The Safeguards Analytical Laboratory: Its Functions and Analytical Facilities

International safeguards agreements give to the IAEA the responsibility and the obligation to perform independent measurements to verify the accountancy of safeguarded fissile materials. The accountancy is generally based on three categories of measurements: (1) the volume or the mass of material; (2) the uranium, plutonium and thorium content of the material; and (3) the abundance of the fissile isotopes in the material.

In the past few years, non-destructive techniques have made striking progress and this often makes it possible for IAEA safeguards inspectors to perform a complete measurement verification in the field. These non-destructive techniques are discussed in another article in this issue. When a satisfactory non-destructive technique is not available, the inspector in the field can only witness the measurement of the volume or the mass of the material and obtain from the operator a representative sample of this material which is sent to an analytical laboratory for destructive analysis.

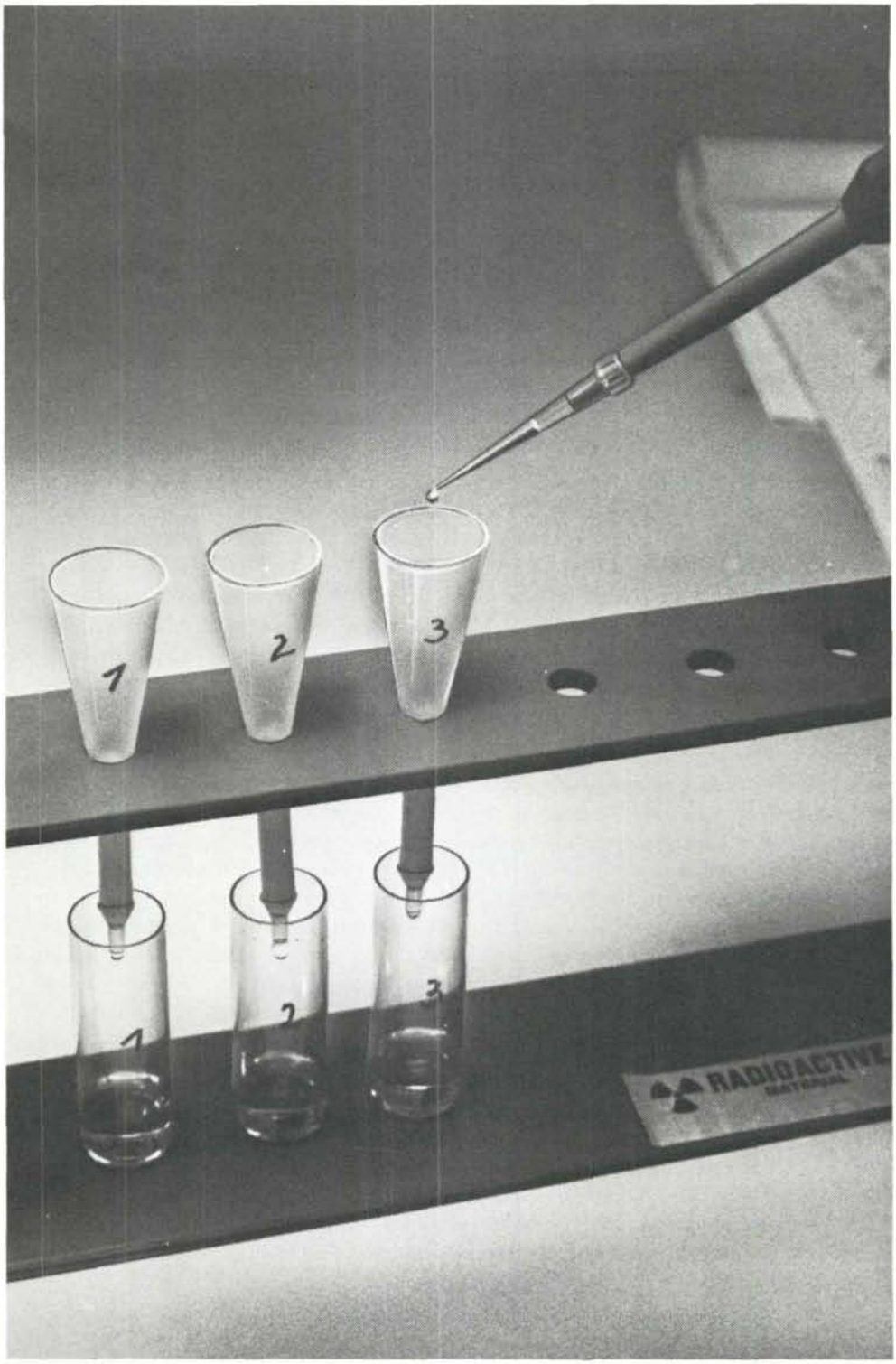
The availability of suitable non-destructive techniques has already a definitive impact on the analytical programme of safeguards, as it has considerably contributed to limit the growth in the number of samples submitted for destructive analysis. The use of non-destructive techniques, however, brings a concomitant need for well-characterized standard materials for calibration, and the characterization of standard materials is usually achieved by means of destructive analytical methods.

