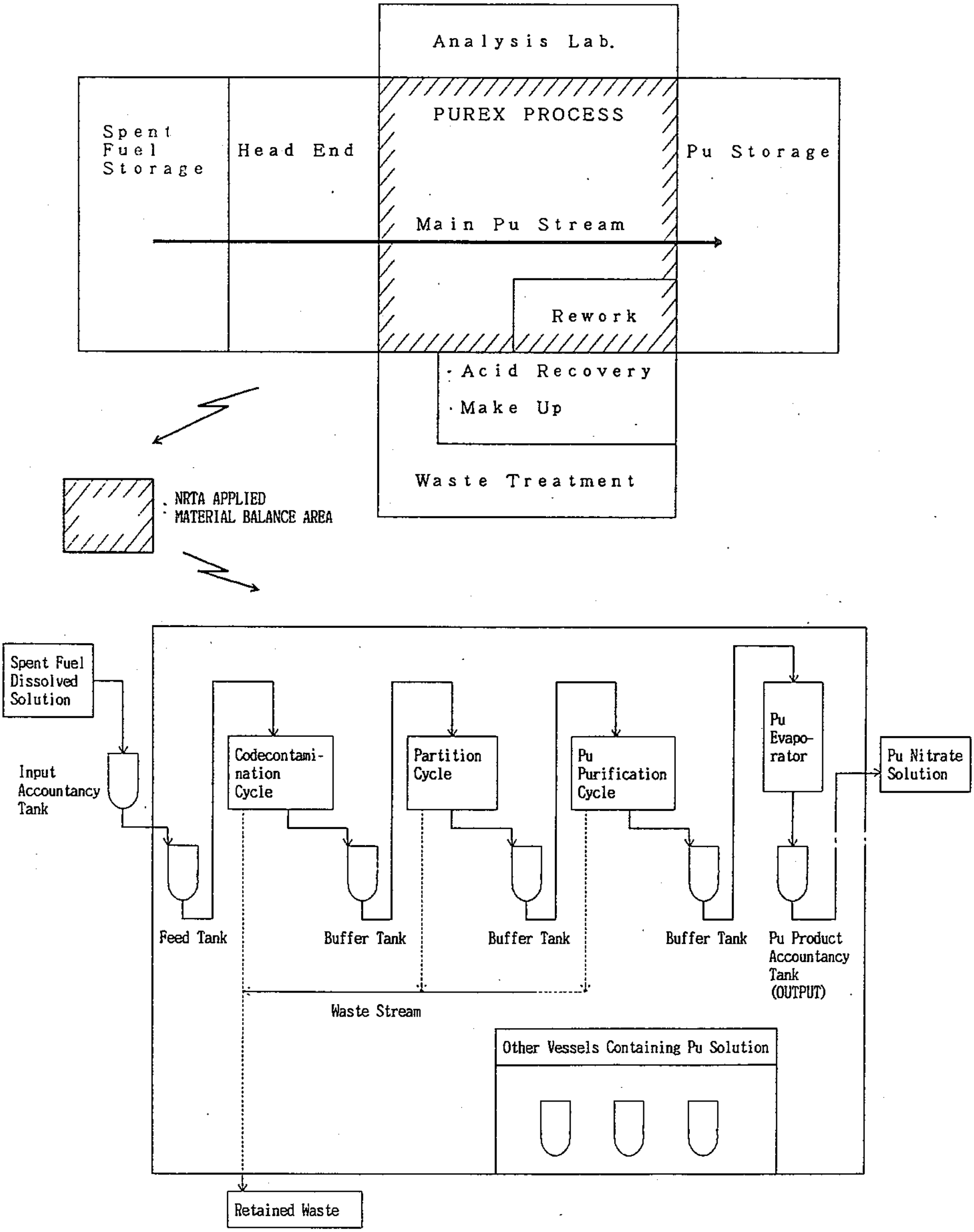
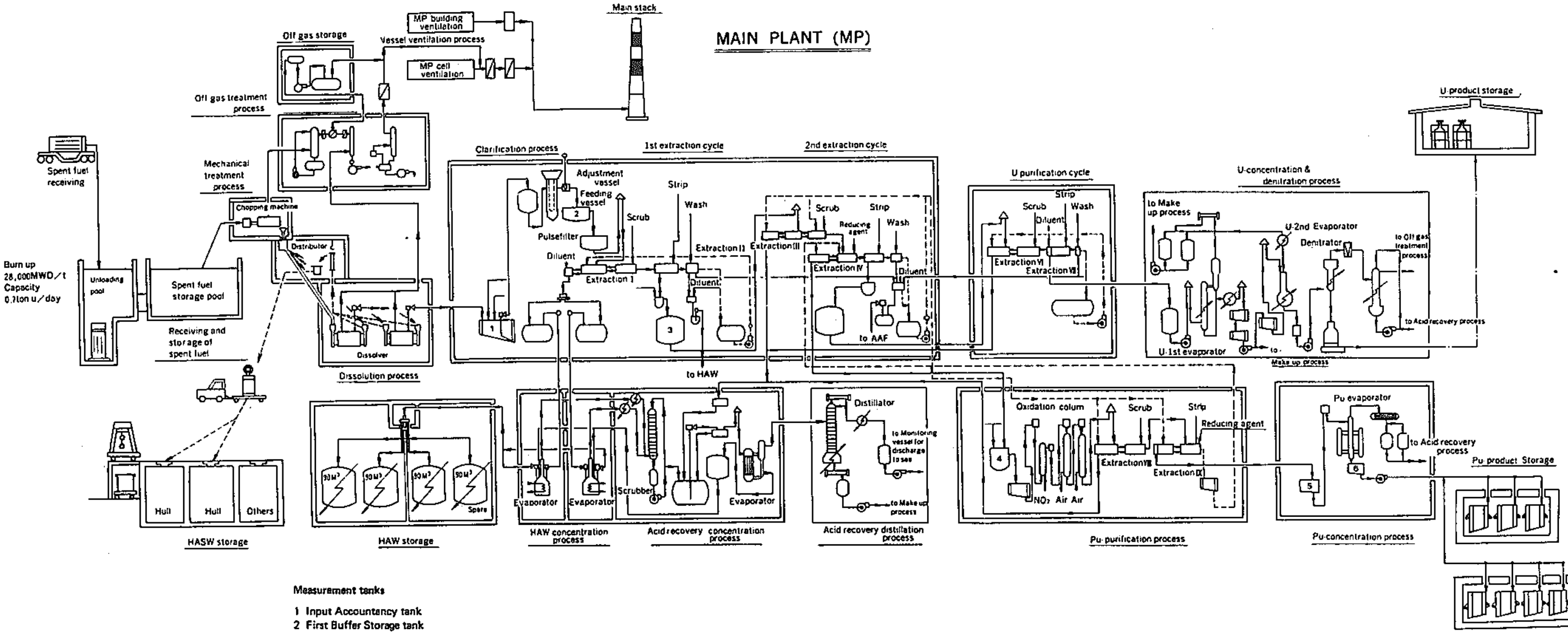


Fig. 2.1
NRTA applied material balance area, and its main equipments in the main Pu Stream. Inventories in above described tanks are directly measured as in-process inventory, when Pu product evaporator is empty. Three extraction cycle inventories are basically regarded as constant values.

Figure 2.2

PNC REPROCESSING PLANT PROCESS FLOW DIAGRAM



Measurement tanks

1 Input Accountancy tank
2 First Buffer Storage tank
3 Second Buffer Storage tank
4 Third Buffer Storage tank
5 Evaporator Feed tank
6 Product Accountancy tank

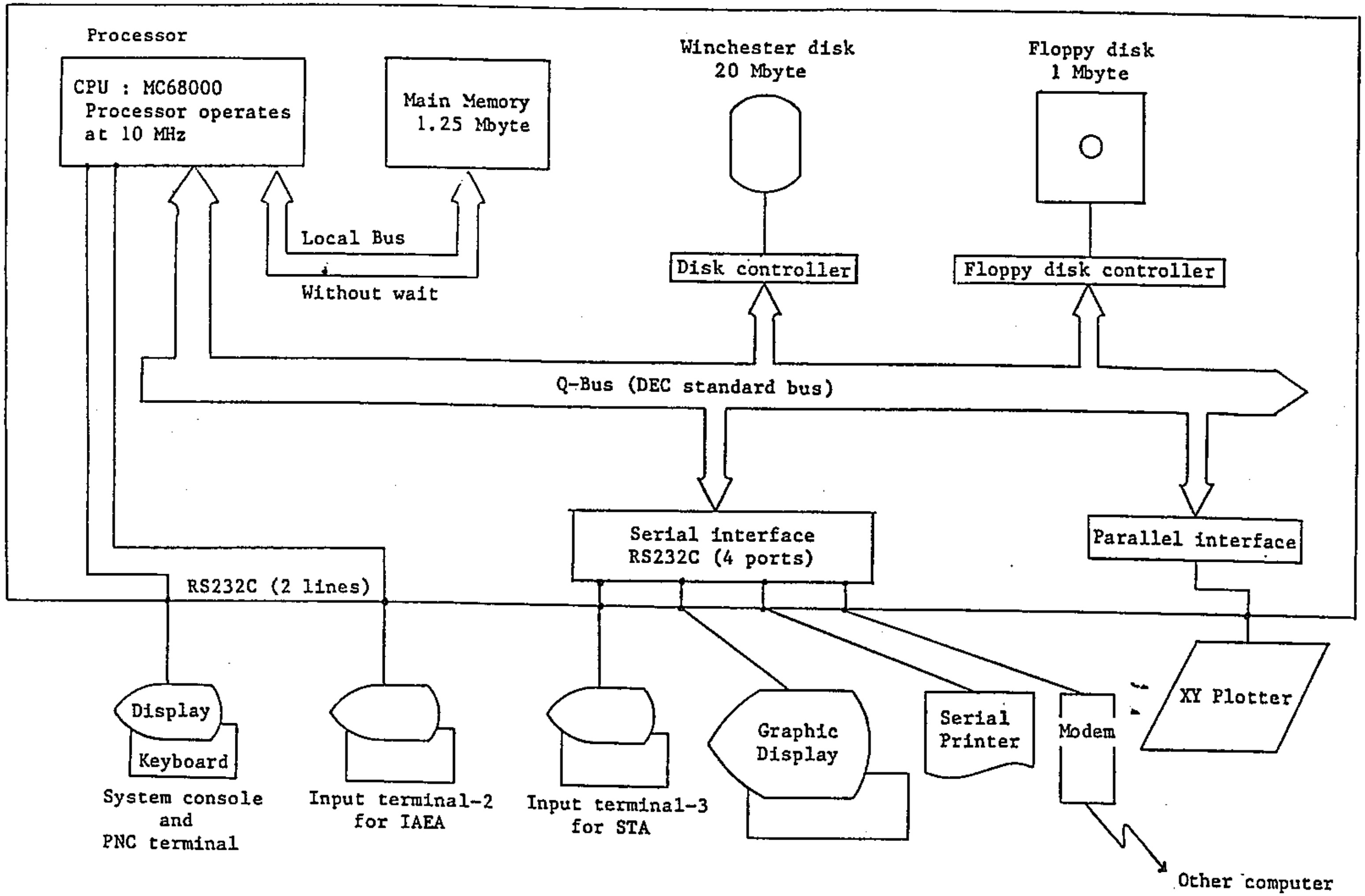


Fig. 2.3 Hardware Configuration of Data Processing System for Near-Real-Time Materials Accountancy

3. The 85-2 Campaign Field Test

A joint IAEA-PNC field test of near-real-time accountancy was discussed informally as early as 1981, and was proposed formally by the IAEA in 1982. Since the reprocessing plant operated very little in 1983 and not at all in 1984, discussions continued intermittently until the Summer of 1985, when the government of Japan formally proposed that the field test be included in the JASPAS programme and performed during the 85-2 campaign scheduled to begin in September 1985. This Chapter describes the procedures followed. The resulting data is presented in Chapter 4, and an "engineering evaluation" is given in Chapter 5.

Since PNC had already collected a significant body of field test data, this joint field test should be viewed as adding two new elements. One is the involvement of IAEA staff (in a developmental role) in the actual collection of in-process inventory data. The other is the commitment by PNC to provide provisional material balance data within three days after each in-process inventory, and the corresponding commitment of the IAEA development staff to perform preliminary data evaluations in a timely manner.

The paragraphs that follow have been summarized from the written working procedures agreed to by PNC and the IAEA.

Research Status. The demonstration is understood to be a field test of near-real-time materials accountancy concepts, undertaken as part of a broader IAEA programme to develop more effective safeguards for reprocessing. The participants hope that a successful demonstration can contribute not only to improved safeguards effectiveness but also to reduced inspector effort. The demonstration is understood not to be connected automatically to any future implementation of near-real-time materials accountancy in the Tokai reprocessing plant.

Overview Description. The suggested advanced safeguards approach for PNC-Tokai [3], contains several elements that are new or different in comparison with current safeguards practice. The demonstration relates to one element, the use of near-real-time materials accountancy in the process MBA to achieve both an increased detection timeliness and an increased detection sensitivity. The field test of near-real-time accountancy involves the sequential analysis of material balance data generated at nominal weekly intervals and based on in-process physical inventories. (A "daily" material balance was also studied during the 85-2 campaign, see Section 4.4.) The in-process physical inventories, in turn, are based on routine process measurements in volume and Pu concentration at the following process points.

- total solution volume in each of the buffer tanks feeding the three cycles of solvent extraction, the buffer tank feeding the product evaporator, and three other recycle storage buffer tanks; and
- Pu concentration at each of the seven inventory points identified in Figure 2.1.

In-process inventories were scheduled at times when the product evaporator was empty, eliminating the need for special inventory procedures for the evaporator.

The following current routine inspection procedures were used to observe and verify input and product output measurements by the IAEA staff members.

- electromanometer for input and output volume measurement
- K-edge densitometer for product Pu concentration measurement
- chemical analyses in the IAEA analytical laboratory.

The demonstration also included the following elements not present in routine safeguards procedures.

- collection of in-process inventory measurement data in the presence of IAEA staff members; and
- statistical analysis of the resulting sequential material balance data on a near-real-time basis, using techniques which specifically recognize the sequential nature of the data.

Data evaluation was performed by IAEA staff members with the assistance of PNC and JAERI. For this purpose, PNC and JAERI prepared a mini-computer which could be used on a current basis. Designated inspectors were included among the IAEA staff members participating in the demonstration. However, the non-routine data collected in the demonstration were still recognized as demonstration data and were not included with routine inspection data.

<u>Detailed Data Collection and Verification Procedures.</u> Input flow batches consisting of spent fuel solution were measured at the input accountancy tank according to the following PNC procedure.

- Sample taking and volume measurement
  - (i) sample taking at the sampling bench
  - (ii) water column manometer reading for measuring full level and solution density
  - (iii) water column manometer reading for measuring heel level
- Chemical analysis in the analytical laboratory
  - (i) solution density measurement for accountancy
  - (ii) Pu concentration measurement for accountancy

Solution density can be measured not only by analysis in the analytical laboratory but also from the manometer readings. The analytical data are usually reported later than one week after the sample was taken. Thus the manometer data were used as provisional data which was later replaced by the analytical data. Similarly, Pu concentration analytical data used for accountancy are usually reported later than one week after the sample was taken. Again, a quick analysis normally used only for process control was used to determine a provisional Pu concentration where necessary. (This problem of analytical measurement timing is discussed in Chapter 6. Under current guidelines for detection timeliness provisional measurements probably are unnecessary.)

PNC staff members attempted to deliver at least provisional data not later than three days after the time of each in-process physical inventory. Provisional data was later replaced by the more accurate data when the latter became available. Following the standard routine inspection procedures a portion of all input samples was prepared for transportation to the IAEA safeguards analytical laboratory.

Product flow batches consisting of concentrated plutonium nitrate were measured at the product accountancy tank according to the following PNC procedure.

- Sample taking and volume measurement
    - (i) sample taking at the sampling bench
    - (ii) water column manometer reading for measuring full level and solution density
    - (iii) water column manometer reading for measuring heel level
- Chemical analysis in the analytical laboratory
    - (i) solution density measurement for accountancy
    - (ii) Pu concentration measurement for accountancy

As with input flow measurement, IAEA staff members observed the sampling and manometer readings, and recorded all verification data in accordance with routine inspection procedures. The electromanometer was used for the verification of this volume measurement.

Solution density can be measured not only by analysis in the analytical laboratory but also from manometer readings. The analytical data are usually reported later than five days after the sample was taken. Thus, the manometer data was used as provisional data and was later replaced by analytical data. Similarly, Pu concentration analytical data for accountancy are usually reported later than five days after the sample taking. In this case the K-edge densitometer measurement performed for the verification was used for the provisional data determination. This K-edge densitometer measurement data likewise was later replaced by analytical data.

Waste discards from the process MBA were measured at the highly active liquid waste (HAW) concentration evaporator, because almost all waste from the process MBA are collected and concentrated at this evaporator. PNC measured the volume of HAW and the concentration of Pu, and delivered these values to the IAEA staff members, as the data became available. Verification was in accordance with routine inspection procedures.

