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ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

A WASTE CHARACTERIZATION MONITOR FOR LOW-LEVEL RADIOACTIVE WASTE MANAGEMENT

Détecteur de caractérisation des déchets destiné à la gestion des déchets radioactifs de faible activité

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Presented at the Sixth Annual Conference of the Canadian Nuclear Society Ottawa, Ontario 1985 June 2-4

Chalk River Nuclear Laboratories

Laboratoires nucléaires de Chalk River

Chalk River, Ontario

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Résumé

L'exploitation de la technologie et des procédés nucléaires pour le bien des Canadiens donne lieu à la production courante d'environ 12 000 m par an de déchets radioactifs solides de faible activité. Ces déchets doivent être isolés aussi longtemps qu'ils pourraient faire courir un risque au public et à l'environnement. Au Canada, la planification actuelle prévoit le développement et l'utilisation de divers types d'installations de stockage et d'enfouissement ayant différentes caractéristiques de confinement. Pour démontrer l'efficacité de l'isolement et pour minimiser le coût total, il faut quantifier la teneur en radionucléides des colis de déchets afin que les risques radiologiques de chaque colis puissent être assortis aux caractéristiques d'isolement des installations de confinement qui conviennent.

On décrit dans ce rapport un détecteur non invasif de caractérisation des déchets radioactifs qui permet de quantifier la teneur en radionucléides des colis de déchets de faible activité jusqu'au niveau de 9 Bq/g (250 pCi/g). La technique d'essai est fondée sur la spectroscopie du rayonnement gamma passif où la concentration des rayons gamma émetteurs de radionucléides dans un colis de déchets peut être estimée à partir de l'analyse des spectres du rayonnement gamma du colis et de normes étalonnées.

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ABSTRACT

The exploitation of nuclear processes and technology for the benefit of Canadians results in the routine generation of ~12 000 m^3 of solid low-level radioactive waste annually. To protect the public and the environment, this waste must be isolated for the duration of its potential hazard. In Canada, current planning foresees the development and use of a range of storage and disposal facilities exhibiting differing containment capabilities. To demonstrate adequate isolation safety and to minimize overall costs, the radionuclide content of waste items must be quantified so that the radiological hazards of each waste item can be matched to the isolation capabilities of specific containment facilities.

This paper describes a non-invasive, waste characterization monitor that is capable of quantifying the radionuclide content of low-level waste packages to the 9 Bq/g (250 pCi/g) level. The assay technique is based on passive gamma-ray spectroscopy where the concentration of gamma-ray emitting radionuclides in a waste item can be estimated from the analysis of the gamma-ray spectra of the item and calibrated standards.

INTRODUCTION

The routine operation of nuclear reactors for research and electricity production purposes and the widespread use of radioisotopes in industrial and medical applications results in the generation of a variety of radioactive solid wastes. On a volume basis, the largest portion of this waste is classified as Low-level waste (LLW); that is waste items with levels of radioactivity that require neither shielding nor cooling during normal handling or transportation [1]. Typically, such wastes consist of disposable paper, plastics, wood and cleaning materials which are loosely packaged in polyethylene bags. In practice, much of this waste contains little if any measurable radioactivity and in principle a large fraction could be segregated and disposed of as non-radioactive waste.

For the past 39 years at the Chalk River Nuclear Laboratories (CRNL) LLW has been placed into interim storage facilities awaiting disposal at a later date. At present, the interim storage facilities contain approximately 85 000 m^3 of LLW [2]. However, as annual waste volumes have increased, it has become evident that this form of LLW management is not cost effective and land use efficient. By the mid-seventies, LLW handling experience had matured and annual waste volumes (~3500 m^3/year) had become sufficiently large to provide the necessary incentives for alternative LLW management practices.

At that time, a two step program was identified for improving LLW management at CRNL. Firstly, where practical, the volume of future LLW would be reduced prior to storage. This step was realized with the commissioning of incineration and compaction equipment at the Waste Treatment Centre (WTC) in 1982. The incineration and compaction techniques resulted in waste volume reduction factors of 187 (31% of site LLW) and 7.8 (20% of site LLW) respectively in 1984. Thus, use of volume reduction techniques has significantly reduced the need for new interim storage facilities for LLW [3,4].