The IAEA Safeguards Analytical Laboratory (SAL) at Seibersdorf, Austria, has been designed to perform rapid, accurate analyses of nuclear material. It has been staffed and equipped to cope with a maximum sample load of about 2000 samples a year. The Agency has thus with this laboratory a solid tool to obtain the necessary basic measurements to fulfill the analytical requirements of its international safeguards responsibilities.

The laboratory, its main installations and its general functions were described in the December 1975 issue (Vol. 17, No. 6) of the *Bulletin*. The building had then just been completed and the first analytical equipment was being moved in. The laboratory is presently operating under a limited type B license issued by the Austrian authorities. Full operation as a type A laboratory is pending the signature of a further agreement with the Austrian Government. This agreement will establish the procedures of co-operation between the Österreichische Studiengesellschaft für Atomenergie (SGAE) and the IAEA for a safe operation of the Safeguards Analytical Laboratory.

However, practically all foreseen equipment has been installed in the new laboratory (the last major instruments to be installed were the two mass spectrometers) and virtually all expected services can be fulfilled. We present here briefly the analytical capabilities of the new Safeguards Analytical Laboratory.

Ion-exchange micro columns containing tiny resin beads (under the funnels) extract pure fractions of uranium and plutonium from samples of irradiated fuel. The purified fractions are then submitted to mass spectroscopy for measurement. ▶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GENERAL ORGANIZATION

The laboratory is functionally organized into five specialized areas: (1) wet chemical analysis of uranium-containing and thorium-containing materials; (2) wet chemical analysis of plutonium-containing materials; (3) mass spectrometric analysis; (4) radiometric measurements; (5) emission spectrography.

The samples are first received in the sample reception and store rooms and then routed to the appropriate wet chemical analysis laboratories. There they are analysed for uranium, thorium or plutonium content, and purified aliquots are prepared for the isotopic analysis of these elements. This isotopic analysis is routinely performed by mass spectrometry, although radiometric techniques are necessary for back-up. Emission spectrography serves to detect the presence of impurities which could interfere and bias the results of the chemical and isotopic analysis of uranium, thorium or plutonium. This combination of techniques provides the assurance of selective and accurate analyses.