In-process physical inventories were taken at nominally weekly intervals. All inventories were scheduled to coincide with discharge of the plutonium product evaporator, and PNC attempted to schedule in-process inventories as close to weekly as was practical. However, in-process inventory schedules depended solely on the process status. PNC advised IAEA staff members participating in the field test concerning proposed inventory schedules as early as possible. It was then the responsibility of the IAEA staff member to be available at the time the inventory was scheduled. (In the event, all in-process inventories were observed.)

In-process physical inventories were taken by PNC in accordance with the following procedures.

- Timing of inventories. From immediately after the discharge of the Pu product evaporator to before the evaporator is refilled to begin the next cycle. Since the discharged product batch was not yet measured, it was counted as still being in the process MBA.
- Volume measurements Recording the solution levels (%) from the strip chart recorder, in each of four buffer storage tanks feeding the three extraction cycles and feeding to the Pu product evaporator, and three other recycle buffer storage tanks (total seven tanks), at the same time. Data conversion was performed using conversion equations developed by PNC.
- Sample taking Sample taking from the seven tanks at the same time (within about one hour). For the three recycle buffer storage tanks, samples were not taken if the solution level(%) had not changed from the level determined during the previous inventory. In this case, previous analytical values were used.
- Chemical analysis Pu concentration and solution density was measured at the analytical laboratory, using a quick analytical procedure normally used for process control but not for accountancy.

IAEA staff members observed the recording of all tank solution levels(%) from the appropriate strip chart recorders, but did not observe the sample taking from all inventory tanks. Chemical analytical data was delivered to the IAEA staff members not later than three days after the date of inventory.

4. The Field Test Data: I. "Raw" Data

As noted in the Introduction, the field testing of near-real-time materials accountancy following the "ten day detection time" model began at PNC in 1980, and with a few exceptions PNC has taken in-process physical inventories at approximately weekly intervals during every reprocessing campaign since that time. Figure 4.1 shows the general operating history of the PNC reprocessing plant and the timing and duration of the reprocessing campaigns. Table 4-1 summarizes the dates of the eight campaigns of interest here, together with the number of material balances (DPITs in the table) prepared, the total quantities of material processed, etc.

Table 4.1 Historical Data of Field Test of NRTA in the PNC Tokai Reprocessing Plant

Campaign Name	Field Test Period from ~ to	days	Number of DPIT	Reprocessed Ammount(MTU)
C-1	80/ 3/ 3 ~ 80/ 8/ 1	153	6	28.5
C-2	80/ 8/ 1 ~ 80/12/ 9	132	7	19.5
81-1	80/12/ 9 ~ 81/ 7/ 7	212	4	26.8
81-2	81/ 7/ 7 ~ 81/12/18	166	5	14.0
82-1	81/12/18 ~ 82/ 7/13	209	16	30.9
82-2	82/ 7/13 ~ 82/12/15	157	11	20.1
85-1BC	85/ 3/31 ~ 85/ 8/20	173	15	49.2
85-2	85/ 8/20 ~ 85/12/10	114	11	29.5
Total	80/ 3/ 3 ~ 85/12/10		75	218.5

Table 4-1. Summary of Reprocessing Campaigns and Field Test Data at PNC-Tokai

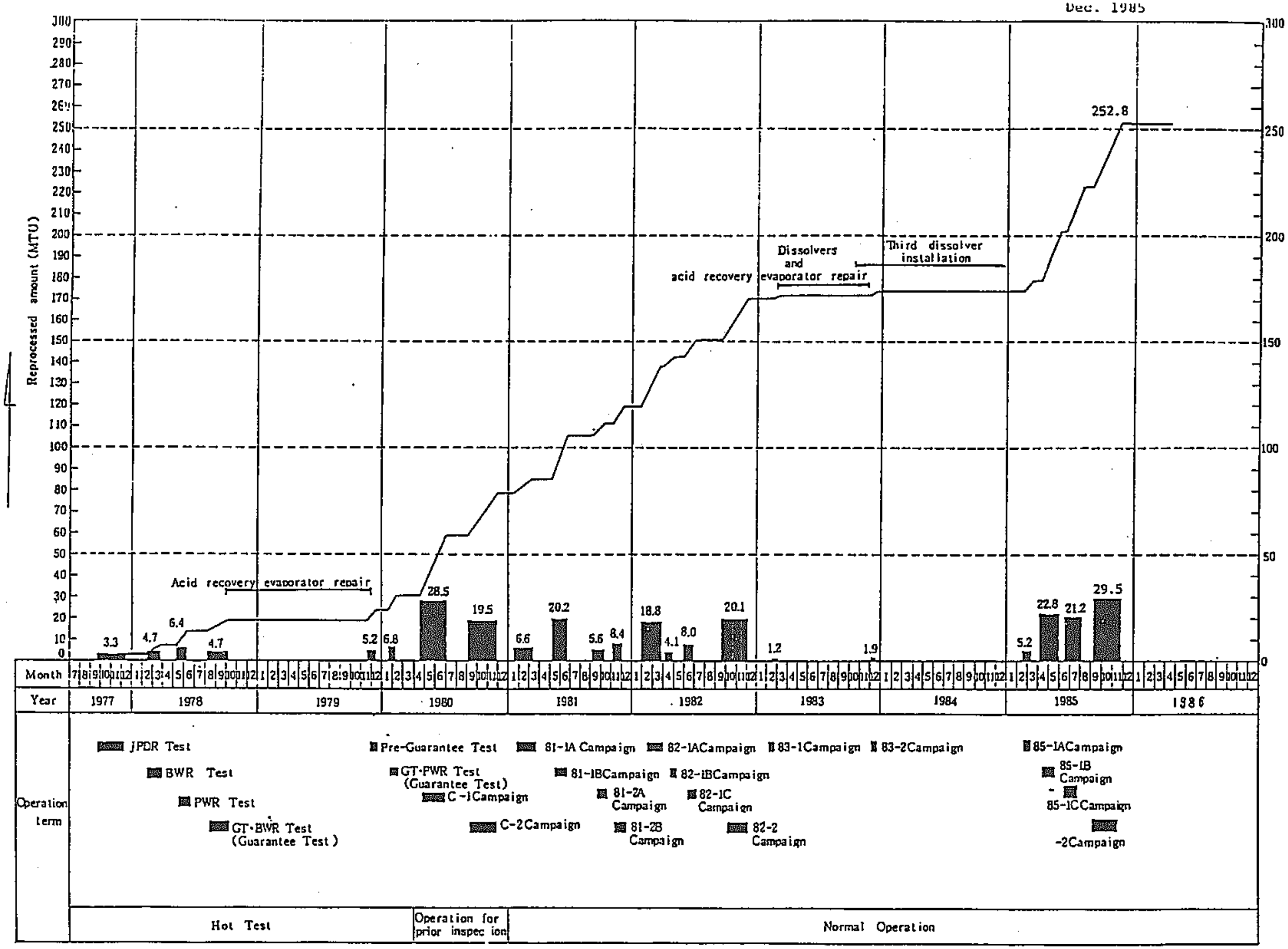


Fig.4.1 Operation on Tokai Reprocessing Plant

For reasons which will become apparent in the discussions that follow, it is convenient to divide this data into three groups, as follows:

a. campaigns C-1, C-2 and 81-1, representing all fuel reprocessed during 1980 and the first half of 1981.

b. campaigns 81-2, 82-1 and 82-2, representing all fuel reprocessed during the second half of 1981 plus all of 1982.

c. the 85-1 B/85-1C campaign, representing fuel reprocessed during the Spring of 1985, plus the 85-2 campaign, which is the specific subject of the near-real-time accountancy field test.

Various adjustments and corrections have been made to the field test data, notably in connection with the unmeasured solvent extraction system inventory. In order to minimize confusion and give the interested reader the opportunity to review both the adjustments and their effect on the data and the conclusions to be drawn from it, this Chapter presents the original "raw" data. Statisticians and others who wish to perform their own data analyses are, of course, free to analyze whatever data they choose, but the authors believe that the adjusted data presented in Chapter 5 gives the best available estimates of the total in-process physical inventories and the corresponding short term MUF values.

4.1 Campaigns C-1,C-2 and 81-1

Figures 4.2, 4.3 and 4.4 show the operating history for the C-1, C-2 and 81-1 campaigns respectively. The C-1 and C-2 campaigns were the first reprocessing campaigns involving more than 5 - 6 tons of spent fuel, and they were the first campaigns in which near-real-time materials accountancy was tried. The result is that in-process inventories were sometimes taken at intervals which differed from the intended weekly schedule. In most cases this was the result of the boundary constraint that in-process inventories can only be taken when the product evaporator is discharged.

The reader's attention is also drawn to the fact that there was an extended period of reprocessing during the latter part of the 81-1 campaign during which no in-process inventories were taken.

Table 4-2 presents the material balance data which was collected during the C-1, C-2 and 81-1 campaigns. (Waste discards, returns of analytical residues to the process area and other minor entries have been combined into general input (I) and output (O) columns.)

Table 4-2

N.R.T. Accountancy Data, Campaigns C-1, C-2, and 81-1

(not adjusted for "unmeasured inventory," see Section 5.1)

(all data in grams)

No.	BI	I	O	EI	MUF	CUMUF
1	7662*	6611	5	10433	3835	3835
2	10433	24708	25933	10621	-1413	2422
3	10621	24772	24507	11594	-708	1714
4	11594	27232	25214	11228	2384	4098
5	11228	20392	19518	14050	-1948	2150
6	14050	25163	27545	13179	-1511	639
7	13179	16471	31657	2416*	-4423	-3784
8	2416*	12997	10	12268	3135	-649
9	12268	17390	19101	10314	243	-406
10	10314	30930	33073	9984	-1813	-2219
11	9984	21983	19794	11245	928	-1291
12	11245	23802	27344	11225	-3522	-4813
13	11225	13934	12449	12018	692	-4121
14	12018	10396	13377	8456	581	-3540
15	8456	194	6476	6289*	-4115	-7655
16	6289*	25060	0	26085	5264	-2391
17	26085	0	5154	22390	-1459	-3850
18	22390	6838	14371	14561	296	-3554
19	14561	3646	5915	15360	-3068	-6622
20	15360	117215	136098	2160*	-5683	-12305