Secondly, as a long term solution, techniques and facilities for the safe and economic disposal of LLW would be developed. As part of this step, a Waste Disposal Project was established in 1982 to pioneer the transition of CRNL managed waste from interim storage to permanent disposal. Since the bulk of LLW presents only a short-lived hazard, the intent is to optimize disposal procedures by classifying and segregating wastes according to their potentially hazardous lifetimes and to match waste segments to disposal options selected for their isolation and containment abilities [2,5]. From the outset, a key element of the project has been the demonstration of measurement techniques for the radionuclide characterization of waste items.

Quantitative assays of the radionuclide content of bulk materials have traditionally been performed via radiochemical procedures. With this technique, the radionuclides from a representative sample of the assay material are first chemically separated from the material matrix and then analyzed using spectroscopy techniques. Chemical separation and purification steps are time consuming, expensive, laboratory techniques that are unsuitable for fast routine LLW characterization, where rapid assay of many items with a minimum occupational radiation exposure and overall cost is required.

Non-destructive gamma-ray spectroscopy of waste items has been pioneered in the United States over the past twenty years by several groups [6,7,8,9]. Originally, efforts were focussed on the assay of reprocessed fuel wastes for the identification of transuranic elements. Recently, with the announcement of the United States (US) Nuclear Regulatory Commission (NRC) directive 10CFR61 "Licensing Requirements for Land Disposal of Radioactive Waste", efforts have been redirected to the development of assay techniques for packaged LLW prior to shipment from the waste generator to disposal sites [6]. In assaying LLW, most of which has already undergone volume reduction and packaging to minimize storage and shipment costs, complex correction procedures are usually required to allow for the increased sample self-attenuation and density variation effects.

At CRNL, an alternative approach to LLW characterization by gamma-ray spectroscopy has been taken. In accordance with the program goals of segregating wastes according to hazard duration, wastes will be characterized prior to volume reduction and standardized packaging. Such an approach minimizes assay procedures, since a single characterization assay can be used to ensure compliance with activity administrative targets at both waste treatment and disposal facilities. In addition, assay procedures are simplified since self-attenuation and density variation effects are minimal for such low density wastes. However, a potential disadvantage of this approach is the need to assay a greater volume of waste.

BACKGROUND CHARACTERIZATION STUDIES

In 1982, experimental work began to characterize site generated LLW that was being processed at the WTC prior to placement in storage facilities. Waste feed items, on average of 0.04 m^3 volume and 3.9 kg weight, and packaged in polyethylene bags, were monitored for total gamma-ray activity and for gamma-ray isotopic composition. The results of these assays were then compared with the results of radiochemical assays of incinerator ash for verification purposes. The principal radionuclides, their halflives and relative abundances in CRNL LLW are listed in Table 1. On average, about eighty percent of the activity is associated with only five radionuclides: Nb-95, Zr-95, Co-60, Cr-51 and Cs-137 [10].

TABLE 1: GAMMA-RAY EMITTING RADIONUCLIDES IDENTIFIED IN CRNL LLW

RADIONUCLIDE	HALFLIFE (DAYS)	PER CENTAGE OF TOTAL ACTIVITY (%)
Nb-95	35	30.9
Zr-95	64	16.4
Co-60	1924	15.0
Cr-51	28	10.7
Cs-137	10,987	7.1
Ce-144	284	5.0
Zn-65	244	3.5
Ce-141	33	2.6
La-140	1.7	1.9
Ag-110m	253	1.7
I-131	8.1	1.1
Ru-103	40	0.9
Co-58	71	0.7
Ba-140	13	0.6
Cs-134	752	0.5
Ru-106	369	0.5
Fe-59	45	0.5
Sb-124	2.7	0.1
Mn-54	313	0.1

From an analysis of the monitoring and radiochemical data, three main conclusions were drawn. Firstly, a large fraction of the total gamma-ray activity (>90%) is concentrated in a small number of waste items (<10%). This result agrees with past estimates of waste item activity based on monitoring with portable gamma-ray survey meters [11].

Secondly, an acceptable isotopic signature for shipments of low density waste items can be obtained from the monitoring of only the most active waste items using gamma-ray spectroscopy.