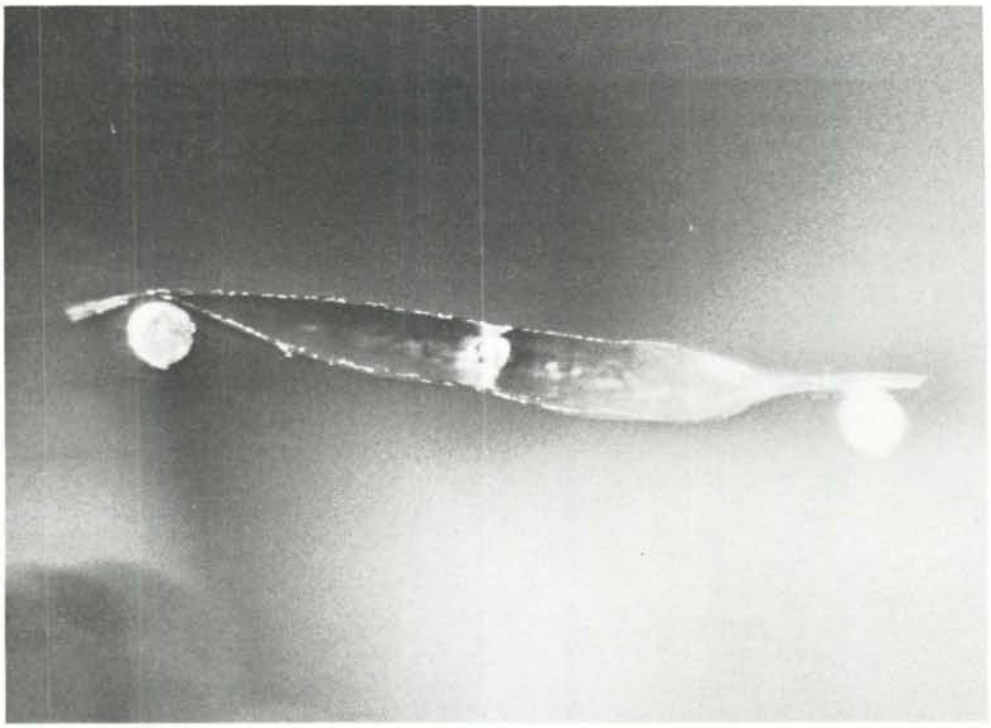
DETERMINATION OF FISSILE CONTENT

For non-irradiated materials, standard methods of chemical assay apply when the sample aliquots contain at least 1 mg to 1 g of the fissile elements. For example, the uranium content of oxide material may be determined by igniting 1 to 20 g of the material at 900°C to form a well-defined U_3O_8 oxide which is weighed. This gravimetric procedure can yield excellent relative precision, i.e. better than 0.01%, but its accuracy depends entirely upon the purity of the ignited oxide. The purity of the oxide is determined by emission spectrography. In general, the method is used for the analysis of uranium oxides with impurities of 0.01 to 0.1%.

Much more selective is the titration method which constitutes the basic procedure in the laboratory. This method, frequently referred to as Davies and Gray's method, is now widely used especially where accuracy and therefore selectivity and precision are paramount. Davies and Gray developed the titration method at Winfrith (UK) and presented it in 1964. It was further refined by Lerner and his co-workers at the New Brunswick Laboratory (USA) to relative precisions and accuracies of 0.01%. The method requires aliquots containing 10 to 300 mg of uranium, and it applies to the assay of uranium in a wide range of materials. A point not to be neglected: the accuracy of this method is conveniently verified by a direct comparison of a uranium and a potassium dichromate primary standard. This method has been in use in Seibersdorf with very good results since 1974 with a semi-automatic installation. A fully automatic unit has recently become operational in SAL with astonishingly good precision and reliability.

Electrochemical techniques, like controlled potential coulometry, are even more sensitive. SAL is equipped with instrumentation designed for the analysis of aliquots containing 2 to 20 mg of uranium or plutonium. A compact automatic instrument is installed in a glove-box for the routine determination of plutonium. Its precision is expected to be within 0.2–0.05%. The coulometric determination of plutonium suffers, however, from the interference of iron, which must be corrected for. More selective and accurate results, if required, will be provided by a titration technique installed in a separate glove-box. Such a technique has been used in Seibersdorf since 1969.

The plutonium wet chemical laboratory of SAL is equipped with two chains of six analytical glove boxes. Each box of a chain is equipped to perform a specific step of the



Ion-exchange resin bead (centre) is mounted on a canoe-shaped rhenium filament for analysis in a two-stage mass spectrometer. The resin bead contains from 10 to 50 ng of uranium and plutonium.

analytical procedure: sample aliquoting, weighing, dissolution, purification, measurement.

It should be noted that the methods apply mainly to feed materials, intermediates and products of fuel fabrication plants. The corresponding samples are usually in a solid form and the accuracy of their analysis is usually not limited by the performance of the analytical technique but rather by the representativity and stability of the samples. The minimum sample size is necessarily 2 to 20 g, depending upon the homogenity of the material and therefore well above the quantities actually needed in the final measurement. Fortunately these materials are not irradiated and readily accessible to sampling and their shipment to and their handling in SAL is thus a practical procedure.

This is not the case for irradiated materials, which are extremely radioactive and samples cannot be shipped without heavy shielding against the high-energy gamma rays emitted by the fission products they contain. These samples are therefore accurately diluted, 200 to 500 times, at the facility and under the observation of the Agency's inspectors. The input accountability tank of reprocessing facilities is the main point in the fuel cycle where sampling of irradiated fuel is necessary. One to three millilitre aliquots of the diluted samples are shipped to Seibersdorf: their beta-gamma radioactivity is now only about 10mCi. Each aliquot of diluted solution contains only about 1 mg of uranium and 10–20 μg of plutonium. This is too little for an accurate assay by the standard wet chemical techniques described above, so tracer techniques are used for isotopic dilution analysis.

