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Large-scale automation of the Lawrence Livermore Laboratory x-ray analytical facilities

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Abstract

Lawrence Livermore Laboratory (LLL) has undertaken an ambitious plan to automate its x-ray analytical equipment. This project ultimately will automate 15 x-ray diffraction and 3 x-ray spectrometric systems. All automation is being done by retrofitting existing equipment and combining it with minicomputers to produce "smart instruments." Two types of "smart instruments" have been developed: one that controls an experiment and acquires data and another that analyzes data and communicates with LLL's large computer center. Three of the former type have been built and are operating; seven more will soon be put into service. Only two of the latter type are needed, and both are currently in service. We describe the details of our overall plan, the "smart instruments," the retrofitting, our current status, and our software.

Introduction

At Lawrence Livermore Laboratory (LLL), we use x-ray diffraction in support of all laboratory programs. These programs include research in nuclear-weapons design, laser physics, alternative energy sources, the earth sciences, materials science, and the biomedical program.

Before this project was begun, this x-ray diffraction work was done by six small x-ray facilities located in five different buildings. Nearly all of the instrumentation involved was operated in a manual mode. (The one exception was a 4-circle, single-crystal diffractometer automated about 1968, which has since been decommissioned.)

Two inefficiencies were noted for this mode of operation. These were:

1. The physical dispersal of the laboratories promoted competition rather than cooperation; the separation also confused customers about which lab was best for their purposes.

2. The department and divisions that ran the laboratories had insufficient resources to either a) keep the instrumentation up-to-date or b) maintain staff to run the instrumentation effectively.

To resolve these problems, the six labs were consolidated into two—one for radioactive materials and the other for all other materials—and money

was allocated and people were committed to design and install automated data-collection and analysis systems in both labs.

Two approaches were available to accomplish our task. The first was to buy off-the-shelf commercial units and the second was to retrofit existing equipment. Our principal limitation was financial: \$100,000 was available to buy capital equipment for the project. These funds would have permitted the purchase of only one commercial unit, which would not have satisfied the goal of automating both of our laboratories.

We thus investigated the cost of retrofitting the existing systems. We found that, by using existing LLL technology, the LSI-11 computer (a noncapital item), and retrofitting technology developed by Parrish¹ on a similar project, we could automate nearly all of our existing systems for the dollars available.

The instrumentation that was to be automated included 15 x-ray diffraction experiments (most of them diffractometers) and 3 x-ray spectrometers. The latter instruments are all located in the radioactive-materials laboratory.

The principal benefits expected to accrue from the project were:

- Higher-quality data with much greater inherent precision.
- Expanded data-analysis capability.
- Greater speed of analysis.

These benefits have in fact been realized by others¹⁻⁶ doing automated x-ray analysis, so we

were confident that they were practical goals. In addition to these goals, we also established the ability to perform "routine" phase identification in 10 minutes as a long-term goal for our labs. This latter goal should prove a challenge for years to come.

The x-ray analysis automation plan

An x-ray analysis system involves three distinct areas:

- Instrument control.
- Data acquisition.
- Data analysis.

We decided to approach the automation of these functions in three phases to get the most for our relatively limited dollars. Phase I was the automation of the instrument control and data acquisition. It also provided the flexibility for expansion of any unit, addition of new units, and in-lab analysis. Phase II involved developing programs for in-lab analysis and for obtaining analysis routines from vendors. Phase III was to pursue the possibility of developing a large in-lab analysis system and modifying the very large analysis routines normally run on LLL's time-share system (LTSS) so that they could be run in our laboratories.

Several alternatives were examined to determine the best system architecture for our x-ray labs. We finally settled on using a "smart-instrument" approach. This design makes all experimental instruments independent of one another and independent of any instrument devoted to only data analysis, i.e., no in-lab time-sharing is involved. This modular approach using smart instruments allowed us to adopt a design that met our Phase-I goals for both laboratories.