* cleanout physical inventory data

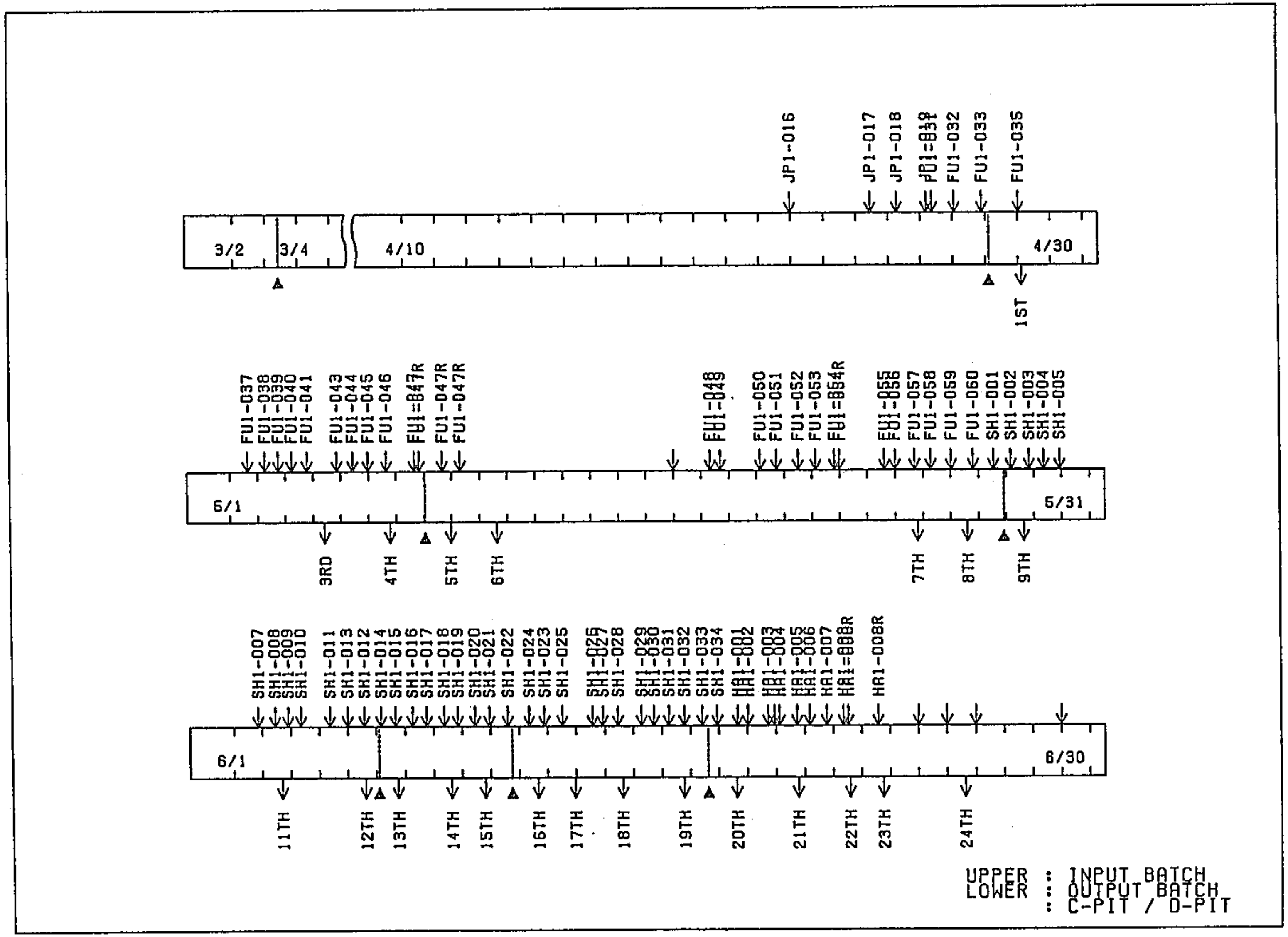


Fig. 4.2 Operation Chart for the C-1 Campaign

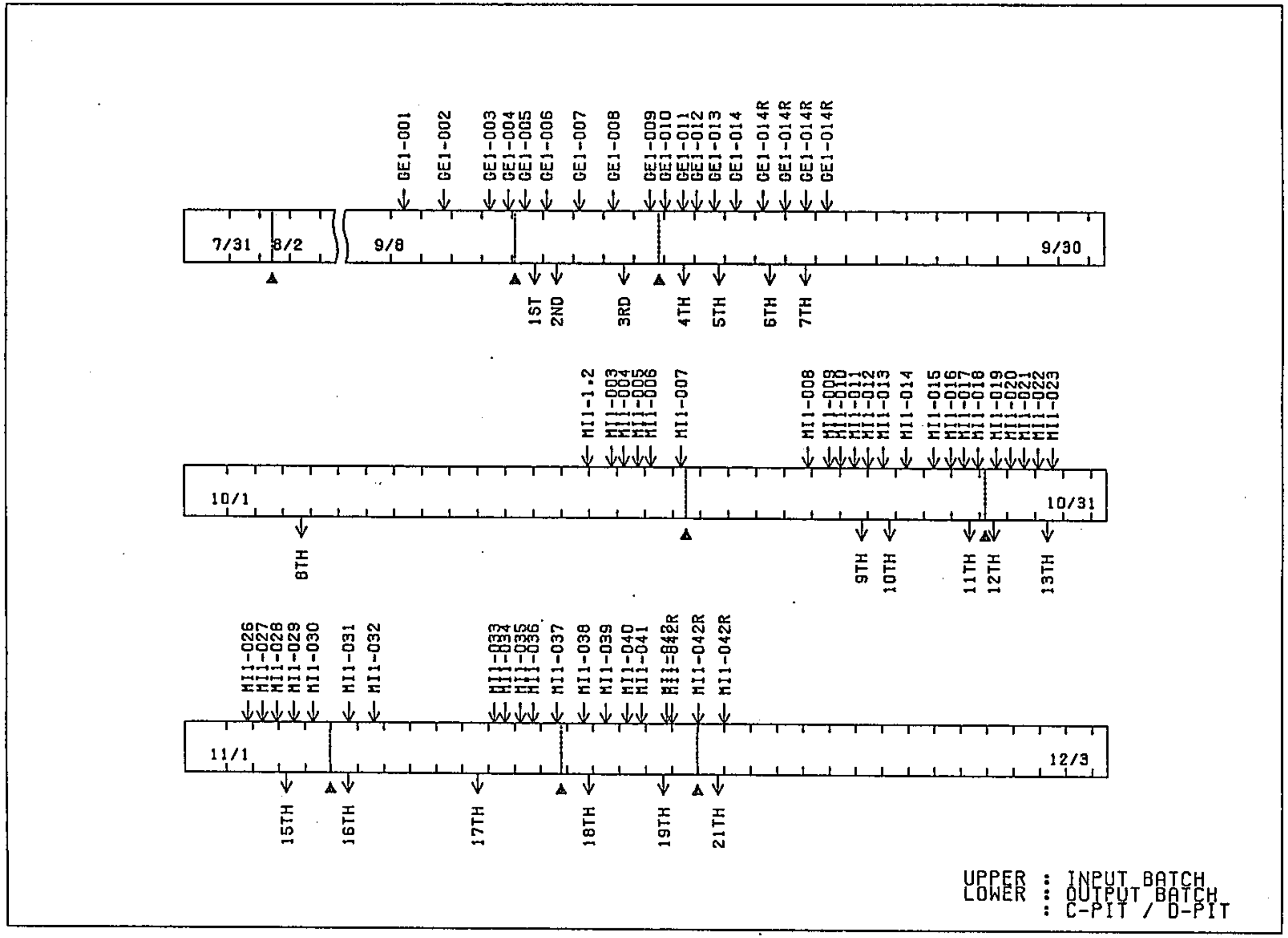


Fig. 4.3 Operation Chart for the C-2 Campaign

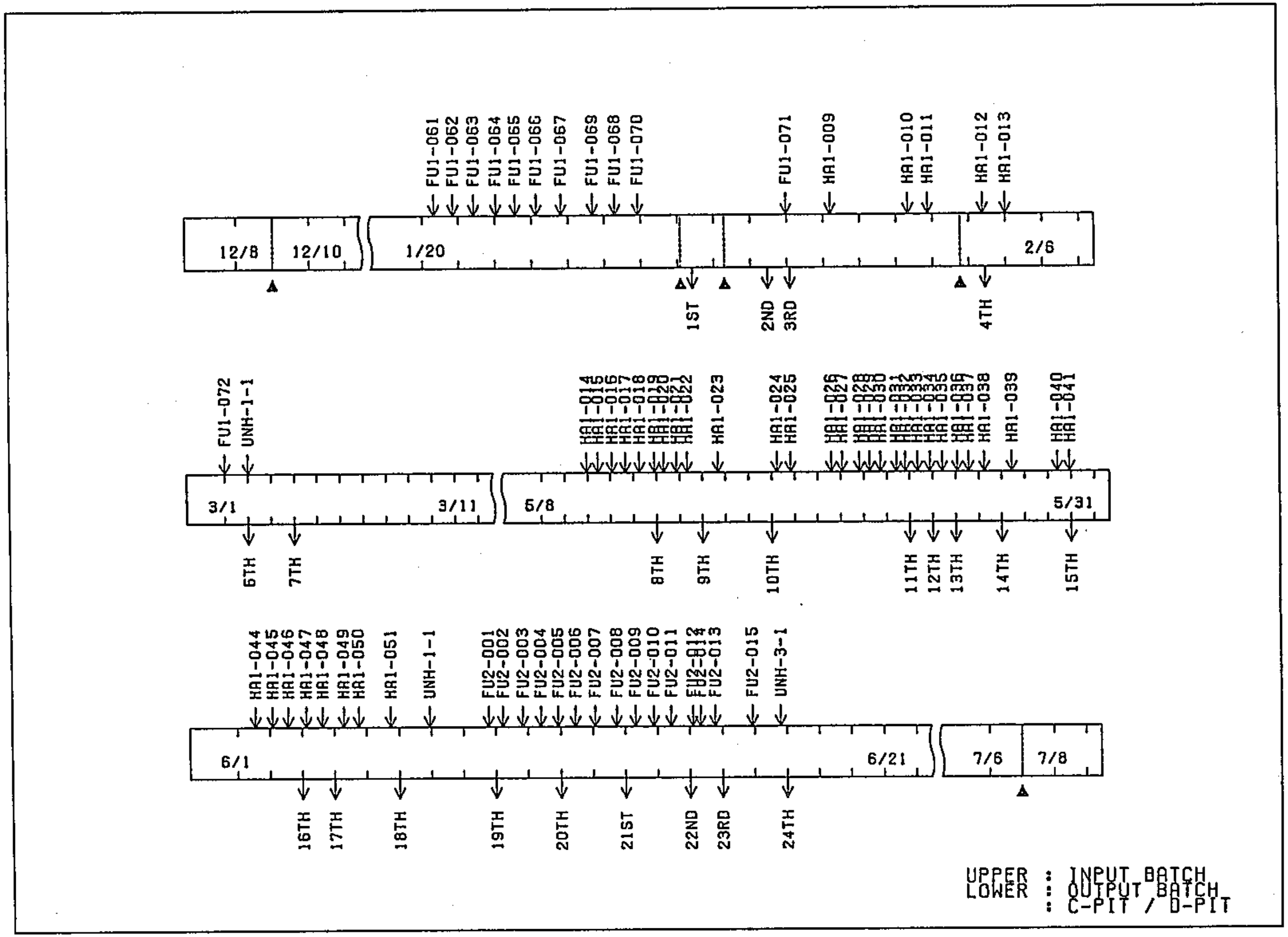


Fig. 4.4 Operation Chart for the 81-1 Campaign

4.2 Campaigns 81-2, 82-1 and 82-2

Figures 4.5, 4.6 and 4.7 show the operating history for the 81-2, 82-1 and 82-2 campaigns respectively. Table 4-3 presents material balance data which was collected during these campaigns.

Table 4-3

N.R.T. Accountancy Data, Campaigns 81-2, 82-1 and 82-2
(not adjusted for "unmeasured inventory," see Section 5.1)
(all data in grams)

No.	BI	I	O	EI	MUF	CUMUF
1	2160*	32899	12621	18862	3576	3576
2	18862	14669	28728	5867	-1064	2512
3	5867	13103	3764	13775	1431	3943
4	13775	4816	5185	12536	870	4813
5	12536	23880	36264	3805	-3653	1160
6	3805	0	3124	1508*	-827	333
7	1508*	9105	0	9672	941	1274
8	9672	19597	16967	11485	817	2091
9	11485	11765	12000	11092	158	2249
10	11092	25411	24335	12834	-666	1583
11	12834	19458	18327	14257	-292	1291
12	14257	14183	15903	12254	283	1574
13	12254	9916	12685	10968	-1483	91
14	10968	6452	4359	13075	-14	77
15	13075	5204	17562	7043	-6326	-6249
16	7043	4862	0	5354	6551	302
17	5354	5674	0	10869	159	461
18	10869	26499	23999	12347	1022	1483
19	12347	13287	11167	13750	717	2200
20	13750	9840	11703	13942	-2055	145
21	13942	4083	11578	8221	-1774	-1629
22	8221	0	7237	3598*	-2614	-4243
23	3598*	0	0	3753*	-155	-4398
24	3753*	8045	0	8301	3497	-901
25	8301	11425	4176	13450	2100	1199
26	13450	15218	12164	16687	-183	1016
27	16687	15973	17206	15553	-99	917
28	15553	18589	16254	19352	-1464	-547
29	19352	16044	17747	14188	3461	2914
30	14188	13418	12519	16788	-1701	1213
31	16788	12716	17609	10696	1199	2412
32	10696	12820	12339	11900	-723	1689
33	11900	20489	19141	14354	-1106	583
34	14354	6485	23058	1524	-3743	-3160
35	1524	0	0	1789*	-265	-3425

* cleanout physical inventory data

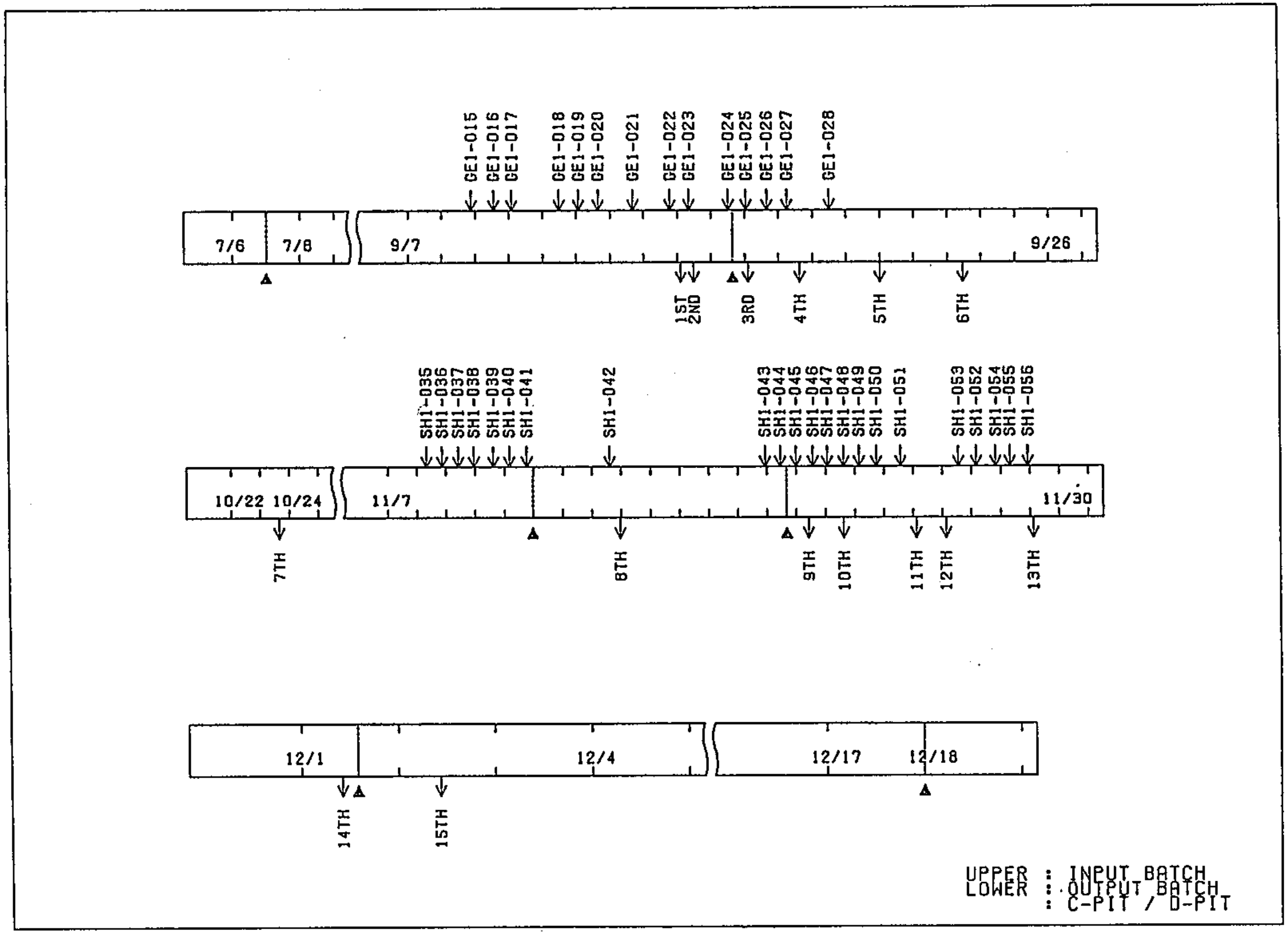


Fig. 4.5 Operation Chart for the 81-2 Campaign

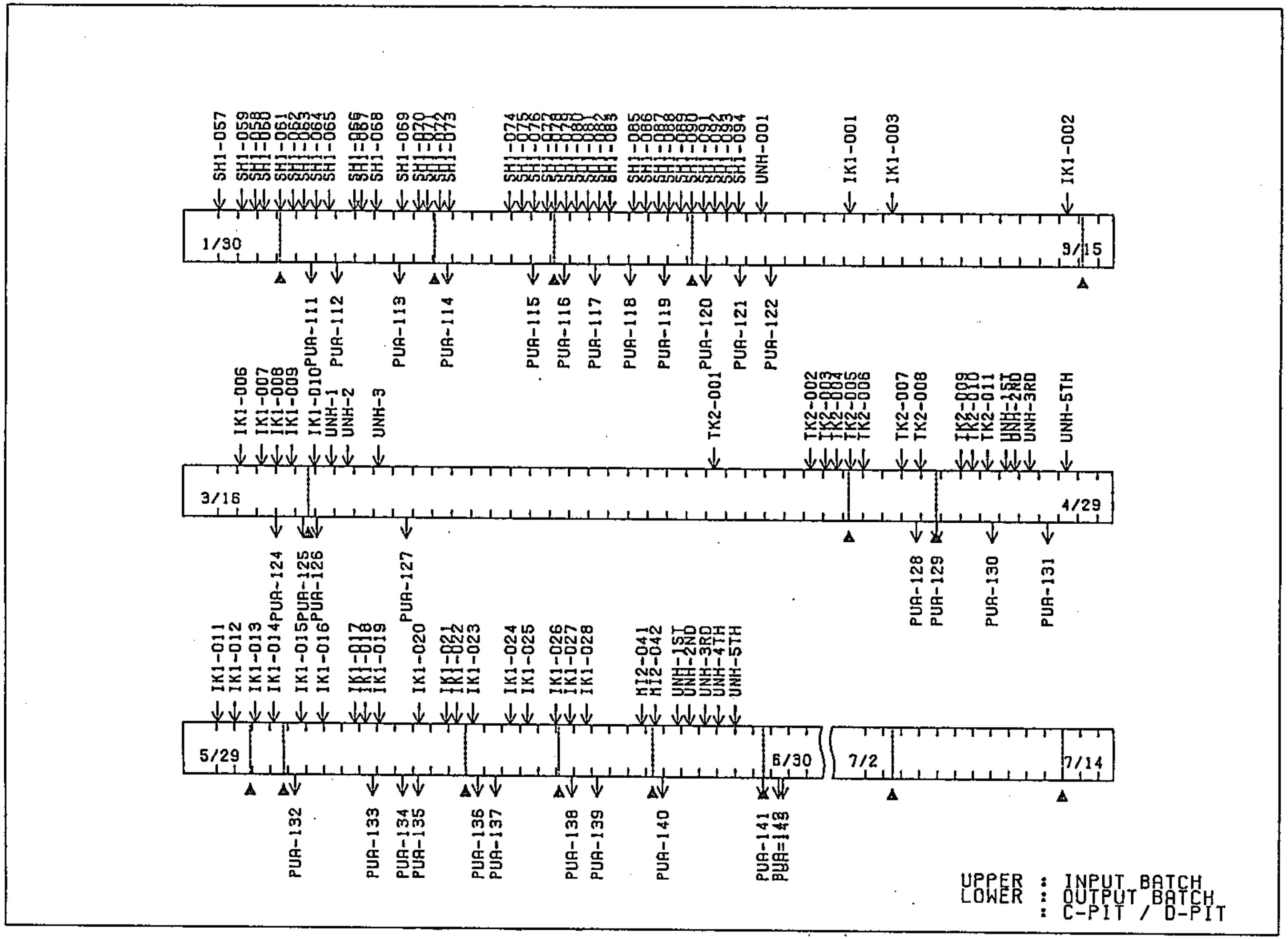


Fig. 4.6 Operation Chart for the 82-1 Campaign

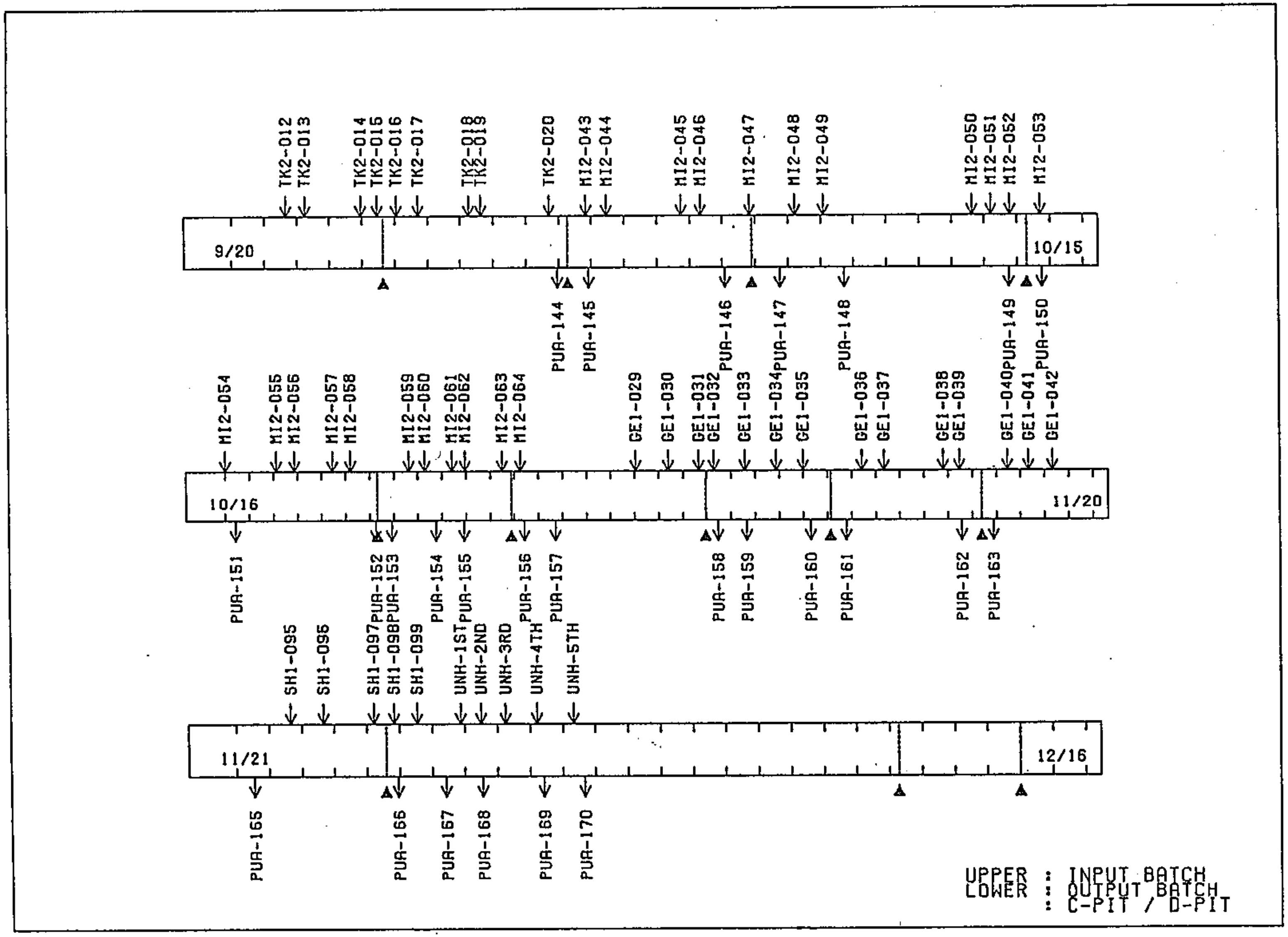


Fig. 4.7 Operation Chart for the 82-2 Campaign

4.3 Campaigns 85-1B,C and 85-2

Figure 4.8 and 4.9 show the operating history for the 85-1B, 85-1C and 85-2 campaigns. (Figure 4.8 shows both portions of the 85-1 campaign.) The 85-1A campaign was a test campaign after the major maintenance shutdown during 1984. Only 5.2 tons of spent fuel were processed, and no in-process physical inventories were taken. Table 4-4 presents the material balance data which was collected during these campaigns.