Two-stage pulse-counting mass spectrometer was specially designed for the IAEA by the Oak Ridge National Laboratory.

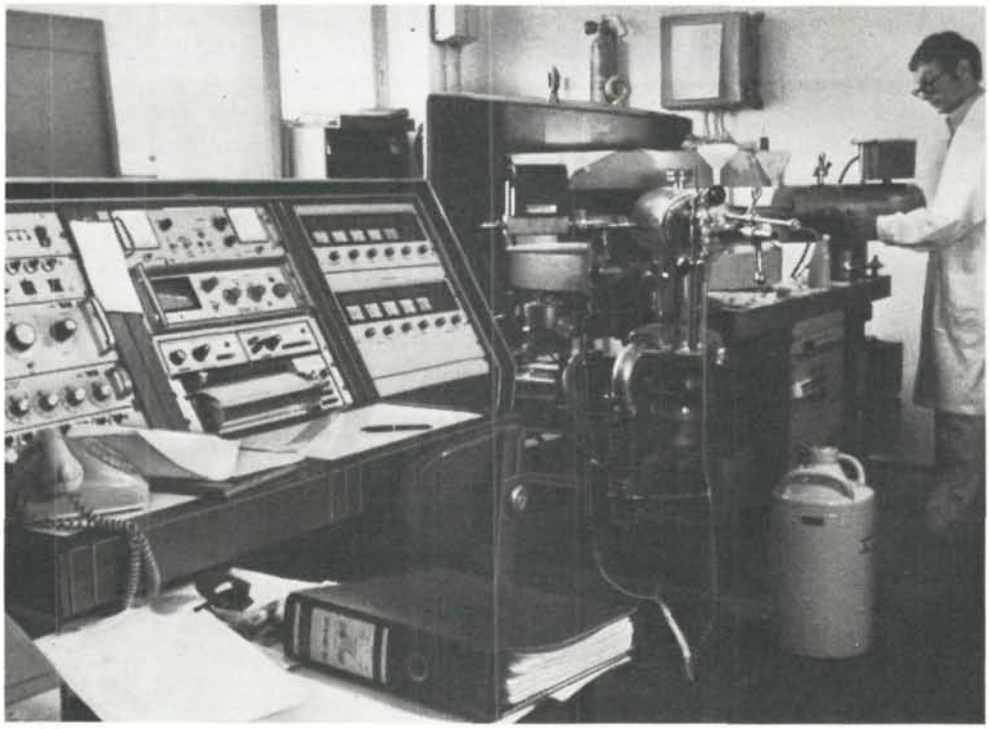
An accurately weighed aliquot of the diluted sample is added to a known amount of U-233 and Pu-242 or Pu-244 isotopes, which are practically absent of the sample. The quantities of U-233 and Pu-242 or Pu-244 tracers, also called spikes, are chosen to match the quantities of uranium and plutonium expected in the aliquot of the diluted sample.

Another aliquot is obtained and left unspiked for the isotopic analysis. These sampling and dilution operations are normally performed at the reprocessing plant. The necessary spikes are prepared and shipped to the plant by the Safeguards Analytical Laboratory at the request of the Agency's inspectors.

When received in Seibersdorf, both unspiked and spiked aliquots are processed behind a local shielding of lead bricks in special fume hoods. Pure fractions of uranium and plutonium, free from Am-241 isotope and fission products, are isolated by an anion exchange procedure on disposable micro-chromatographic columns and submitted to mass spectrometry for measurement.

OTHER CHEMICAL ANALYSIS MEASUREMENTS

As stated above, emission spectrography is our primary survey technique to ascertain the conditions of accurate assay. With minimal sample preparation, it can simultaneously provide semi-quantitative or quantitative determination of 10 to 30 elements that are present at concentrations between 0.01 and 0.1%. Of interest is the presence of elements which require special dissolution procedures or which interfere in the final measurement.



Single-stage surface ionization mass spectrometer (AVCO-type) requires 0.1 to 1 μg samples of plutonium or 1 to 10 μg of uranium.

The same instrument serves for the analysis of materials containing uranium, thorium and/or plutonium.

If an interfering element is found to be present at concentrations above 0.5 to 1%, emission spectrography may not provide appropriate results for correction of the chemical assay. Calorimetric and electrochemical techniques must then be used.