Data-collection units and analysis units are the two building blocks on which the automation system is based. Data-collection units control the diffractometer (or spectrometer) and collect and store data on floppy disks. At present, some minimal data analysis can also be done on these units.

The information stored on the floppy disks by the data-collection units is then transferred to an analysis unit. There, the data can either be analyzed using in-lab software or they can be transported to LTSS for analysis by advanced crystallographic (or spectrometric) codes. Figure 1 shows the conceptual layout of this system.

The money available to us permitted construction of 10 data collectors and 2 analysis units. The radioactive-materials laboratory was allocated 4 data collectors to be shared by 7 x-ray instruments. This laboratory was also allocated 1 analysis unit. The other lab was equipped with 6 data collectors shared by 11 instruments; it also has 1 analysis unit. The data collectors are portable and can be easily moved as needed.

Data collector unit

A goniometer (or spectrometer) combined with a data-collector unit becomes a "smart instrument" capable of controlling an experiment and collecting data independently of any other experimental station. Our data collection units include the following:

- An LSI-11 computer with 28K words of memory.
- A dual floppy-disk unit.
- A KSR-43 terminal.
- A scaler.
- A 2θ display.
- An LLL-designed interface/controller.
- A conventional solid-state x-ray pulse-counting system.

This system is shown schematically in Fig. 2.

The LSI-11 runs under the RT-11. Version IIIB, single-job monitor with BASIC as the primary system language. A 1-MHz programmable clock is used as a precision timing source. Assembly-language subroutines have been installed in BASIC to control the stepping motor, scaler, 2θ display, clock, and strip-chart recorder.

A Slo-Syn M062-FC03 stepping motor was chosen for angle control, and many of these have been installed on experiments. STM-1800 translator modules (Slo-Syn) are being used to control and drive these motors. One drive pulse sent from the LSI-11 turns the motor axis 1.8 deg. On a Philips diffractometer, this is equivalent to a minimum step size of 0.005 deg 2θ .

FIG. 1. Conceptual layout of the automated data-gathering and analysis system at LLL. Data-collection units control the goniometer (or spectrometer) and collect and store data on floppy disks. The stored information is then transferred to an analysis unit where the data is either analyzed using in-lab software or transported to LTSS. In the radioactive-materials laboratory, a data-collection unit serves a spectrometer, as shown here.

The KSR-43 terminal provides hard-copy capability for each data collector as well as communication with the LSI-11. Data, run information, and rough graphical output can be printed on the 132-character-wide paper.

Floppy disks are used to store programs and data. The output data from the data collector are then taken to an analysis unit either for in-lab graphical analysis or for transmission to LTSS.

Analysis unit

These units permit the user to run in-lab analysis codes or transmit data to LTSS. Each analysis unit contains:

- An LSI-11 computer with 28K words of memory.
- A dual floppy-disk unit.
- An interface to the large time-shared computers at either 300 or 9600 baud rate.
- Tektronix 4012 graphic-display terminal.
- Tektronix 4631 hard-copy unit.

A schematic representation of an analysis unit with its connections to our large computer system is shown in Fig. 3.

In addition to these items, the analysis unit in the nonradioactive-materials laboratory has a 10-Mbyte hard-disk unit to permit data archiving and greater program flexibility. A KSR-43 terminal will also be added to this system to serve as a line printer.

The analysis unit runs under the RT-11, Version IIIB, single-job monitor and with either BASIC or FORTRAN as the system language. On the analysis units, however, a FORTRAN compiler and support libraries are used extensively. Various plotting routines are available to provide interactive graphics capabilities.

The LTSS interface uses a board previously designed at LLL and modified for this project. The interface ties directly into a LTSS terminal port and allows either the LSI-11 or the LTSS to use the line. Data transfer rates from the LSI-11 depend on baud rate of the port (either 9600 or 300 baud).

FIG. 2. Principal components of a "smart instrument." The data-collector unit serves as the core of our automated x-ray instruments. Each "smart instrument" can control an experiment and collect data without interacting with any other experimental station.