As has been discussed throughout this report, a joint PNC-IAEA field test was conducted during the 85-2 campaign, with active participation by inspectors from both the IAEA and the Japan Nuclear Safety Bureau. Data collection procedures were the same, however, and the authors see no reasons to treat this data in any separate or different manner.

Table 4-4

N.R.T. Accountancy Data, Campaigns 85-1B,C and 85-2

(not adjusted for "unmeasured inventory," see Section 5.1)

(all data in grams)

No.	BI	I	O	EI	MUF	CUMUF
1	1002*	6994	0	5388	2608	2608
2	5388	18482	12264	11366	240	2848
3	11366	19769	18898	11951	286	3134
4	11951	19576	18820	11689	1018	4152
5	11689	13251	13239	12038	-337	3815
6	12038	21826	18884	13497	1483	5298
7	13497	16643	19022	11029	89	5387
8	11029	24425	31804	6130	-2480	2907
9	6130	8493	6051	7793	779	3686
10	7793	27958	19879	14390	1482	5168
11	14390	18723	19697	12796	620	5788
12	12796	15557	18516	10876	-1039	4749
13	10876	19296	19094	10389	689	5438
14	10389	29479	26463	12380	1025	6463
15	12380	27255	32562	9667	-2594	3869
16	9667	13	8943	1944*	-1207	2662
17	1944*	13715	6	12870	2783	5445
18	12870	41028	40059	13340	499	5944
19	13340	25973	25359	13862	92	6036
20	13862	14672	18464	9431	639	6675
21	9431	24958	20554	14422	-587	6088
22	14422	17424	20208	10596	1042	7130
23	10596	193	7149	3870	-230	6900
24	3870	12021	0	15674	217	7117
25	15674	28050	26364	15868	1492	8609
26	15868	33756	33813	15578	233	8842
27	15578	4926	18604	4085*	-2185	6657
28	4085	0	3497	1018	-430	6227

* cleanout physical inventory data

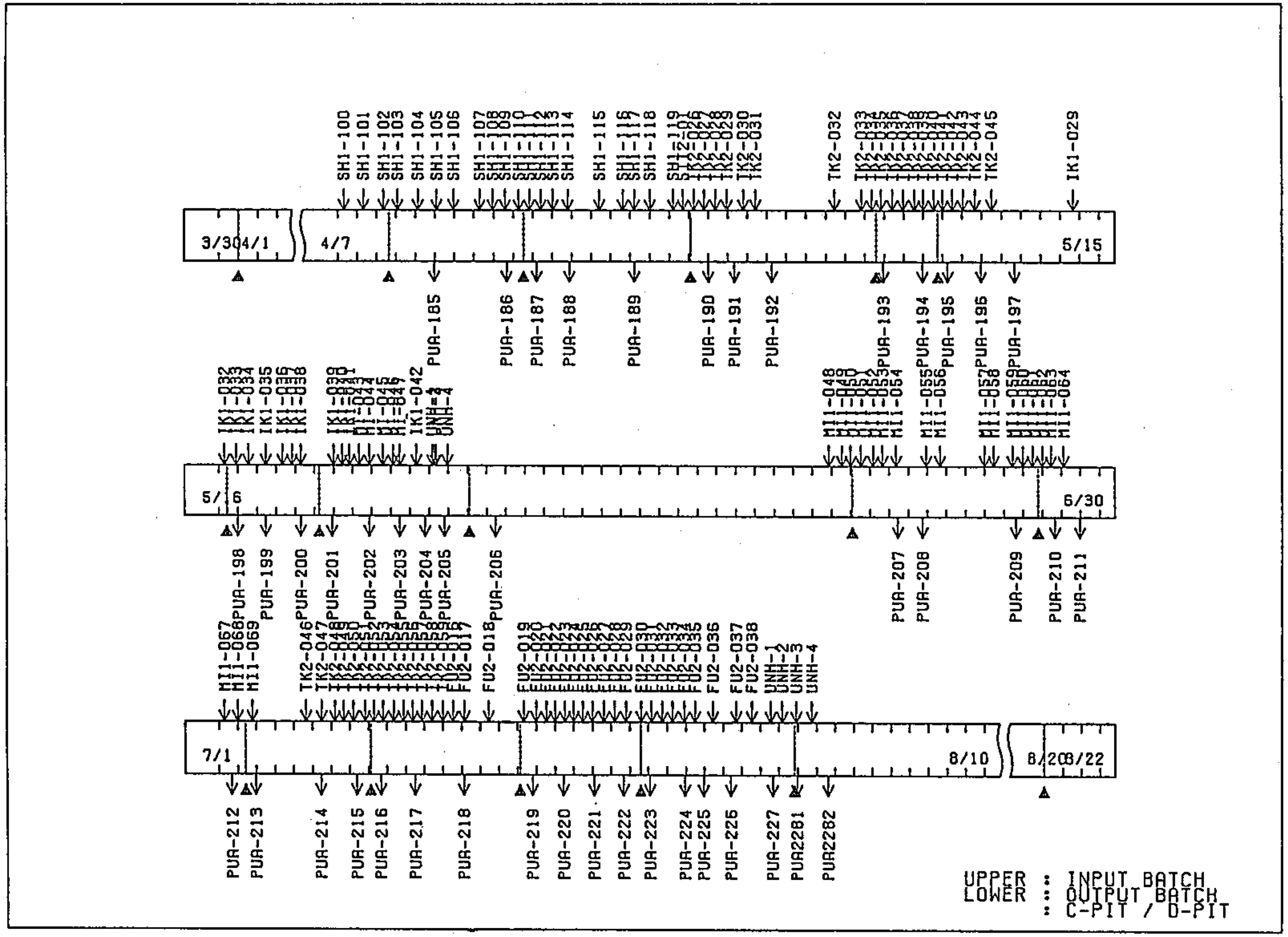


Fig. 4.8 Operation Chart for the 85-1 Campaign

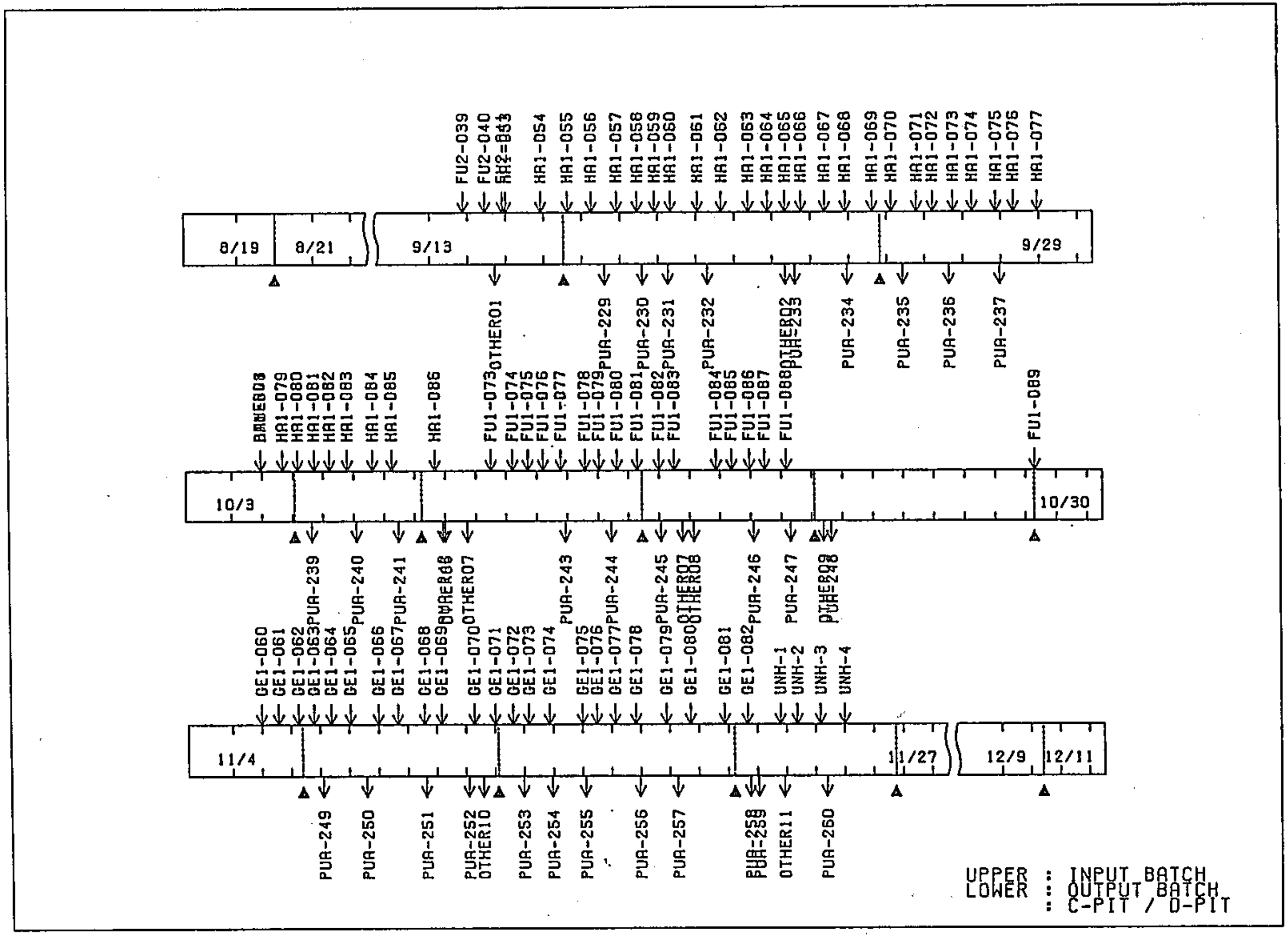


Fig. 4.9 Operation Chart for the 85-2 Campaign

4.4 Campaigns 85-2, "Daily" Material Balances

During the 85-2 campaign it was decided that an investigation based on daily material balances should be carried out. The balances would be based solely on process control data (see below), and would be used to develop better information regarding the magnitude and fluctuation of the unmeasured solvent extraction inventory. Since inventories can only be taken, and material balances prepared, when a product transfer measurement occurs, the following procedures were followed.

a. Every time the product evaporator was emptied (nominally every 24-36 hours) the volumes in the seven buffer tanks were measured. Since all seven tanks are instrumented with strip chart recorders, the volume measurements consisted of reading the strip chart record for the time in question.

b. The Pu concentration in each tank was assumed to be equal to the measured concentration at the time of the next (or in some cases the last previous) process control measurement. Since process control samples are normally taken at 8 hour intervals this measurement may not match the exact time of the in-process inventory, but it should not be further than 3-4 hours from the inventory time.

c. Process control samples are not taken from the buffer tank feeding the first extraction cycle, because the solution has just been analyzed in the immediately preceding input accountancy tank. For the "daily" material balance data, concentrations were calculated from the known concentration of the last batches transferred to the tank.

Table 4-5 presents the material balance data which was collected in this manner. The timing of the "daily" inventories can be derived from Figure 4.9 by noting that an inventory was calculated every time a product batch transfer occurred, identified by PUA numbers in the Figure. Since one of the objectives in doing the daily balances was to estimate the unmeasured solvent extraction system inventory, the CUMUF column in Table 4-5 omits the first and last periods that included cleanout inventory data.

Table 4-5

N.R.T. Accountancy Data, Campaigns 85-2 "Daily" Material Balance Data

(not adjusted for "unmeasured inventory," see Section 5.1)

(all data in grams)

No.	BI	I	O	EI	MUF	CUMUF
1	1944	13715	6	12870	2783	
2	12870	8522	5726	14743	923	923
3	14743	5840	6587	14377	-381	542
4	14377	6178	7379	13098	78	620
5	13098	7820	6907	13576	435	1058
6	13576	7883	6585	14883	-9	1049
7	14883	4785	6876	13340	-548	501
8	13340	5251	6200	12215	176	677
9	12215	7561	6257	13349	170	847
10	13349	7172	6476	13170	875	1722
11	13170	5990	6427	13862	-1129	593
12	13862	7324	6178	14497	511	1104
13	14497	4866	6172	12922	269	1373
14	12922	2483	6114	9431	-140	1233
15	9431	11931	7152	14333	-123	1110
16	14333	5134	6386	12846	235	1345
17	12846	7893	7016	14423	-700	645
18	14423	9633	7021	16314	721	1366
19	16314	5046	7058	14232	70	1436
20	14232	2745	6129	10596	252	1688
21	10596	193	7149	3870	-230	1458
22	3870	12021	0	15674	217	1675
23	15674	6279	6596	14253	1104	2779
24	14253	6253	6625	13960	-79	2700
25	13960	9179	6295	16807	37	2737
26	16807	6339	6846	15868	432	3169
27	15868	6705	6855	15194	524	3693
28	15194	3933	5805	13540	-218	3475
29	13540	9423	7391	15124	448	3923
30	15124	4077	6753	12768	-320	3603
31	12768	9618	7008	15578	-200	3403
32	15578	3748	6448	11851	1027	4430
33	11851	727	6372	9376	-3170	1260
34	9376	452	5784	4085	-41	1219
35	4085	0	3497	1019	-431	

5. The Field Test Data: II. Engineering Evaluation

5.1 "Unmeasured Inventory" Estimation

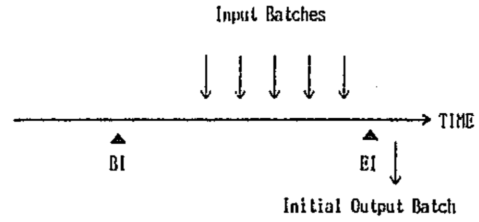
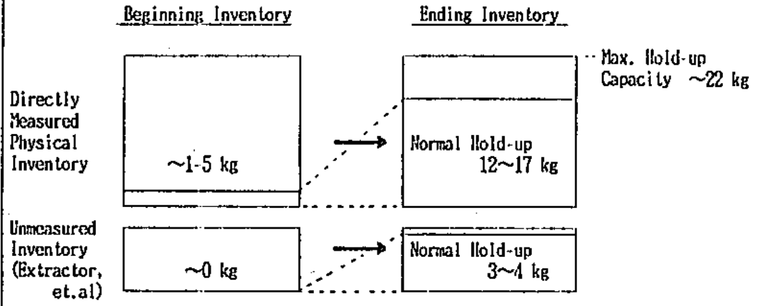
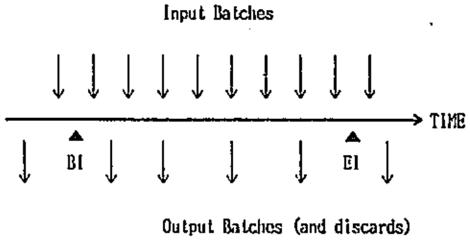
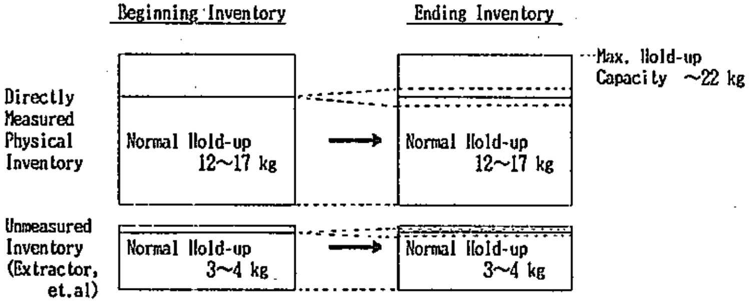
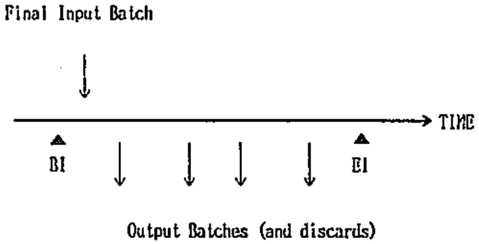
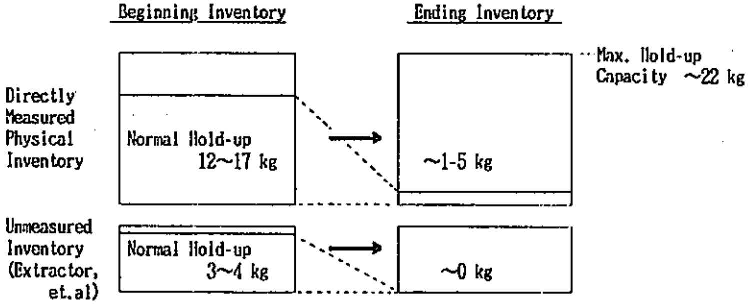
Inspection of the raw data for the C-1 and C-2 campaigns (or for any other campaign) will show that the observed MUF during the first material balance period is abnormally large, on the order of 3 - 4 kgs Pu, and that there is an offsetting MUF gain during the last material balance period. These result from the fact that the raw data includes only the measured portion of the in-process inventory.