Thirdly, non-gamma-ray emitting radionuclides can likely be estimated with acceptable confidence limits from the gamma-ray isotopic signature of current waste feed; that is, wastes associated with fission fuel cycles.

MONITORING CONCEPT

Based on the results of these early monitoring experiments, a decision was made to develop a demonstration, non-invasive waste monitor for use at the CRNL WTC. The monitoring concept chosen combines both gross gamma-ray and gamma-ray spectral monitoring. For each waste item, an initial gross gamma-ray activity measurement is used to determine the activity level of the item. Based on this initial activity measurement, only the most active items are selected for gamma-ray spectral characterization. At the completion of the monitoring session, the gamma-ray spectra are analyzed and the radionuclide content of gamma-ray emitting isotopes is quantified. Next, an estimate of the radionuclide inventory for all items in a given shipment is estimated by scaling up the radionuclide content determined for the most active items by a correction factor determined from gross gamma-ray activity measurements of all shipment items. Finally, estimates of the activity levels of non-gamma-ray emitting radionuclides can be made from an examination of the scaled gamma-ray spectral signature and radiochemically determined correlation factors between non-gamma-ray and gamma-ray emitting radionuclides for the type of LLW monitored.

DESIGN REQUIREMENTS

At the onset of the monitor development program, the following design criteria were established to guide the developers. The monitor should:

- be capable of a measurement resolution of <370 Bq/g (10 nCi/g) for the major isotopes of interest in conformance with the United States Nuclear Regulatory Commission directive 10CFR61,
- be flexible so that wastes of various physical forms, shapes and sizes can be easily monitored,
- permit short assay times per waste item in order not to impair normal waste handling and processing procedures,
- be capable of being operated by non-specialist personnel,
- provide convenient data accumulation and storage facilities compatible with the current conversion of site waste management records to a computer database,
- be designed as a transportable instrument package to allow convenient use at a number of assay sites, and
- be assembled from commercially available equipment wherever possible to minimize development effort and to simplify future maintenance requirements.

EQUIPMENT DESCRIPTION

The waste monitor consists of a partially shielded enclosure that contains the gamma-ray detection equipment, weigh scale platform and liquid nitrogen storage dewar; and a single instrumentation cabinet containing nuclear counting and weigh scale electronics, a computer and an interactive terminal for operators. A block diagram of the equipment configuration is shown in Figure 1 and a photograph of the monitor during preliminary testing at the WTC is shown in Figure 2.

The partially shielded enclosure consists of two 2 cm thick lead walls mounted on moveable steel frames. The walls are normally positioned 60 cm apart for monitoring small sized waste items such as waste bags but can easily be reconfigured with greater spacings to accommodate larger sized waste items. A wall spacing of approximately 1.5 m is shown in Figure 2 to accommodate the monitoring of a compacted bale of waste. Such a shielding enclosure was judged necessary to suppress the high background gamma-ray fields of ~1 μGy/h (100 μrad/h) to be encountered at the WTC monitoring location. The background gamma-ray field is primarily due to the nearby storage of active waste items awaiting volume reduction processing.

Two gamma-ray detection systems are mounted on the shield enclosure frame. For a measurement of the gross gamma-ray activity of a waste item, two NaI detectors are mounted in collimated and back-shielded plugs, one in each enclosure wall. The position of the detector assembly within the collimator is variable so that the effective field of view can be altered to accommodate objects of varying physical size. These detectors are connected to conventional nuclear counting electronics for count accumulation.

The gamma-ray spectral measurement is made with a hyper-pure germanium semiconductor detector. This detector is normally mounted on a crossbar that interconnects the two shielding walls and views each waste item to be assayed from above. The detector is a portable, multi-axis mount model and can be removed easily from the crossbar location and positioned to view the item for assay from alternative views. This flexibility is illustrated with the stand-mounted detector configuration shown in the foreground of Figure 2. A liquid nitrogen dewar with one week cooling capacity is mounted on one side of the shielding enclosure frame. The detector is interfaced to a conventional multi-channel analyzer (Nuclear Data model 65) positioned on top of the supporting instrument cabinet.