Retrofitting the x-ray equipment

To date, the only systems that have been retrofitted use the Philips Wide-Range Goniometer. We mount the stepping motor on this instrument using the technology developed by Parrish.¹ In addition, because the goniometers must be partially disassembled during this step, we take advantage of this opportunity to:

- Remove all mechanical linkages to the original synchronous motor.
 - Remove the motor itself.
 - Remove all unneeded switches and wiring associated with manual operation of the instruments.
 - Change the detector counterweight from the lead unit supplied by the vendor to a thicker one made of tungsten.
- The counterweight change is made only for the diffractometer systems and not for the spectrometers. For the diffractometers, the change is necessary because the weight of the diffracted-beam monochromator is not properly balanced by the lead counterweight.

We then add the wiring appropriate for the stepping motor. However, in stripping the instrument of unneeded items, we do not remove the half-angle and limit switches. These two switches are wired into our data-collector system. The half-angle switch permits us to compare motor pulses sent to the motor with the actual distance travelled. The limit switches are used to protect the instrument from being driven into the tube tower or the table top.

The diffractometer retrofitting described here is identical to the procedure that will be used to modify the two wavelength-dispersive x-ray spectrometers located in the radioactive-materials laboratory. This is possible because those spectrometers and diffractometers use the same type of goniometer.

Other retrofitting projects now either in progress or under consideration include:

- Mounting stepping motors on Weissmann double-crystal diffractometers.
- Mounting a stepping motor on a small-angle diffractometer.
- Picking up the temperature and pressure

FIG. 3. Principal components of an analysis unit showing its links with LTSS components located both in and outside of the x-ray diffraction laboratory. The analysis units permit the user to run in-lab analysis codes or transmit data to LTSS.

controls on our high- and low-temperature x-ray diffractometers.

- Interfacing to a position-sensitive detector system and its multichannel analyzer.

- Automating a film densitometer.

- Interfacing to an energy-dispersive spectrometer system.

- Automating a diffractometer sample changer.

Of these retrofitting projects, the position-sensitive detector system is the closest-to-completion. This system, when complete, will permit the rapid transfer of 4096 channels of information from the memory of the multichannel analyzer to a floppy-disk unit. This retrofitting required only that the analyzer (a Northern Scientific 720) be modified for parallel data transfer and that new cabling be fabricated.

Work on the remaining projects is, for the most part, in the planning stage.

Software

We divide our discussion of software into two parts: the software used to run the data-collector and analysis units (designated user software) and analytical software that is (or will be) used on our data-analysis system.

User Software

Three user programs written in BASIC are available for data collectors: SETUP3, SCANPL and SCANPR. SETUP3 is the standard program for running routine samples in the automated mode. SCANPL simulates the continuous manual-mode operation using only a strip chart. SCANPR digitizes the data and prints out a graphic plot on the terminal with 2θ angle and counts. Because the SETUP3 program cannot format data in a manner directly usable by our analysis program, a FORTRAN program (GFORMT) is used to reformat the data from SETUP3.

In addition to these programs, two user programs are available on the analysis units. One is a general-purpose routine that sends any ASCII file from the analysis unit to the LTSS. The other reformat the d-spacings and intensities calculated by an analysis program and sends them to LTSS where

they are analyzed by a crystallographic Search/Match program.⁷

Analysis Software

Currently, only one FORTRAN program (DIFPLOT) is being used on the analysis units; DIFPLOT analyzes and plots the x-ray diffraction patterns produced by our data collectors. This program is based on a program originally written by Goehner^{8,9} to run on a Honeywell 600 computer using a Tektronix 4010 graphics terminal for input/output.