ISOTOPIC ANALYSIS BY MASS SPECTROMETRY

Two special surface-ionization mass spectrometers have been in operation at SAL since November 1976. Either instrument can be used for the isotopic analysis of uranium, thorium and/or plutonium, or other heavy elements, but they differ basically in their design and their characteristics. The first and more conventional spectrometer is a single magnet instrument with an electrometer read-out. It requires sample loadings of 0.1–1 μg of plutonium or 1–10 μg of uranium and can provide the determination of a 1% U-235 abundance with a relative precision of the order of 0.2%.

The second instrument was specially designed for the Agency by Oak Ridge National Laboratory (USA) and was installed in September 1976. It is a two-magnet instrument with pulse counting read-out. It is slightly less precise than the other spectrometer: a 1% U-235 abundance can routinely be measured with a 0.3 to 0.4% relative precision. But its sensitivity is about 100 times greater and optimal precision is obtained with sample loadings of only 10 to 50 ng.

This feature led R. Walker and his co-workers in Oak Ridge to propose in 1974 an elegant way to combine the purification and loading of the sample in a single operation. In the conventional procedure, a 1 to 10 μl drop of a dilute solution of the purified sample is transferred by pipetting to the surface of a very pure rhenium or tantalum filament. The filament is heated electrically to evaporate the liquid, and the filament is then mounted in the ion source chamber of the spectrometer. In the resin bead procedure proposed by the Oak Ridge National Laboratory, 10 to 20 beads of anion exchange resin beads are immersed overnight in about 0.2 ml of a nitric acid solution containing 0.1 to 0.5 μg of plutonium. The resin beads absorb uranium and plutonium but leave the americium and the fission products in solution. The acid concentration is adjusted so that the resin beads absorb equal amounts of uranium and plutonium, about 10–50 ng per bead. A loaded bead is taken out of the solution and transferred to a boat shaped rhenium filament.

The technique is extremely attractive for the safeguarding of irradiated fuel solutions since the resin beads can be immersed in the solution sample at the plant. The loaded beads, essentially free from fission products, can then be shipped to SAL in type A of even conventional industrial containers.

The other spectacular feature of the tandem instrument is its ability to measure the abundance of minute amounts of a minor isotope in presence of a neighbouring major isotope, for example parts per million of the U-236 and U-234 isotopes with respect to the U-238 isotope. Such information is of value in safeguarding procedures based on isotopic correlation techniques.