Between the shielding walls, an industrial weigh scale is placed for the measurement of the weight of each item to be assayed. This scale has a range of measurement from 0 to 100 kg with a resolution of 0.1 kg.

The single, half-height instrumentation rack is normally positioned adjacent to one of the shielding walls for ready access to the monitoring area for insertion and removal of waste items and control of the monitor functions from the operator display/keyboard interface. The display/keyboard interface has been placed on top of the instrumentation rack at waist level to facilitate convenient equipment control from a standing position.

A Digital Equipment Corporation MICRO-11 computer controls the system. It is interfaced to the gross gamma-ray counting equipment via the IEEE-488 interface standard and to the weigh scale and gamma-ray spectral measurement electronics via RS-232 serial interfaces. Two additional serial interfaces are used for connection of the operator console and a data link via standard telephone lines to the site computing facility.

The computer is used to perform three main functions. Firstly, it controls the acquisition and temporary local storage of data during a monitoring session in accordance with operator commands. Secondly, data analysis programs for peak identification, background subtraction, nuclide identification and quantification are executed with the spectral data from the germanium detector assay measurements. These data reduction functions are normally performed in a background mode simultaneously with the execution of monitoring sequences during an assay session. Thirdly, the computer is used to establish a communication channel to the site central computer during the evening and to control the uploading of daily monitoring data and their archival in the waste management database.

OPERATIONAL DESCRIPTION

The monitor operates under direct control of process operators who must manually insert and remove waste items from the counting position and designate the waste processing option to be selected for each item. The operator interface for the monitor consists of a CRT display for status information and prompting display, and a keyboard for command and data input.

To perform a given function, the appropriate routine is loaded from disk into computer memory and execution is initiated. Throughout the execution sequence, the operator is prompted to select assay parameters and/or command options. All prompting messages display a default value for the particular response requested. To resume execution, the operator enters a new parameter or command option or instructs the program to continue with the default value previously displayed. All operator inputs are examined before use to ensure that they fall within defined ranges. The interactive nature of the routines reduces the requirements for operator training to a minimum and eliminates the need to consult written manuals during normal monitor operation.

The monitor software is partitioned into two routines; a monitoring routine and a data transmission routine. The monitoring routine is used to perform all aspects of assay data acquisition.

The sequence of operations performed in a typical monitoring session is as follows. Following sequence initiation, the operator is requested to enter the current date, time and monitoring location. Next, he is instructed to install a flexible disc in the computer for backup storage of the monitoring data for data safety purposes. The routine then offers the user the ability to tailor the assay parameters for the monitoring session to the specific requirements of the waste to be monitored. For example, assay count durations and the activity level to initiate a complete spectral assay of a given item can be changed from their default values via a prompt/response dialogue.

Three calibration measurements are performed next. Firstly, a calibration check of the weigh scale with known weights is performed and the zero and gain of