During the first period the total quantity of plutonium in the solvent extraction systems (and in miscellaneous piping, small vessels, oxidation columns, etc.) appears as MUF. During the second and subsequent material balance period only the fluctuation in this unmeasured inventory (UMI) appears as MUF. When the plant is shut down for a cleanout physical inventory this UMI is reduced to zero (or nearly zero), and the previously omitted material appears as part of the measured material balance. This phenomenon is explained graphically in Figure 5.1. The same effect occasionally occurs within various campaigns when the plant is partially flushed between fuel types and an in-process inventory is taken during the flushing period.

In order to make any rational use of the raw near-real-time accountancy data, clearly it is necessary to make some adjustment for this unmeasured inventory. In most of the early evaluations an empirically estimated 4000 grams UMI was added to all in-process inventories. In this report the value of 4000 is retained for the C-1, C-2 and 81-1 campaigns (Section 5.2 explains why) and a value of 3200 grams Pu is used for all other campaigns. The paragraphs that follow review the work which led to the adoption of this revised value.

Fig. 5.1

Effect of Plant Operation Status on NRTA Data

Operation Status	Flow	In-process Inventory	MUF
I. Start Up 	Input ~Normal Process Hold-up (see next column) Output ~0		(positive MUF) ~Normal Hold-up of Unmeasured Portion + Measurement Error
II. Normal Operation 	Input ~Output (max~50kg-Pu at nominal operation) Output ~Input		(positive or negative MUF) ~Variation of Hold-up of Unmeasured Portion + Measurement Error
III. Flush Out 	Input ~0 Output ~Normal Process Hold-Up (see next		(negative MUF) ~Measurement Error − Normal Hold-up of Unmeasured Portion

For the 85-2 campaign PNC used the transient code MIXSET to calculate the complete inventory profile as a function of time. The result of this calculation is shown in Figure 5.2. Neglecting the "spike" at about 740 hours when the systems were flushed prior to a change from BWR to PWR fuel, the inventory appears to have fluctuated in the range of 3100 g Pu during the BWR part of the campaign, and then to have increased to about 3600 g Pu during the PWR part of the campaign. The increase in going from BWR to PWR is consistent with the increased burnup and Pu content of the PWR fuel.

PNC also used this MIXSET data to calculate the solvent extraction system inventory at the time of each of the 34 "daily" in-Process inventories discussed in Section 4.4. The mean value for the first 21 data points (prior to the change in fuel types) is 3128 ± 608 g Pu. For the ten data points collected during PWR fuel processing (neglecting the data point during the flushout and two data points at the end of the campaign when the system was being rinsed for cleanout physical inventory taking) the mean value is 3602 ± 199 g Pu. For the data set as a whole (omitting the three points noted) the mean value is 3281 ± 556.

In addition PNC and JAERI separately used multiple linear regression to develop a simplified inventory relationship. The models differ slightly; the JAERI model was of the form $H = aX_1 + bX_2 + cX_3$,

where H is the total solvent extraction system inventory (in grams Pu) and X_1, X_2 and X_3 represent the Pu concentration in the buffer tank feeding the first, second and third cycles of solvent extraction.

Using the difference between inputs and outputs in the 85-2 campaign "daily" data as an estimate of the solvent extraction system inventory, JAERI obtained the relationship

$$H = 701.9\ X_1 + 2684.3\ X_2 + 842.9\ X_3 \qquad \text{(eq. 5.1)}$$

During the 85-2 campaign the mean value of X_1 was 1.384 g Pu/l, the mean value of X_2 was 0.4222 g Pu/l, and the mean value of X_3 was 1.3444 g Pu/l, yielding a mean value for the total inventory of 3238 ± 428 g Pu.

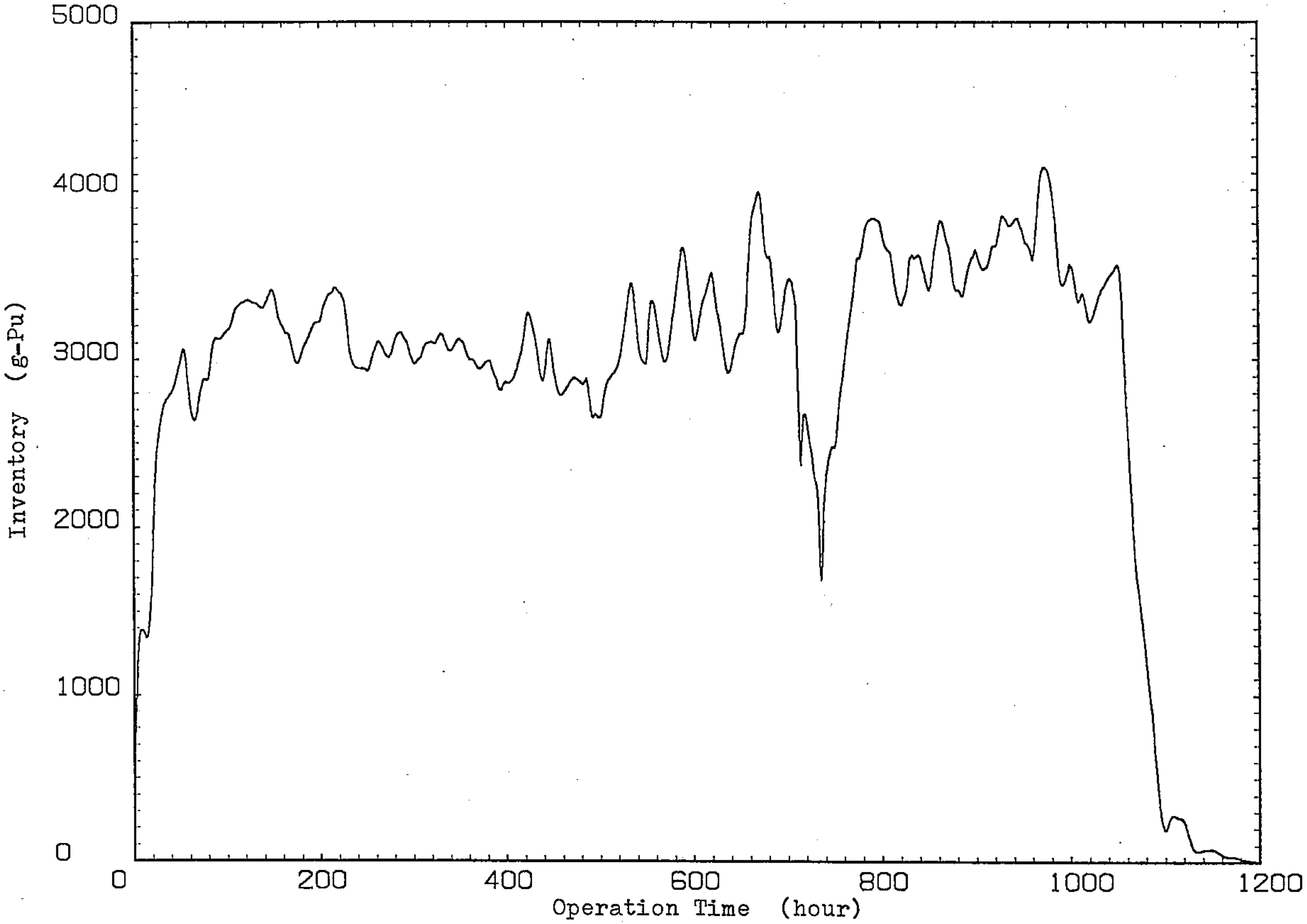


Fig. 5.2 Transient of Pu Inventory in Mixer Settlers through the 85-2 Campaign

PNC used a model which also allowed a constant term, and used the MIXSET values for the solvent extraction inventories. They obtained the relationship

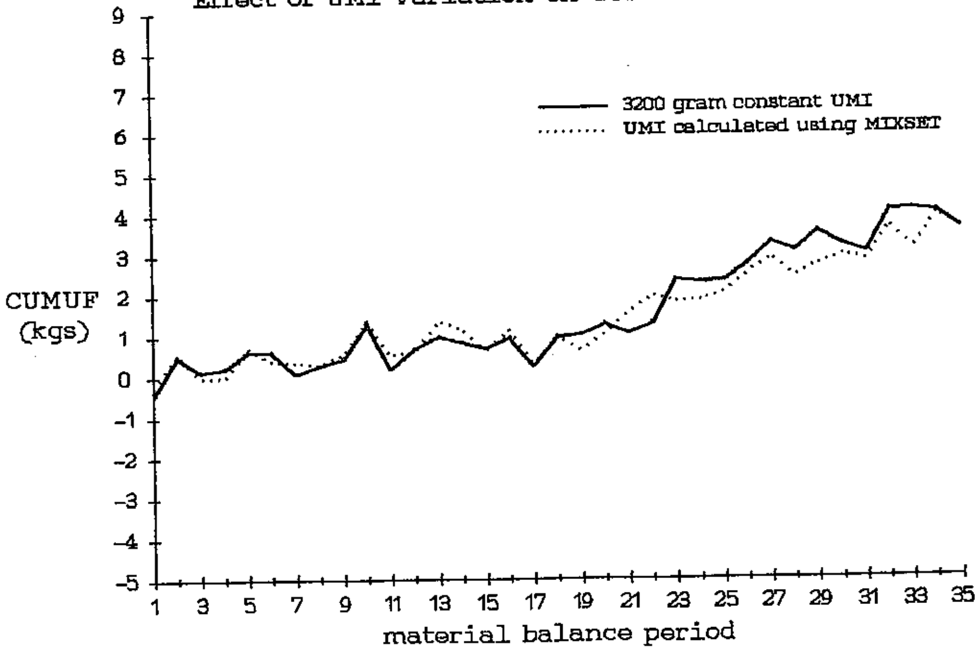
$$H = 20\ X_1 + 4366\ X_2 + 732\ X_3 + 390 \qquad \text{(eq. 5.2)}$$

Using the same values for X_1, X_2 and X_3 as JAERI, the PNC relationship yields a mean inventory value of 3244 ± 237 g Pu, in excellent agreement with JAERI.

It is worth noting in passing that the strong dependence of the total inventory on the concentration of the aqueous feed to the second cycle is not unexpected, because in the PNC flow sheet partition occurs in the second cycle.

The various calculations reported above have estimated standard deviations in the range of ± 15% of the solvent extraction system inventory. However, the solvent extraction system inventory itself is no more than 25% of the total in-process inventory, and the effect of this variation on near-real-time accountancy in general appears to be unimportant. This can be seen graphically in Figure 5.3, which compares the "daily" CUMUF data for the 85-2 campaign with and without an adjustment for variations in the solvent extraction system inventory.

Figure 5.3
Effect of UMI Variation on CUMUF Data



5.2 Campaigns C-1, C-2 and 81-1

As previously noted, the first near-real-time accountancy data was collected in 1980, during the C-1 and C-2 campaigns. Several lessons were learned in this initial field testing effort, The first was the importance of the plutonium inventory in the solvent extraction system. In the evaluations performed in 1981, the unmeasured inventory was estimated to be about 4000 grams Pu, and Table 5-1 below shows the result of this adjustment. This adjustment applies to all in-process physical inventories, but not to the clean-out physical inventories, indicated in Table 5.1 with an asterisk.

The estimate of 4000 g Pu, admittedly based primarily on the magnitude of the first weekly MUF in the C-1 campaign, appears to be remarkably close to the nominal average value when the process is operating normally, for the campaigns in question. It is not a good estimate when the process is being flushed between fuel types.

Table 5-1

N.R.T. Accountancy Data, Campaigns C-1, C-2, and 81-1
(including 4000 grams Pu for "Unmeasured Inventory")
(all data in grams)

No.	BI	I	O	EI	MUF	CUMUF
1	7662*	6611	5	14433	-165	-165
2	14433	24708	25933	14621	-1413	-1578
3	14621	24772	24507	15594	-708	-2286
4	15594	27232	25214	15228	2384	98
5	15228	20392	19518	18050	-1948	-1850
6	18050	25163	27545	17179	-1511	-3361
7	17179	16471	31657	2416*	-423	-3784
8	2416*	12997	10	16268	-865	-4649
9	16268	17390	19101	14314	243	-4406
10	14314	30930	33073	13984	-1813	-6219
11	13984	21983	19794	15245	928	-5291
12	15245	23802	27344	15225	-3522	-8813
13	15225	13934	12449	16018	692	-8121
14	16018	10396	13377	12456	581	-7540
15	12456	194	6476	6289*	-115	-7655
16	6289*	25060	0	30085	1264	-6391
17	30085	0	5154	26390	-1459	-7850
18	26390	6838	14371	18561	296	-7554
19	18561	3646	5915	19360	-3068	-10622
20	19360	117215	136098	2160*	-1683	-12305

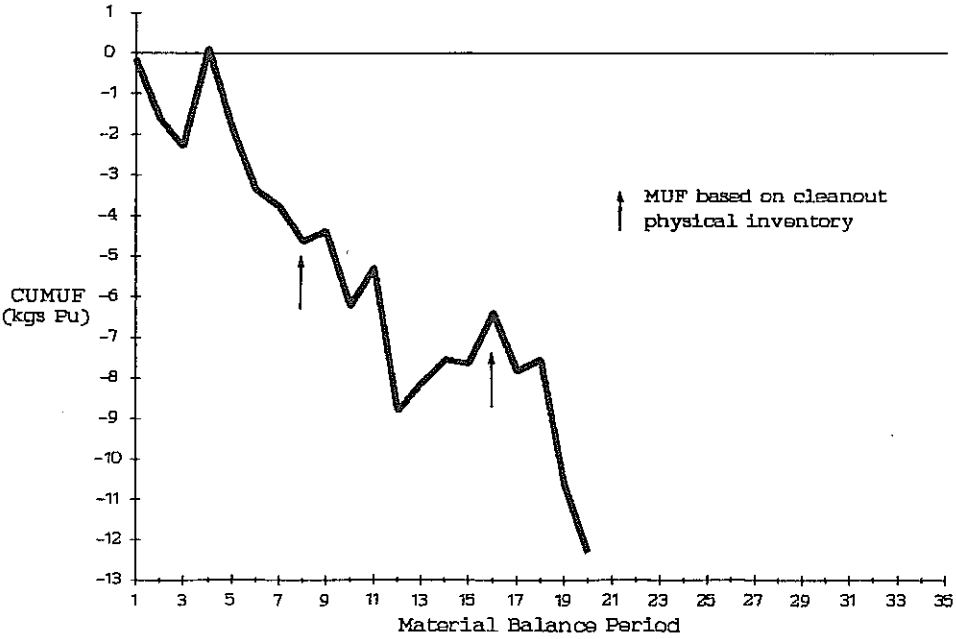
* cleanout physical inventory data

The careful reader will have noted the difference between the estimate of 4000 grams given here and the estimate of 3200 grams developed in Section 5.1. When it was first noted that 4000 grams appeared to be an over-estimate for the 81-2, 82-1 and 82-2 campaigns, it was assumed that PNC had modified its flow sheet in a manner which reduced the solvent extraction inventory. Investigation revealed, however, that the cleanout inventory data for the C-1 and C-2 campaigns included about 500 grams Pu in retained waste. The remainder of the discrepancy is partially explained by the processing of high burnup PWR fuel; part of the discrepancy remains unexplained. Since the C-1 and C-2 campaign data has already been published with a 4000 gram UMI adjustment, the authors have chosen to leave the UMI estimate at 4000 grams, and to use the better estimate developed in Section 5.1 only for the later campaigns.