The program offers several analysis features. If the data are noisy, they can be smoothed by a least-squares, moving-average technique developed by Savitzky and Golay.¹⁰ A background pattern can also be determined and subtracted from the raw or smoothed data using a linear-least-squares, background-subtraction algorithm. To facilitate peak identification, $K\alpha_2$ peaks can be stripped from background-subtracted data using one of two algorithms.^{11,12} (A pair of peaks, $K\alpha_1$ and $K\alpha_2$, represents each diffraction line in the original pattern; only one peak is required for identification.) Peaks are located using a second-derivative peak-search algorithm, and the resulting peak intensities and areas can be calculated and normalized to the peak with the highest value. Finally, DIFPLOT can order the peaks by normalized peak intensities. Figure 4 shows an example of DIFPLOT output.

Analysis modes include:

- Full-screen display of raw data, analyzed data (including identified numbered peaks), and background.

- Overlaying of two patterns (both to the same scale).

- Split-screen display of two patterns (one on the upper half of the screen, the other on the lower half, but both to the same scale).

- Full-screen display of the difference between two patterns.

Figure 5 shows the logic flow for the full-screen complete-analysis mode of DIFPLOT. To date, this software mode has been used the most.

A second analysis program, called SPECPLT, has also been developed. This program takes data from automated wavelength-dispersive x-ray spectrometers and analyzes and plots those patterns. Most features are the same as those of DIFPLOT except that this code produces

FIG. 4. Example of DIFPLOT output. DIFPLOT analyzes and plots the x-ray diffraction patterns produced by our data collectors. Noisy data (a) can be smoothed by a least-squares, moving-average technique and the background subtracted to give (b). The identified peaks are numbered sequentially. The spectrum shown here is the x-ray diffraction pattern obtained from volcanic ash produced by the May 18, 1980, eruption of Mount St. Helens. The sample was collected at Ellensburg, Washington.

FIG. 5. Logic flow for the full-screen complete-analysis mode of DIFPLOT, a program that analyzes and plots the x-ray diffraction patterns. Raw input data are plotted during the first pass and full analyses are available during the second and subsequent passes.

energies and wavelengths instead of d-spacings. The program will be put into service when the first spectrometer is automated.

In the future, we plan to convert many of our LTSS FORTRAN programs to run on the analysis unit. All but the largest programs will be converted.

Project status

Table 1 summarizes the current situation with respect to the hardware necessary to build our automated systems. In a nutshell, we now have three operating automated diffractometers, all in the main laboratory. We also have two complete analysis units, one in each laboratory. Our present emphasis is to install the remaining data collectors and to align goniometers as rapidly as they are modified.

To date, we have spent about two years on the project, and we anticipate that at least another year will be needed to finish the work. The charge-rate (and thus time charges) should be lower during the next period, however, because the heaviest project costs were incurred during the planning and construction phases, which have now been completed.

TABLE 1. Current status of the various hardware items required to complete the original automation plan. This plan calls for one analysis unit in each of two x-ray laboratories and ten data collectors (six in one lab and four in the other).

Hardware item	Number required	Built to date	Electronics debugged	System in use
Analysis unit	2	2	2	2
Data collector	10	10	7	3
Modified goniometer	16	7	7	3
Interface unit	10	10	7	3

Conclusions

The results we are now obtaining from our automated instrumentation amply justify the time and money spent on this project. The increases in sensitivity, precision, and accuracy noted by others¹⁻⁶ are quite apparent in our data. Because of these advantages, virtually 100% of the nonradioactive-materials work we do is now being done on automated systems.

We believe that large-scale automation is a practical concept for x-ray laboratories such as ours. The combination of minicomputers and the retrofitting of existing equipment permits most systems to be automated cheaply. Thus, each con-

verted manual system has the chance (if the hardware is in good shape) to produce the high quality data mentioned above. Best of all, of course, is that the unit cost of automation done in this fashion can be minimized. Our costs will ultimately be spread over at least 18 instruments of various types.

We are still committed to maintaining the overall goal of a ten-minute routine phase analysis. To achieve this, we expect that we will adopt some form of the analysis procedure developed by Parrish¹ and that we will also modernize our sample-preparation area.

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