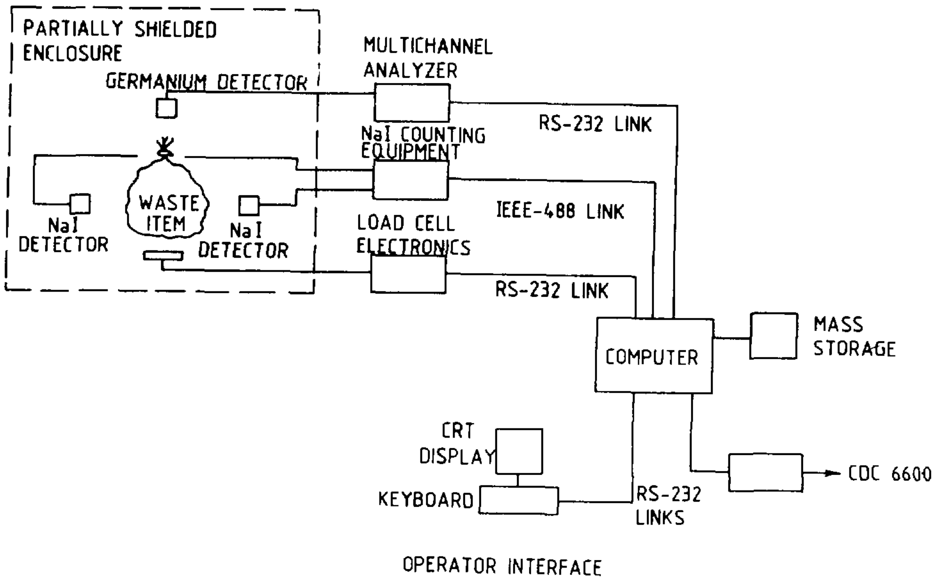

FIGURE 1: CONFIGURATION OF WASTE CHARACTERIZATION MONITOR

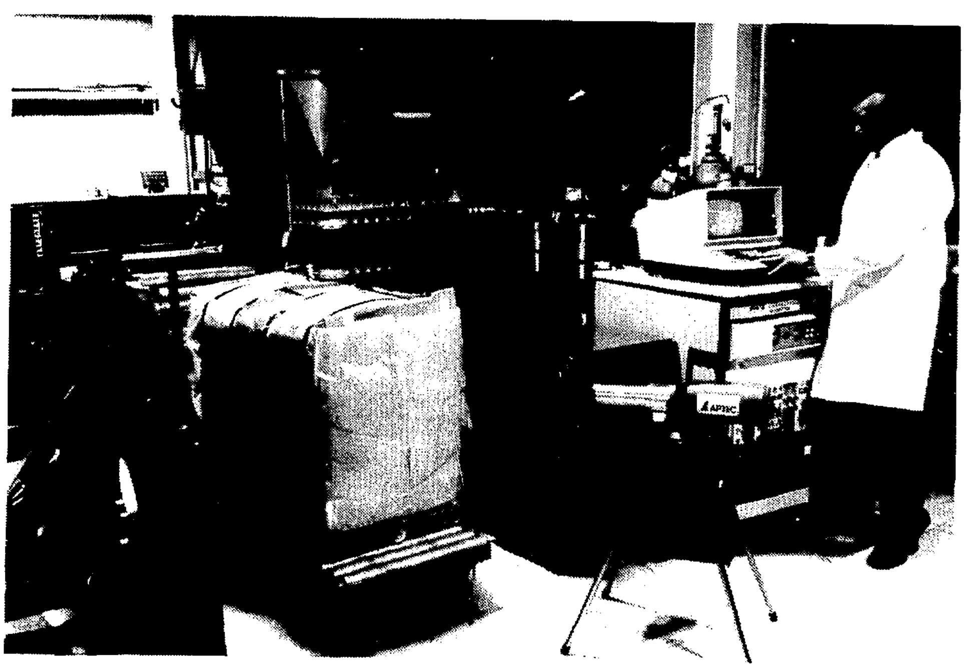

FIGURE 2: WASTE CHARACTERIZATION MONITOR

the scale electronic output is adjusted if necessary. Secondly, a calibration assay of the two radiation detector systems is performed using calibrated radionuclide sources in a matrix of waste material representative of the items to be assayed. Lastly, an assay with the two radiation detector systems is performed, with the monitor position clear of waste items, to establish an initial activity baseline for background correction purposes.

The monitor is now prepared to begin assays of individual waste feed items. The assay of each item begins with the operator being prompted to load a waste item onto the weigh scale in the monitor position. Following acknowledgment from the operator that this action has been completed, the computer initiates both a gross and spectral gamma-ray assay of the item and a measurement of the item weight. After a five second counting period, the count recorded with the gross gamma-ray assay system is compared to a predetermined counting limit identifying those items for which a spectral assay is required. If the gross gamma-ray assay count is greater than the predetermined limit, the spectral assay for the item is permitted to continue to completion, otherwise the spectral assay for the item is terminated and the partial spectral data erased. During the assay count period, the operator is prompted to enter additional information such as the waste item type, source and reference labelling to identify the assay data collected. At this point, the operator must select a waste category in which to route the monitored item. Current categories in use for bagged waste are incinerable, compactible, rejection on physical or activity properties and too active to monitor. Once a category is selected, the assay and reference data for the monitored item are stored in a temporary data file according to waste category. The routine then offers the operator the option of monitoring more waste items or terminating execution.

Following the completion of monitoring a single item via the CRT display (see Figure 3). This display provides a summary on the left side of the waste category designation for previously monitored items, lists on the right side reference and assay data for the last item monitored and displays in the lower portion of the CRT operator prompts.