Another lesson learned early in the evaluation of the first field test data was that there was no reason to terminate a graph (or a statistical calculation) simply because a cleanout physical inventory occurred. The initial graphs of the C-1 and C-2 campaigns separately (not shown here) included too little data for meaningful evaluation. Combined, as shown in Figure 5.4, the field test data clearly show that a positive measurement bias (or a combination of biases having a net positive effect) exists. The combined data also justifies the combination of data across cleanout inventories. One cannot tell by inspection which intermediate data points (marked on the graph with arrows) represent cleanout physical inventories.

Figure 5.4

Campaign C-1, C-2, 81-1 (Adj. Data)



One does not need complex statistical calculations to conclude that the CUMUF data graphed in Figure 5.4 show a definite trend in the direction of an MUF gain. Since negative diversion is not defined, this trend must have been caused by measurement bias. It is immaterial whether the bias is (or the biases are) within estimated systematic measurement variances. The magnitude of the bias, calculated on a Pu throughput basis, is 2.8%.

During the early part of 1981 PNC undertook an intensive investigation of a wide variety of possible measurement errors. This investigation has been separately reported, see [5,6]. In summary, the investigation showed that:

a) The product accountancy tank, a reinforced thin-wall slab design, buckled inward when filled with hot solutions, causing product measurements to be biased high.

b) Heel measurements performed on the product accountancy tank immediately after each transfer inadequately reflected the extent to which solution transfer lines drain back into the accountancy tank after a transfer.

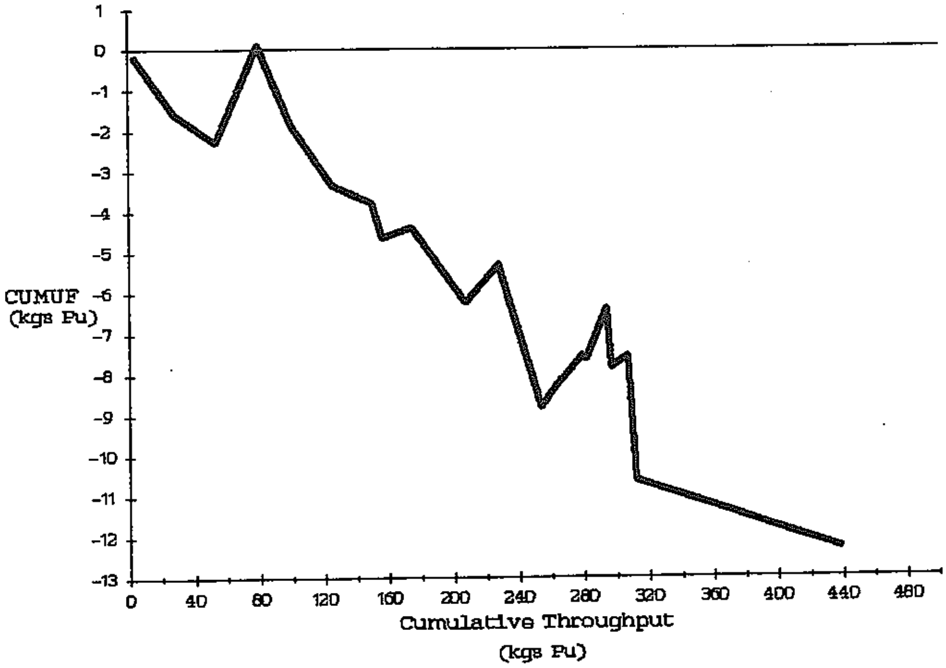
Unfortunately, the estimated magnitude of these two measurement biases does not fully account for the observed trend. The estimates themselves were subject to considerable uncertainty, and it was not unreasonable to believe that these two biases could account for the observed effect. In any case, as shown in Section 5.3, the bias was effectively eliminated in the 81-2, 82-1 and 82-2 data.

The C-1 and C-2 campaigns were still in the nature of shakedown campaigns for PNC, and process operations were correspondingly uneven. Since near-real-time material balances could only be prepared when the product evaporator was emptied, and since at times the evaporator was not emptied for periods considerably longer than the desired seven days, the intervals between balances varied widely.

Initially this problem was ignored. Figure 5.4 is as close to a straight line as one has any right to expect; replotting the data in a different form simply did not occur to the authors. In the course of the total re-evaluation of all data leading to the preparation of this report, however, it was realized that errors generally fall into two basic categories; those that are independent of the length of the material balance period (e.g., inventory measurement error, inventory clerical error, etc.) and those that are logically a function of throughput (e.g. flow measurement error).

This led to a suggestion that n.r.t. accountancy data should be plotted and evaluated as a function of throughput, not as a function of material balance period. Figure 5.5 shows the C-1, C-2 and 81-1 campaign data as a function of (Pu) throughput, not as a function of material balance period. Whether balances should be taken at intervals which are defined in terms of throughput is a question which is still open. (In passing, the authors note that the same conclusion was reached independently in the Dounreay field test work.)

Figure 5.5
Campaign C-1, C-2, 81-1 (Adj. Data)



Visually, Figure 5.5 does not present quite the straight line observed in Figure 5.4. However, this is largely an artifact of the graphs and of the previously noted fact that no in-process inventories were taken during the latter part of the 81-1 campaign. As discussed below, the correlation is if anything slightly better when the data are plotted on a throughout basis.

Evaluation of materials accountancy data in terms of historical MUF data has been proposed a number of times, but rarely wins much support because the available historical data usually is limited both in quantity and in defensible validity. The PNC field test work, however, includes some 75 historical data points grouped into three sets, each of which arguably represents consistent plant operation and measurement conditions.

A linear regression analysis of the C-1, C-2 and 81-1 campaigns, after adjusting for an assumed 4000 g UMI, shows that the data can be represented by a straight line defined by:

$$\text{CUMUF} = 101 \text{ g Pu} - 532 \text{ g (m.b.p.)} \qquad \text{(eq. 5.3)}$$

The uncertainty in this estimate is given by

$$\sigma_{s.e} = \pm\ 1267 \text{g Pu}$$

Or, on the more logical basis of cumulative throughput, the linear regression equation is:

$$\text{CUMUF} = 218 \text{ g Pu} - 0.0283 \text{ (cum. throughput)} \qquad \text{(eq. 5.4)}$$

The uncertainty in this estimate is:

$$\sigma_{s.e.} = \pm\ 1087 \text{ g Pu}$$

The correlation coefficient on a material balance period basis is 0.931; on a cumulative throughput basis it is 0.950. Both values are very high, confirming the visual interpretation that a throughput-related measurement bias is the major factor affecting the data.

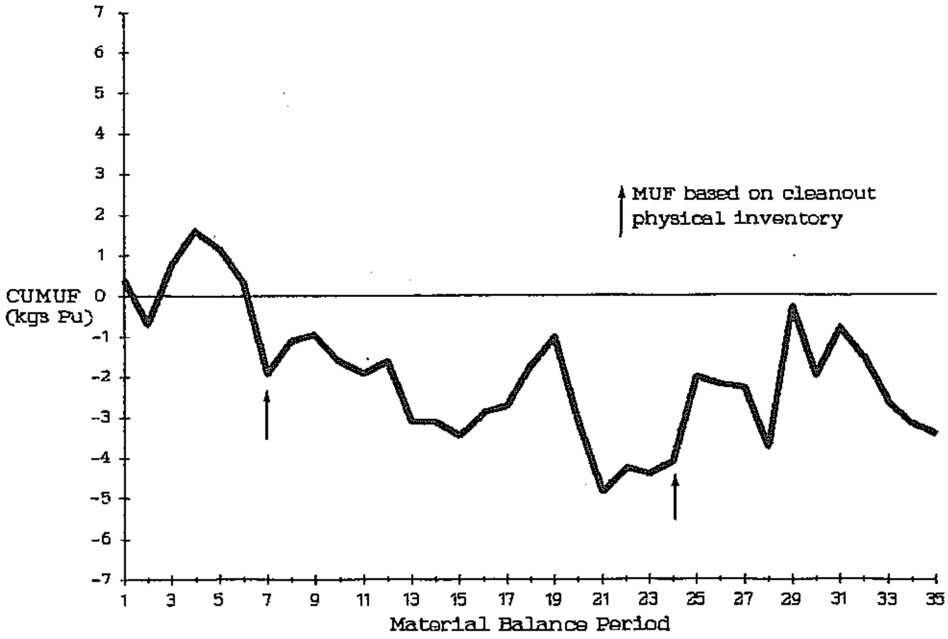
5.3 Campaigns 81-2, 82-1 and 82-2

Careful inspection of Table 4-3 will reveal a "spike" at the 15th material balance, an MUF gain which is almost twice as large as any other MUF in the series and which is matched by an almost identical MUF loss in period 16. Such spikes are characteristic of inventory errors, and an investigation was conducted in an effort to identify and correct the error. It was found that the contents of the product accountancy tank, which were off-spec., had already been transferred to the recycle tank at the time of the in-process inventory, and were recorded as being in both places. Table 5-2 presents corrected material balance data for the 81-2, 82-1 and 82-2 campaigns.

Although in the present instance data evaluation did not occur until several years after data collection, it is worth noting that the ability to recognize probable data errors and make appropriate corrections is one of the strengths of near-real-time accountancy. Errors of this nature are inevitable, but timely data evaluation for near-real-time accountancy data greatly facilitates data correction. In a routine situation, the inventory error in period 15 probably would have been noted before the offsetting MUF in period 16 made it obvious, but in all events it would have been noted and corrected by period 17 or 18.

Figures 5.6 and 5.7 show the corrected data graphically as a function of material balance period and as a function of cumulative throughput, again defined as input plus output divided by two.

Figure 5.6
Campaign 81-2, 82-1, 82-2 (adjusted)

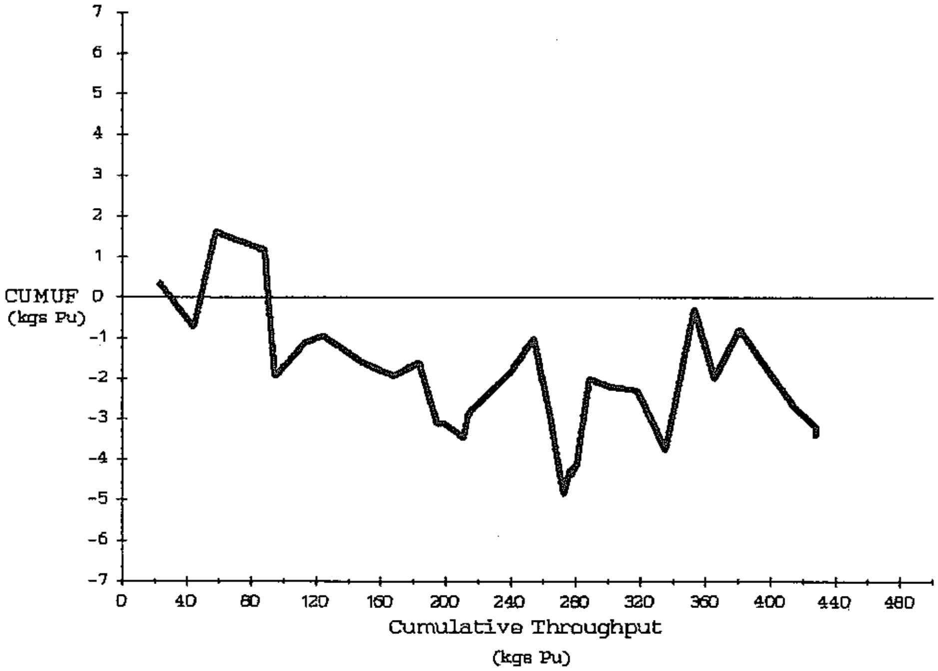


As noted, serious evaluation of the 81-2, 82-1 and 82-2 campaign data did not occur until 1985, during the course of preparing this report. One could ask several questions, for example, is there a negative bias extending through about period 23, and if so, what change occurred to reverse the bias during the remaining material balance periods. Alternatively, one could note that if the first six periods are ignored the remaining data show little visual evidence of bias, especially if one could find an explanation for the large MUF gains at periods 20 and 21.

In view of the long time interval between data collection and data evaluation, the authors have chosen not to pursue the investigation of this dataset in detail. The plant was shut down at the end of these campaigns and remained shut down for most of the following two years, during which period all measurements of interest were recalibrated. The data are at least approximately within combined random and systematic measurement error variances, so no detailed investigation or explanation is required.

Figure 5.7

Campaign 81-2, 82-1, 82-2 (adjusted)



It is interesting, however, to compare linear regression estimates for the 81-2, 82-1 and 82-2 campaigns with the previous estimates for the C-1, C-2 and 81-1 campaigns (and with the 85-1 and 85-2 campaigns, given in the next Section). On a material balance period basis the regression equation is:

$$\text{CUMUF} = -\ 439 \text{ g Pu} - 85 \text{ g (m.b.p.)} \qquad \text{(eq. 5.5)}$$

The uncertainty in this estimate is given by

$$\sigma_{s.e} = \pm\ 1397 \text{ g Pu}$$

Or, on the more logical basis of cumulative throughput, the linear regression equation is:

$$\text{CUMUF} = -\ 208 \text{ g Pu} - 0.0076 \text{ (cum. throughput)} \qquad \text{(eq. 5.6)}$$

The uncertainty in this estimate is:

$$\sigma_{s.e.} = \pm\ 1380 \text{ g Pu}$$

Consistent with the visual interpretation of figures 5.6 and 5.7, these equations show that the measurement system, although perhaps not completely bias free, is much closer to that elusive goal than during the earlier campaigns. Correlation coefficients are in the range of 0.54, indicating that there is some correlation with throughput but that at least two-thirds of the variation in the data must be explained in terms of factors other than flow measurement bias.

Table 5.2

N.R.T. Accountancy Data, Campaigns 81-2, 82-1 and 82-2

(corrected for inventory error at period 15, see text)

(adjusted for 3200 gram UMI)

(all data in grams Pu)

No.	BI	I	O	EI	MUF	CUMUF
1	2160	32899	12621	22062	376	376
2	22062	14669	28728	9067	-1064	-688
3	9067	13103	3764	16975	1431	743
4	16975	4816	5185	15736	870	1613
5	15736	23880	36264	3805	-453	1160
6	3805	0	3124	1508	-827	333
7	1508	9105	0	12872	-2259	-1926
8	12872	19597	16967	14685	817	-1109
9	14685	11765	12000	14292	158	-951
10	14292	25411	24335	16034	-666	-1617
11	16034	19458	18327	17457	-292	-1909
12	17457	14183	15903	15454	283	-1626
13	15454	9916	12685	14168	-1483	-3109
14	14168	6452	4359	16275	-14	-3123
15	16275	5204	17562	4243	-326	-3449
16	4243	4862	0	8554	551	-2898
17	8554	5674	0	14069	159	-2739
18	14069	26499	23999	15547	1022	-1717
19	15547	13287	11167	16950	717	-1000
20	16950	9840	11703	17142	-2055	-3055
21	17142	4083	11578	11421	-1774	-4829
22	11421	0	7237	3598	586	-4243
23	3598	0	0	3753	-155	-4398
24	3753	8045	0	11501	297	-4101
25	11501	11425	4176	16650	2100	-2001
26	16650	15218	12164	19887	-183	-2184
27	19887	15973	17206	18753	-99	-2283
28	18753	18589	16254	21552	-1464	-3747
29	22552	16044	17747	17388	3461	-286
30	17388	13418	12519	19988	-1701	-1987
31	19988	12716	17609	13896	1199	-788
32	13896	12820	12339	15100	-723	-1511
33	15100	20489	19141	17554	-1106	-2617
34	17554	6485	23058	1524	-543	-3160
35	1524	0	0	1789	-265	-3425

5.4 Campaigns 85-1B, 85-1C and 85-2

Table 5-3 presents the material balance data for the 85-1B, 85-1C and 85-2 campaigns. All inventories except those associated with the cleanouts after the 85-1C campaign and the 85-2 campaign have been adjusted by 3200 grams Pu for the unmeasured solvent extraction systems. Although one might wish for a smaller UMI adjustment for periods such as no. 8 or no. 15, where 3200 grams appears to be too large, the constant value was used. No other data corrections or adjustments appear to be necessary. Figures 5.8 and 5.9 show the adjusted data graphically.

Both Figures 5.8 and 5.9 suggest the existence of a small bias in the direction of MUF loss, on the order of 1.1% of throughput. This is the reverse of the biases observed during 1980 and 1981, and as noted the 1982 data suggested the absence of significant measurement bias. As of the date of this report no investigation of this bias has been conducted, and no explanation is offered.