RADIOMETRY

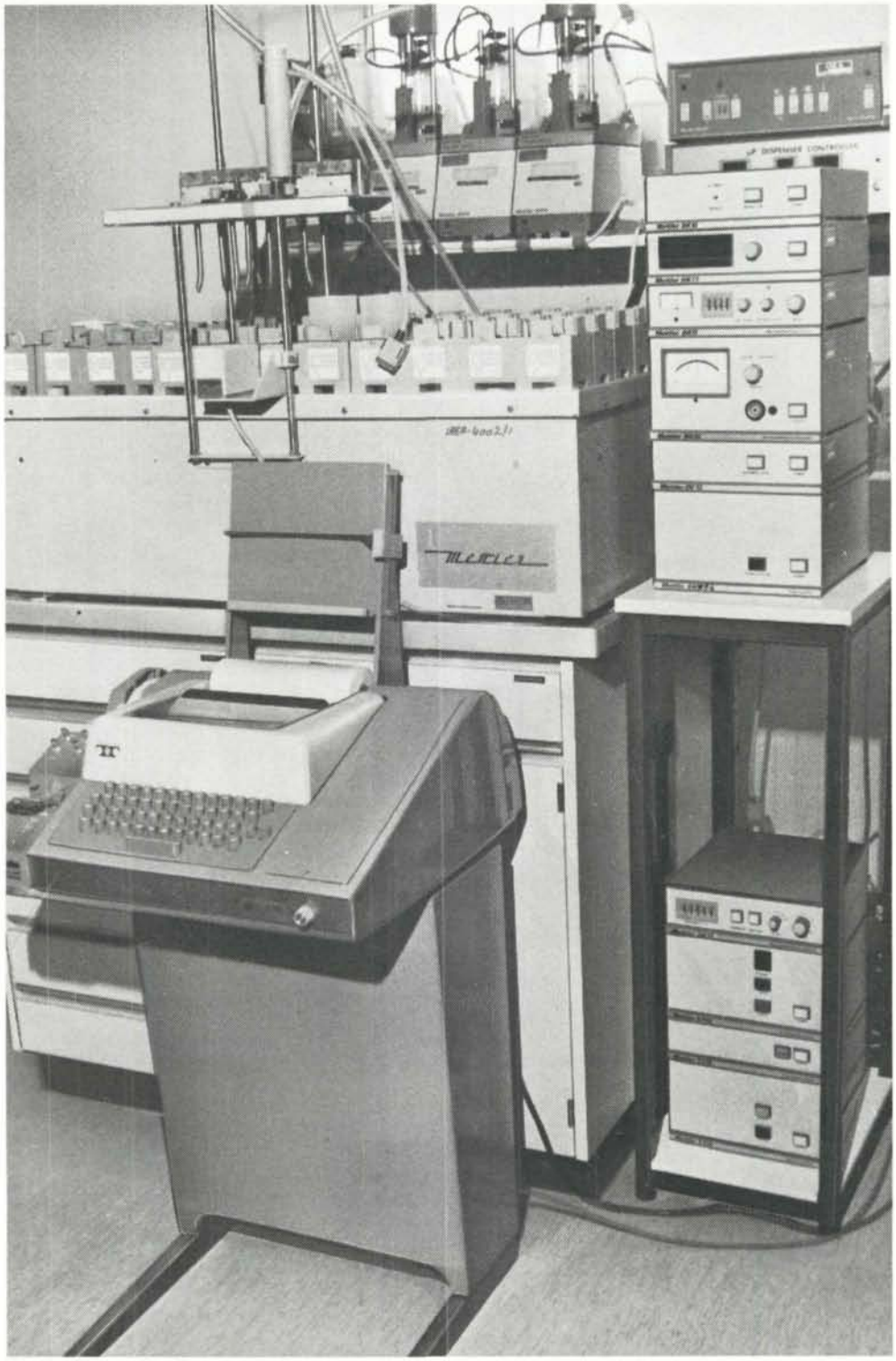
SAL is also equipped for scintillation and high-resolution gamma spectrometry, gamma transmission measurements and alpha spectrometry. These techniques are primarily intended as a back-up to mass spectrometry. U-235 and Pu-239 isotopic abundances can be measured by gamma spectrometry and the Pu-238 isotopic abundance can be verified by alpha spectrometry. They are also essential in the characterization of reference materials for non-destructive assay techniques.

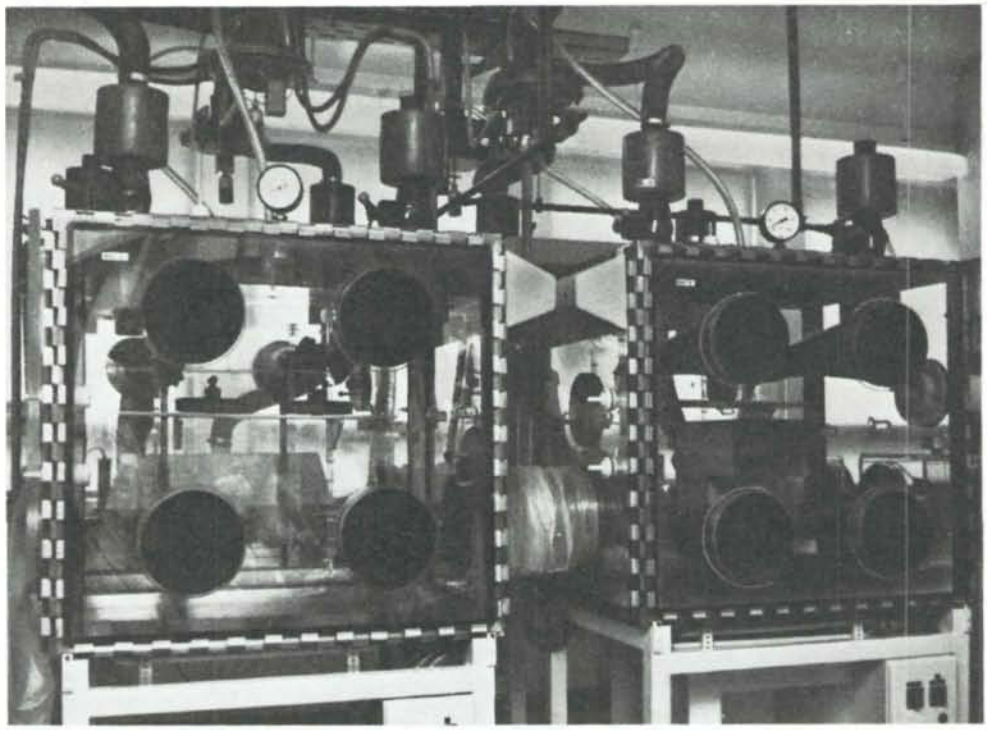
High-resolution spectrometry, for example, detects eventually radioisotopes such as Pb-212 which can interfere in the non-destructive measurement of the U-235 abundance. Gamma spectrometry is also used for rapid non-destructive testing of inspection samples and trouble shooting. Radiometric techniques are also basic for the control of radioactive wastes produces in the laboratory.