During a monitoring session, assays of the gamma-ray background are initiated automatically at regular time intervals via computer control or manually by the operator via keyboard command. Data reduction of the spectral assay data is performed, transparent to the operator, in a background mode simultaneously with the monitoring sequence.

At the termination of a monitoring session, the operator is instructed to initiate the data transmission routine. This routine controls the transfer of daily assay information to the CRNL site Control Data Corporation (CDC) 6600 central computer for entry of the data directly into the waste management database. The operator can select to initiate the transfer via an interactive mode on a file (waste category) by file basis or select an automatic transfer mode where the data is transferred during evening hours automatically.

Maintenance activities for the monitor consist of general cleaning of the equipment to minimize the background effects of fixed contamination and weekly replenishment of the liquid nitrogen dewar supply.

PERFORMANCE

Although the monitor has not been fully commissioned and evaluated in a field trial, experimental measurements with monitor subsystems and simulations of the monitoring sequences have characterized the limits to monitor performance.

85-05-14 **MONITOR STATUS** **13:42**

SUMMARY			CURRENT ITEM		
CATEGORY	**CURRENT BIN**	**MONITORED ITEMS**	**TYPE:**	**BAG**	
			SOURCE:	**BLDG 150**	
			REFERENCE:		
INCINERATE	**3**	**42**			
COMPACT		**35**	**MASS (kg):**	**4.2**	
			VOLUME (m^3):	**0.04**	
REJECT - PHY.		**3**			
REJECT - ACT.		**1**	**NET NaI COUNTS:**		**29 250**
			BACKGROUND COUNTS:		**25 198**
REJECT - TOO ACT.		**0**			
			SURVEY METER (mGy/h):		**0.01**
BALE		**2**			
ASH DRUM		**0**			

(PROMPTING MESSAGE AREA)

FIGURE 3: DISPLAY OF MONITOR SEQUENCE STATUS

The range and resolution of waste item activities that the monitor can accommodate is a function of the source to detector separation, background gamma-ray activity levels, assay count durations and counting electronics capabilities. The lower range limit or resolution can be best expressed by an estimate of the minimum detectable activity for a given radionuclide. This quantity is defined as the minimum activity that can be detected with a probability of 95% greater than the pseudo-activity due to random natural variation of the background. For a background activity level of 1 μGy/h (100 μrad/h), a source to detector separation of 40 cm and an assay count duration of 60 seconds, the monitor is capable of minimum detectable activities of less than 37 kBq (1 μCi) per waste item for the five most common radionuclides found in CRNL LLW. For a typical 4 kg waste bag, this implies the capability to measure radionuclide activity concentrations of less than 9 Bq/g (250 pCi/g) which is more than an order of magnitude lower than the US NRC 10CFR61 standard of 370 Bq/g (10 nCi/g).

The upper range limit for the standard 40 cm source to detector separation is approximately 44 kBq/g (1.2 μCi/g). Waste items with specific activity contents up to 440 kBq/g (12 μCi/g) can easily be accommodated for assay by increasing the source to detector separation through the movement of the shielding assembly walls.

The throughput capability of the monitor is determined by the assay duration and the number of active items requiring full spectral characterization in a given monitoring batch. For gross gamma-ray activity and spectral assay durations of 30 and 86 seconds respectively and a waste distribution where only 10% of waste items require full spectral characterization, a monitor throughput of approximately 100 waste items per hour is achievable.

PROJECT STATUS

The demonstration waste characterization monitor is at an advanced stage of development. All hardware has been delivered and assembled except for the germanium detector and supporting electronics. These outstanding items are scheduled for delivery in the later part of June 1985. Of the two software packages, the monitoring routine is still undergoing development while the data transmission routine was completed and tested in the fall of 1984. Monitor calibration and initial field evaluation trials are currently planned for August of 1985. Completion of instrument commissioning at the CRNL WTC is scheduled for 1985 October 1.

The experience gained in using the monitor in an operations environment will be gathered over a one year period. Following this period, any deficiencies in monitor performance will be corrected and the expansion of the current system or the construction of additional monitors for characterization of other waste feeds will be considered.

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