Another significant observation is that the spread of the data has been reduced. On a material balance period basis the linear regression equation is:

$$\text{CUMUF} = -\,630 \text{ g Pu} + 215 \text{ g (m.b.p.)} \qquad \text{(eq. 5.7)}$$

The uncertainty in this estimate is given by

$$\sigma_{s.e} = \pm\, 865 \text{ g Pu}$$

Or, on the more logical basis of cumulative throughput, the linear regression equation is:

$$\text{CUMUF} = -\,363 \text{ g Pu} + 0.011 \text{ (cum. throughput)} \qquad \text{(eq. 5.8)}$$

The uncertainty in this estimate is:

$$\sigma_{s.e.} = \pm\, 900 \text{ g Pu}$$

For the 1980-81 data the ability to predict CUMUF from a knowledge of throughput was ± 1087 g Pu (one s.d.). For the 1981-82 data it was ± 1380 g Pu. Although no serious investigation was performed, the explanation for this reduction probably lies in a reduction of random inventory measurement error variances as a result of the long shutdown period during 1983-84. Correlation coefficients are again high, on the order of 0.9 for either material balance period or throughput.

Figure 5.8

Campaigns 85-1B, 85-1C and 85-2 (adjusted)

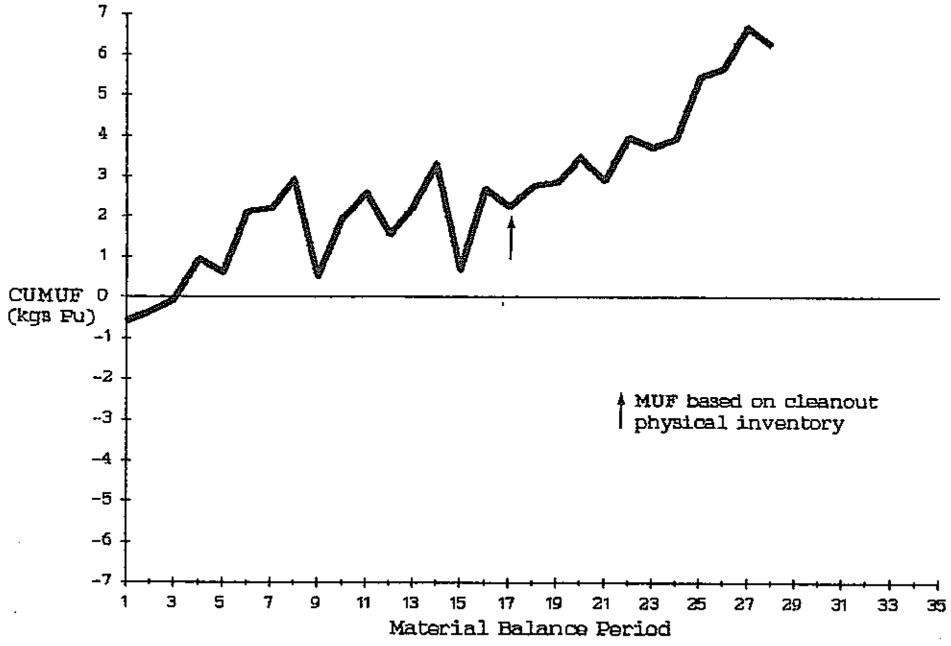


Figure 5.9

Campaigns 85-1B, 85-1C and 85-2 (adjusted)

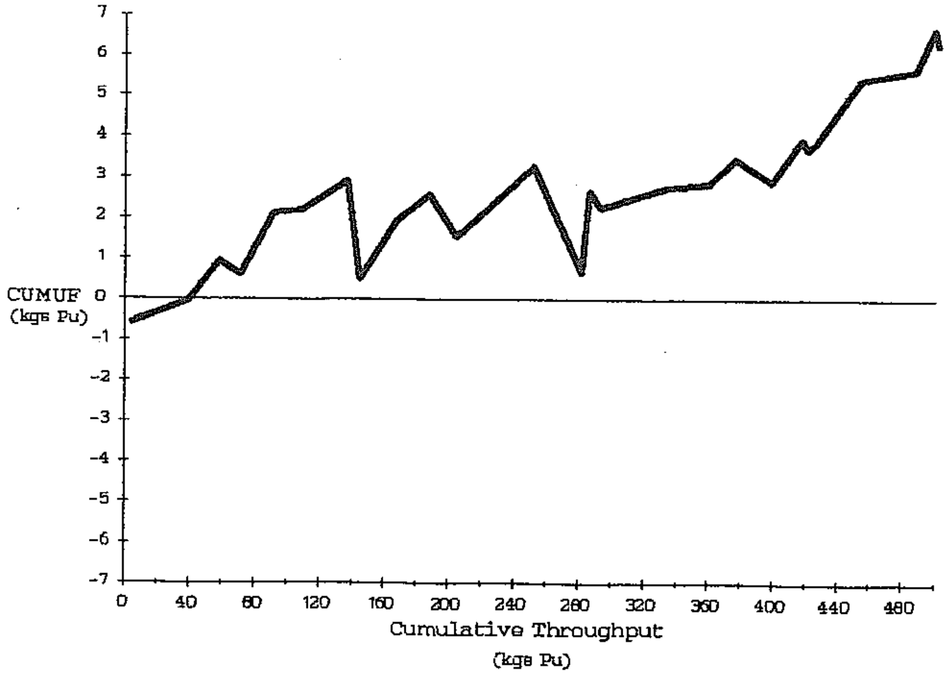


Table 5-3

N.R.T. Accountancy Data, Campaigns 85-1B, 85-1C and 85-2

(adjusted for 3200 grams UMI, see Section 5.1)

(all data in grams)

No.	BI	I	O	EI	MUF	CUMUF
1	1002*	6994	0	8588	-592	-592
2	8588	18482	12264	14566	240	-352
3	14566	19769	18898	15151	286	-66
4	15151	19576	18820	14889	1018	952
5	14889	13251	13239	15238	-337	615
6	15238	21826	18884	16697	1483	2098
7	16697	16643	19022	14229	89	2187
8	14229	24425	31804	6130	720	2907
9	6130	8493	6051	10993	-2421	486
10	10993	27958	19879	17590	1482	1968
11	17590	18723	19697	15996	620	2588
12	15996	15557	18516	14076	-1039	1549
13	14076	19296	19094	13589	689	2238
14	13589	29479	26463	15580	1025	3263
15	15580	27255	32562	12867	-2594	669
16	12867	13	8943	1944*	1993	2662
17	1944*	13715	6	16070	-417	2245
18	16070	41028	40059	16540	499	2744
19	16540	25973	25359	17062	92	2836
20	17062	14672	18464	12631	639	3475
21	12631	24958	20554	17622	-587	2888
22	17622	17424	20208	13796	1042	3930
23	13796	193	7149	7070	-230	3700
24	7070	12021	0	18874	217	3917
25	18874	28050	26364	19068	1492	5409
26	19068	33756	33813	18778	233	5642
27	18778	4926	18604	4085*	1015	6657
28	4085*	0	3497	1018*	-430	6227

* cleanout physical inventory data

5.5 Campaign 85-2, "Daily" Material Balances

As noted in Sections 4.4 and 5.1, "daily" material balances were prepared during the 85-2 campaign. The primary objective of these daily material balances was better estimation of the solvent extraction system inventory, but the balances also provide an interesting comparison with the weekly data. The data given in Section 4.4 require no adjustment, because for this exercise the beginning and ending points based on cleanout physical were omitted. (This is also the reason why the total campaign MUF does not agree with the campaign MUF derivable from Table 5.3.) The data are shown graphically in Figure 5.3, in Section 5.1.

5.6 Running Book Inventory

On various occasions it has been suggested that it really was not necessary to measure or estimate the in-process inventory, all that was necessary was for the inspector to confirm that the book inventory, defined as the physical inventory at the beginning of the campaign plus the net of subsequent input and output transfers, was within pre-determined limits. This suggestion has variously been named "running book inventory" or "input - output analysis". This section analyzes the PNC-Tokai data in terms of the running book inventory concept.

Figures 5.10 through 5.15 show, for campaigns C-1 through 82-2, the running book inventory for the process MBA at PNC-Tokai as a function of material balance period, where here material balance period refers to the period between two product transfer measurements. The running book inventory for any given period is calculated as the book inventory at the beginning of the period plus all measured inputs, minus all measured discards, and minus the product transfer which ends the period. Defining the period in this way, as a function of specific process events, should be more meaningful than any arbitrary definition based on clock time. Data are not combined across campaigns as they have been in all previous discussions; there does not appear to be any easy way to carry a running book inventory analysis across a cleanout physical inventory boundary.

It is a true statement that a would-be divertor cannot divert, in an abrupt diversion mode, more material than is physically present in the area in question. Indeed diversion of any significant fraction, perhaps 1/4 or 1/3 of the material physically present, would seriously disrupt process operations. It is also a true statement that if the plant operator attempts to continue normal operations in a process area from which diversion has occurred, then the apparent book inventory in that process area will necessarily increase. To a first approximation the magnitude of the increase will equal the total amount diverted.

Advocates of the running book inventory approach usually suggest that the operator, State and inspectorate should agree on limits within which the book inventory normally should lie. So long as the apparent book inventory remains within these limits no action is taken. If the apparent book inventory goes outside the defined limits, some investigative action is called for.

For the PNC-Tokai plant the smallest limit which would not have been exceeded at least once during the field test work is 27 kgs Pu. This limit has never been exceeded, although it was closely approached (26+ kgs) at the beginning of the 81-1 campaign. A limit of 16 kgs would have been satisfactory for the C-1, C-2 and 81-2 campaigns, and even a limit of 12 kgs would have been satisfactory for the 82-1 campaign. During the 82-2 campaign, however, the running book inventory was larger than 16 kgs for 15 of the 27 periods, including one sustained sequence of 8 periods. Any limit smaller than 27 kgs would have led to the detection of at least one anomaly requiring subsequent investigation.

What conclusions can one draw from the observation that the apparent book inventory in the process area at PNC-Tokai has never exceeded 27 kgs Pu? First, one can be certain that an abrupt diversion of 27 kgs Pu never occurred. However, the plant operator presumably knows, or could know if he wanted to, why some campaigns were completed within lower running book inventory limits, and could duplicate those conditions when necessary to conceal an abrupt diversion. Thus as much as 11 kgs could have been diverted during the C-1, C-2 or 81-2 campaigns without exceeding the 27 kg limit, and as much as 15 kgs could have been diverted during the 82-1 campaign without exceeding the 27 kg limit.

If the desired detection goal is 8 kgs Pu, these numbers do not sound too bad, except that the PNC-Tokai plant is rated at a throughput of 210 t/a. A 1000 t/a plant presumably would have an inventory limit about five times larger, or in the range of 125 kgs Pu, and presumably could divert close to half that limit by manipulating the burnup level of the fuel processed or other flow sheet paramters not controlled by inspectors.

What about protracted diversion? In theory protracted diversion should lead to a gradual increase in the magnitude of the running book inventory, and this gradual increase is at least theoretically detectable through sequential data analysis techniques. There is considerable scatter in the running book inventory graphs, however, and detection of a pattern would be subject to considerable statistical uncertainty. The reader should recall that the CUMUF graphs shown earlier in this Chapter clearly showed a measurement bias equal to 2.8% of throughput in the C-1 and C-2 campaigns, and should ask whether this bias is discernible in Figures 5.10 and 5.11.

In the event the agreed inventory limit is exceeded, the inspectorate should undertake an investigation to determine the cause. In the ultimate, if the anomaly cannot be resolved in any other manner, the inspectorate logically would ask the facility to shut down for a special cleanout physical inventory. This is a serious request, one not to be made lightly. Should such a request be made, and should the State and facility agree to a special cleanout physical inventory, the inspectorate's ability to decide whether diversion in fact had occurred would be determined by the combined random and systematic error variances for all measurements subsequent to the last prior cleanout physical inventory. This is exactly the uncertainty, however, which has been termed unacceptably large in terms of future large reprocessing plants, and which has provided the primary motivation for the extensive research efforts on safeguards for reprocessing over the last decade.