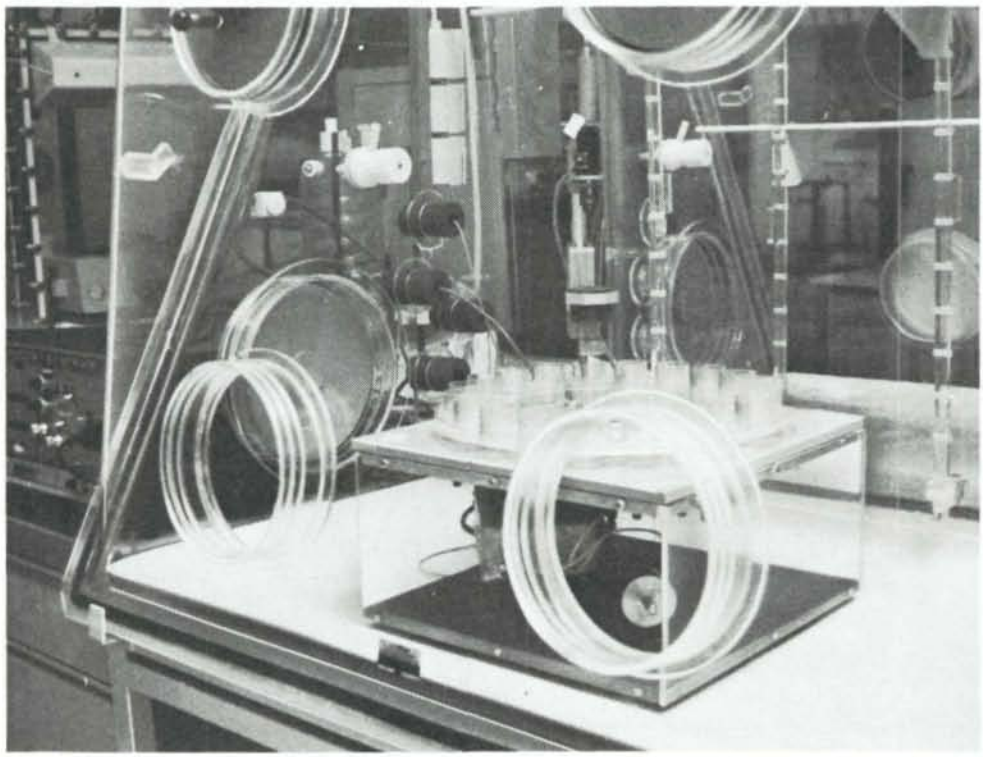
DATA PROCESSING

The mass spectrometers and gamma spectrometers are connected to minicomputers which control the collection of the measurements and perform the calculation of the results. All data reductions are in fact progressively done on these minicomputers. Other analytical instruments also can provide measurements for direct computer processing, so that eventually all analytical results and their reports will be obtained without any hand transcription. The use of minicomputers not only facilitates the analytical processes but

Automatic titrator for the potentiometric titration of uranium.







it also renders practical the application of statistical tests of the quality of the analytical results. Actually little remains to be added to the installations of the Safeguards Analytical Laboratory to permit a direct transmission of its analytical results to the larger computer facilities in the Agency's headquarters. The Division of Safeguards Information Treatment proceeds there to a further evaluation. The results of the Agency's safeguards laboratories are combined with the results provided by the Agency's Network of Analytical Laboratories (NWAL) and further tested in the Network's Quality Control Programme. The Agency's Safeguards Analytical Laboratory participates also in various international intercomparison exercises or international analytical quality assurance programmes.

Part of one of the chains of alpha glove boxes for the wet chemical analysis of unirradiated plutonium materials.

Automatic controlled potential coulometer in a glove box is used for routine determination of plutonium.