Fig. 5.10

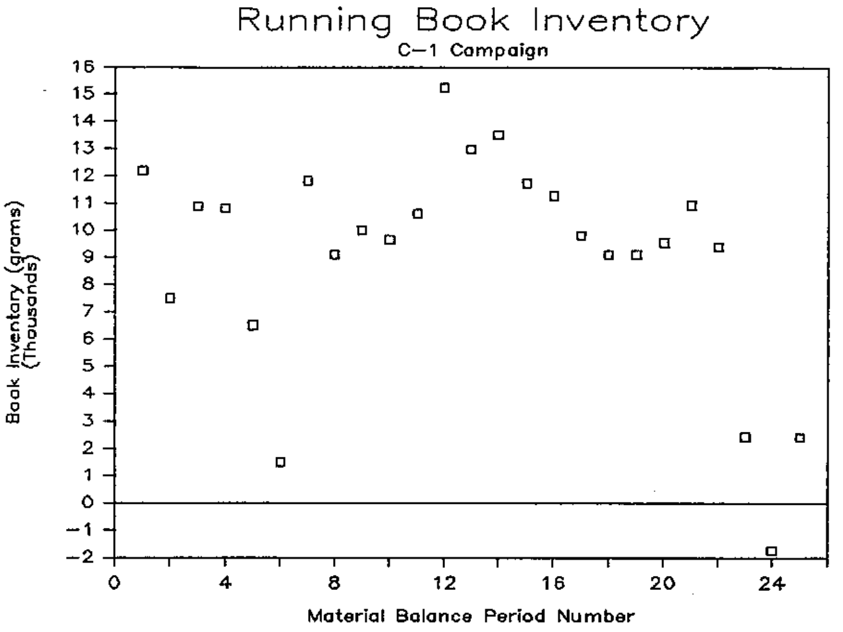


Fig. 5.11

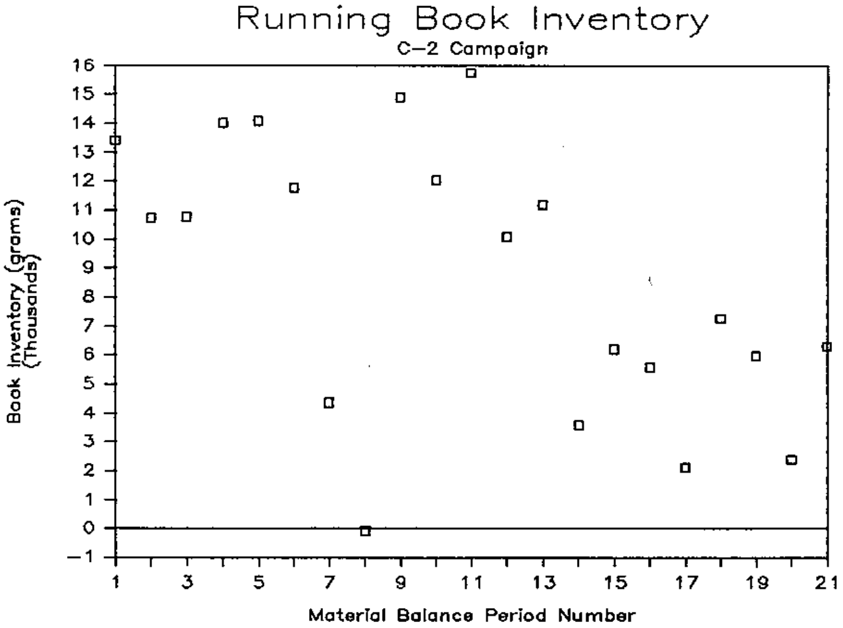


Fig. 5.12

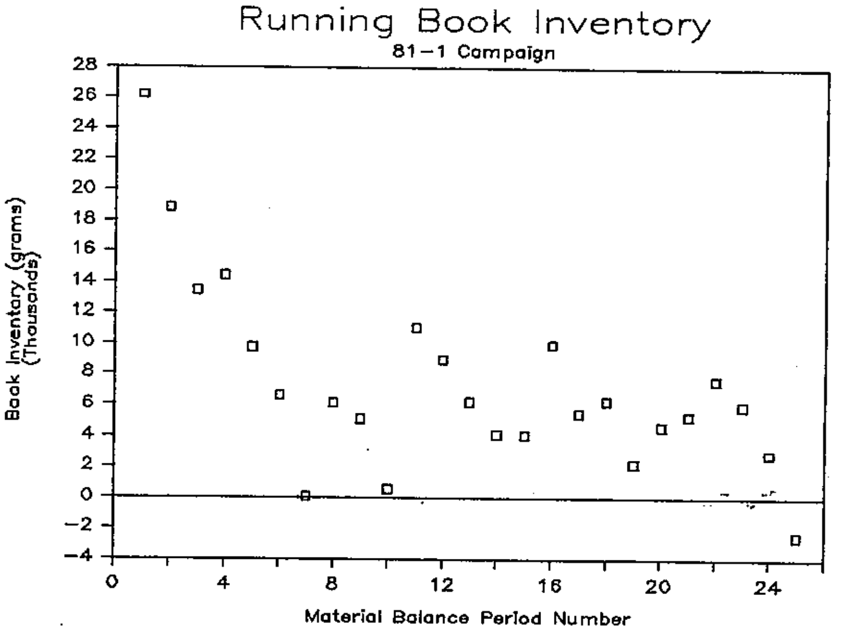


Fig. 5.13

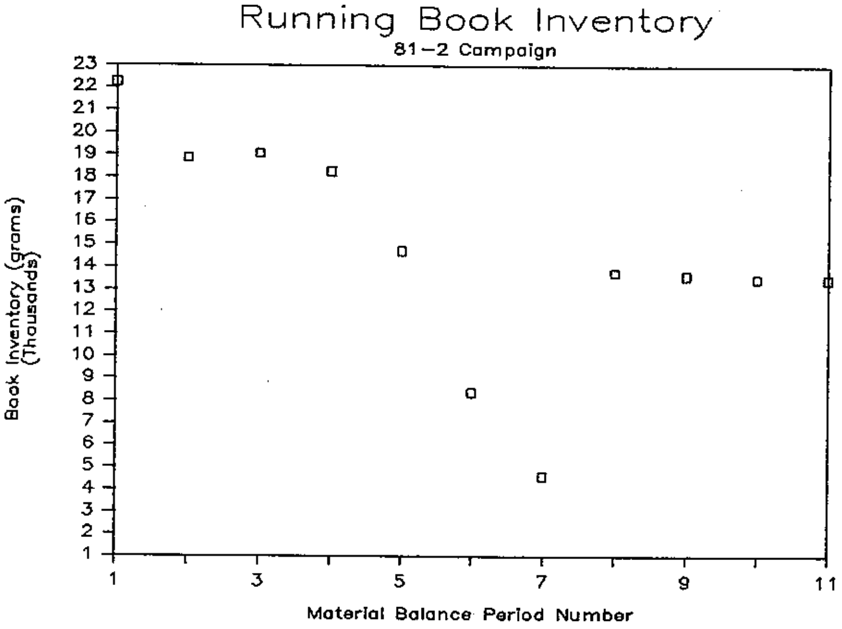


Fig. 5.14

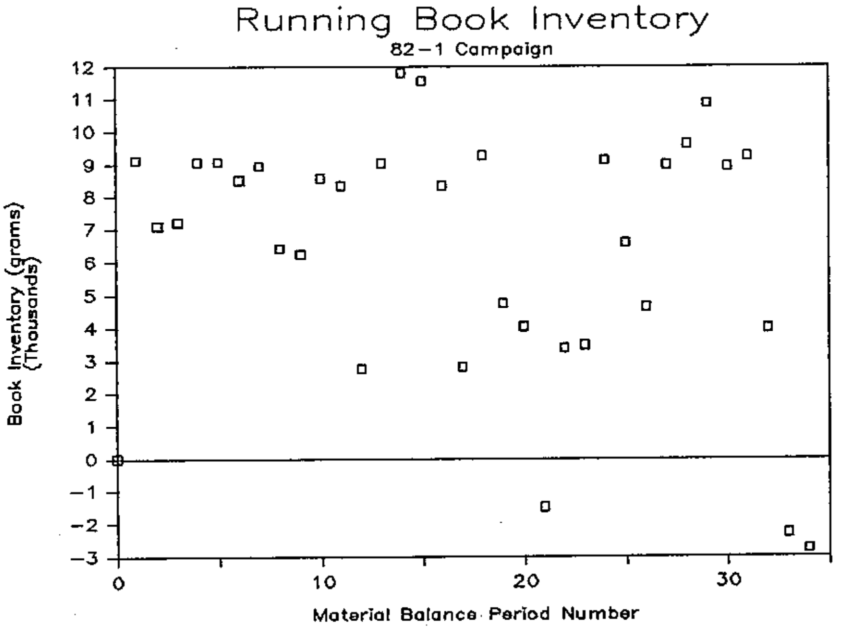
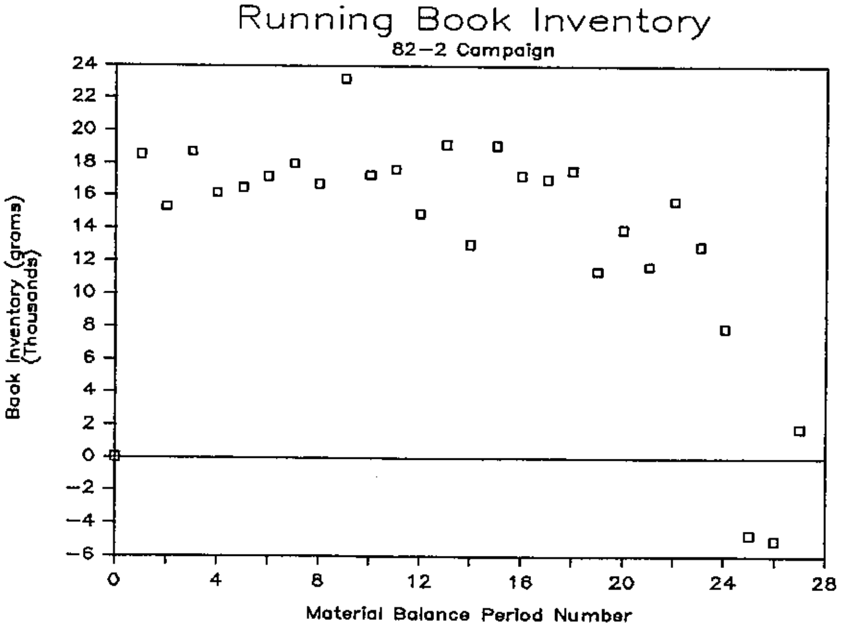


Fig. 5.15



6. Discussion

6.1 Work Load

Although it was originally intended that samples taken for process control should be used for in-process inventory data, PNC found that it was preferable to take extra samples, to be analyzed for Pu only, rather than re-schedule process control samples to match inventory requirements. Thus it is estimated that material balance data acquisition required about 41 man-hours of effort per week (over and above the effort required for process control and conventional materials accountancy). This is shown graphically in Figure 6.1. The mixing and sampling effort was small, only 2.1 man-hours per inventory. The bulk of the effort was in the analytical laboratory, and was about equally divided between provisional analysis of input and product samples to meet the ten day timeliness requirement and analysis of the weekly in-process inventory samples.

Concentration of this effort around weekly inventories led to peak load increases on the order of 10 - 15%.

Plant operations varied widely during the field test work, and in some cases the near-real-time accountancy data responded to these variations, producing short-term anomalies in the data. The short-term nature of these anomalies is considered important, because in all cases offsetting anomalies occurred as the plant operations returned to normal. In no case did anomaly investigation require any excessive effort or lead to any unwarrented suspicions or allegations.

Fig.6.1 Necessery Work Load for NRTA (10 days detection time model). TRP PNC 1985

Measurement Items		Flequency of	Sample Taking			Analysis			Total Load (for 7 days)
Measurements	Vessels	Measurement	Method		Work Load	Method		Work Load	
Flow Measurement	Input 251V10	twice/ a day	Solution Mixing	Air Sparging	Performed as Conventional Maeterials Accountancy routine work.	Pu conc.	Mass Spectroscopy	Conventional MA routine work	Conventional MA routine work + 23.8 man hour / 7 days
							Absorptiometry	1.7 man hour/once	
			Sample Taking	Air Lift		Density	Vibration Method	Conventional MA routine work	
						Treatment of take out samples			
	Output 266v23	once / a day	Solution Mixing	Pump Circulation		Pu conc.	Coulometry	Conventional MA routine work	
							K-edge densitometry		
			Sample Taking	Direct		Density	Vibration Method		
						Treatment of take out samples			
In-process Inventory Taking	251V11	once / 7 days	Solution Mixing	None	Total 2 man hour/once	Pu conc.	Absorptiometry	1.7 man hour/once	17.1 man hour / 7 days (In case of twice/15weeks for 266V24 and 276v20.)
			Sample Taking	Air Lift		Density	Vibration Method	1.0 man hour/once	
	255V12	once / 7 days	Solution Mixing	Air Sparging		Pu conc.	Absorptiometry	2.5 man hour/once	
			Sample Taking	Air Lift		Density	Vibration Method	1.0 man hour/once	
	265V12	once / 7 days	Solution Mixing	Air Sparging		Pu conc.	Absorptiometry	1.7 man hour/once	
			Sample Taking	Air Lift		Density	Vibration Method	1.0 man hour/once	
	266V12	once / 7 days	Solution Mixing	Air Sparging		Pu conc.	Absorptiometry	1.7 man hour/once	
			Sample Taking	Air Lift		Density	Vibration Method	1.0 man hour/once	
	266V13	once / 7 days	Solution Mixing	Air Sparging		Pu conc.	Absorptiometry	1.7 man hour/once	
			Sample Taking	Air Lift		Density	Vibration Method	1.0 man hour/once	
	276V20	once or twica / a campaign	Solution Mixing	Air Sparging		Pu conc.	Absorptiometry	1.7 man hour/once	
			Sample Taking	Air Lift		Density	Vibration Method	1.0 man hour/once	
	266V24	once or twica / a campaign	Solution Mixing	Pump Circulation	0.5 man hour/once	Pu conc.	Absorptiometry	1.7 man hour/once	
			Sample Taking	Direct		Density	Vibration Method	1.0 man hour/once	
Total Load (for 7 days)					2.1 man hour/once / 7 days			38.8 man hour / 7 days	40.9 man hour / 7 days

The clerical effort required during the field test has not been estimated, but was not insignificant. This effort was required to collect data, prepare tables, investigate anomalies, etc. Much of this effort occurred during a period when field testing was an internal PNC project, however, and an additional significant fraction was related to the evaluation of the field test effort and the preparation of this report.

When the ten day detection time model was originally defined, detection of possible abrupt diversion within ten days of the event was considered an important requirement. Through the years this requirement has gradually been relaxed, and it is worthwhile to consider how the model might be modified.

It appears to be desirable to retain the requirement for in-process physical inventories at nominally weekly intervals. However there appears to be no valid reason why all sample analyses must be completed within three days. Substitution of a requirement that sample analyses be completed within say two weeks would eliminate the need for provisional analysis of input and product transfers, and thus would reduce the estimated workload from 41 man-hours per week to about 17 man-hours per week. Relaxation of the timeliness requirement would also give the analytical laboratory more freedom in scheduling analytical work to reduce or eliminate peak workloads.

6.2 Effects of Unmeasured Inventory Fluctuation

In the early stages of the field test, the effect of unmeasured inventory fluctuation was assumed to be a major element of uncertainty in near-real-time materials accountancy. The field test work show that the inventory in the solvent extraction system varies with a standard deviation in the range of ±15%, based on calculations using the transient code MIXSET. However, the inventory of the solvent extraction system is no more than 25% of the total in-process inventory. In comparison, the uncertainty in the measured portion of the in-process inventory which is more than 75% of the total inventory was estimated to be in the range of ±10%.

Accordingly, at PNC-Tokai the uncertainty of MUF can be regarded to be caused mainly by measurement uncertainty in the in-process inventory. The effect of the fluctuation of unmeasured inventory under steady-state conditions on near-real-time accountancy appears to be unimportant.

6.3 Data Requirement

During the field test in the 85-2 campaign, PNC collected near-real-time accountancy data after every product batch, on a "daily" basis as reported in section 5.5. In this study, PNC did not take extra samples for in-process inventory taking; the data required for near-real-time accountancy were extracted or estimated from process operation data. The spread of the obtained MUF data is not significantly different from the spread of the MUF data obtained by taking extra samples. This means that it is not always necessary to take extra samples for near-real-time accountancy to carry it out.

The unmeasured inventory estimates reported in Section 5.1 were performed at PNC, and only the results were made available to the IAEA. Both the need for solvent extraction system inventory estimates on a weekly basis and the manner in which these might be calculated are still under investigation; the authors can only note that if it is decided that weekly UMI calculations are needed then the equations used for those calculations should be examined carefully with regard to potential confidentiality questions.

6.4 Inspection Authentication and Verification

One of the basic principles of IAEA safeguards is that operator-generated materials accountancy data might have been falsified, and therefore can only be used by the inspectorate if it has been verified or authenticated. The distinction is that verification implies independent IAEA measurement of randomly selected items or batches, whereas authentication implies that the IAEA has reason to believe that operator-controlled equipment is producing valid (i.e., authentic) data.

In the PNC-Tokai model of near-real-time accountancy, receipts, shipments and measured discards are based on exactly the same input and product flow measurements as for conventional materials accountancy. Verification of these input and product flow measurements is a part of current routine inspection practice, and need not be discussed further here. The questions which do require discussion are, to what extent does the in-process inventory data used in near-real-time accountancy require verification or authentication, and how might this be done.

Unfortunately, conventional verification theory treats each item of safeguards-relevant information independently. Sampling theory exists to define what fraction of the total data set must be verified in order to achieve some specified confidence that the total set has not been falsified (or that the total number of falsifications, if any, is below some specified threshold considered not to have safeguards significance). This theory, however, assumes that all items within a given stratum are equally likely to have been falsified and that verification of one item gives no information regarding any other item other than through the random selection process.

These assumptions are not applicable in near-real-time accountancy. Any falsification in the in-process inventory data only transfers the diverted quantity to the next material balance period. The falsfication must be repeated to prevent detection (through MUF analysis) in the second period, and it may need to be increased if additional quantities are diverted. Any data element which has been verified in the recent past has a reduced need to be verified again in the near future. Similarly, failure to verify a given data element at any particular in-process inventory taking may be of reduced significance if that data element is scheduled for verification in the near future. A viable theory for the verification of multiple material balances is, to the best of the authors' knowledge, non-existent.

Only limited in-process inventory verifications were performed during the 85-2 campaign field test. All in-process inventory tank volume readings were observed, and the raw data (per cent of full scale on a chart recorder) were converted to volume using calibration data supplied by PNC. This calibration data could have been falsified, but only by making one consistent falsification in the entire data set. This falsification could have concealed a diversion in the first weekly balance period, but it could not have concealed further diversions during subsequent weekly balance periods, because those balances necessarily used beginning and ending inventories based on the same falsification. Even the amount possibly diverted during the first period would be detected during the last period, when a cleanout inventory is taken and the falsified tank calibration data is not used.

Analytical data falsifications (in the in-process inventory data) would still be possible based on the verifications performed during the field test. This was known, and was accepted. The inspectors could have witnessed sampling, using previously identified sample bottles, and then could have followed sample preparation activities through shipment to the Agency's laboratory for verification analysis. However, it would have taken a lot of preparatory work, because SAL is not immediately prepared to handle these intermediate samples. In a routinely functioning system the inspector would need some easily available NDA measurement technique, possibly the K-edge densitometer, which could be used for random spot check analyses.

Only a few verification analyses would be needed, because again the falsification possibilities are severely limited. There has been essentially no theoretical work in this field, but randomly selecting one tank for analytical verification each week probably would be sufficient.

6.5 Effectiveness

It is difficult to discuss the effectiveness of near-real-time accountancy at PNC-Tokai in quantitative terms because planned statistical evaluations have not been completed. However, some preliminary remarks can be made using the linear regression calculations which have been included in this report.

Using linear regression, the standard error of estimate, the ability to predict CUMUF from a knowledge of either material balance period or throughput, was never worse than ± 1100 grams, and for the 1985 data was only ± 900. Multiplying by 3.3 to introduce false alarm and detection probabilities suggests that any abrupt diversion larger than 3.0 – 3.5 kgs Pu would have at least a 95% probability of being detected as an anomaly requiring further investigation. Or, as the authors suggest might be a more appropriate way of looking at the data, an 8 kg abrupt diversion would have at least a 99% probability of detection, even using a false alarm rate of 0.1% or lower.

Abrupt diversion detection sensitivity using more sophisticated statistical tests probably is better than suggested above. More important, however, is the fact that considerable investigative information becomes available quickly and without effort, in the form of the next week's data point. Inventory errors, such as occurred during the 82-1 campaign, are revealed as inventory errors. There never is a need for a special cleanout inventory "to find out how much is really missing".

Detection sensitivity for protracted diversion is difficult to assess from simple linear regression, Several of the graphs show CUMUF lines which have non-zero slopes based on visual observation. Most of these slopes probably are also statistically significant in terms of estimated measurement uncertainties, but that remains to be established. The question then is, are the non-zero slopes the result of uncorrected measurement biases, unrecorded process losses or protracted diversions. This question cannot be answered through statistical analysis of material balance alone. (The conventional approach to statistical analysis of material balance data does not answer this question. Rather it arbitrarily assumes the existence of uncorrected measurement biases up to a limit defined by systematic error variances, and dismisses any MUF and CUMUF smaller than this limit.)

Since most studies based on modelling data conclude that detection sensitivity for protracted diversion is somewhat less (i.e., standard deviations are larger) than detection sensitivity for abrupt diversion, the authors suggest that 6 - 7 kg Pu is probably an upper limit. More precise statements must wait for better statistical data evaluations.

7. Conclusions

The results of these field tests performed during the past six years provided important experiences for better understanding of the characteristics of near-real-time accountancy in the actual field. One of the most important results is that near-real-time accountancy will be effective in detecting possible abrupt diversions. However, another result is that the existence of measurement biases affects the ability of near-real-time accountancy to detect protracted diversion. To improve the effectiveness of near-real-time accountancy, estimation of realistic measurement uncertainties and factor analysis of measurement biases will be important subjects in future, as well as studies on statistical evaluation techniques.

Acknowledgement

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