# RAPID MONITORING OF GASEOUS FISSION PRODUCTS RELEASED FROM NUCLEAR POWER STATIONS

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## ABSTRACT

Quantitative monitoring of gaseous fission products by rapid, *in situ* measurements was conducted around nuclear power stations in Taiwan. A portable high-resolution germanium detector with portable a multichannel analyzer was used in the field monitoring work. The detecting unit was calibrated using activated Ar, Kr, and Xe isotopes dispersed in a large chamber to obtain absolute efficiency curve in terms of counts per gamma per $m^3$ versus gamma-ray energy. The calibrated detecting unit was brought to the nuclear power plants for *in situ* monitoring for both normal operation and nuclear accidental drill. In a typical four-hour measurement, the detection limits for most Kr and Xe fission product isotopes are only 0.0028% ~ 0.98% of the derived air concentration imposed by the local authority. The dose rate caused by gaseous radioisotopes released from nuclear power stations and dispersed to the surroundings can be quantitatively monitored in a short period using this portable unit.

## INTRODUCTION

Monitoring of individual gaseous fission products in the environment, due to releases from a nuclear power station during normal operation, or after an accident, using high resolution gamma-ray spectrometers became possible when semiconductor germanium detectors were commercially available (Beck, *et al.*,1972). The earlier germanium detectors had to be housed in a temperature and humidity controlled laboratory, and gaseous fission products, sampled from around a nuclear power station, had to be transferred to the laboratory for counting. Earlier studies using portable germanium gamma-ray spectrometers to perform quantitative *in situ* measurements of gaseous fission products have been reported; however, details of the absolute calibration for the portable spectrometer were only given for point sources at various angles (Gogolak and Miller, 1974).

An important aspect of environmental monitoring is the measurement of radiation dose due to external exposure to photons emitted by each gaseous fission product dispersed in the air. In this work, the detector efficiency of a portable germanium gamma-ray detecting unit was determined using activated argon, krypton, and xenon radionuclides dispersed in a sealed chamber. The detection sensitivity was established for each specific full-energy peak, and the detection limit for each gaseous fission product was evaluated. A field application using the calibrated portable unit in the area surrounding all nuclear power stations in Taiwan is presented. The detection limits are compared with those regulatory values imposed by the local authority.

## EXPERIMENTAL

During normal operation of a nuclear power station or in the early stage of an accident, some gaseous fission products are released to the surroundings (Gogolak,1984). Assuming the released radionuclides of Kr and Xe are continuously dispersed in the infinite half-space homogeneously and have reached equilibrium state with respect to their half-lives, the detector efficiency, in terms of full energy peak count rate at constant radioactive concentration (cps per $Bq/m^3$), can be determined by calibrating the count rate

of each full-energy peak of interest in a chamber with known concentrations of gaseous fission products. The response in the finite size of the sealed chamber can be corrected for the infinite half-space by a geometric factor related to the limit radius of the chamber (Chung and Tsai, 1996).

Enriched rare gases of stable Ar, Kr, and Xe, sealed in quartz tubes, were irradiated in the 1 MW Tsing Hua Open-pool Reactor (THOR) for three hours. Immediately after the irradiation, the radioactivity in the sealed tube was measured using reference line sources for absolute activity calibration. Each tube was broken open inside the sealed chamber which had a radius of 8.3 meters, in which a portable high-purity germanium (HPGe) detector was positioned at the center, above the chamber floor. The 30% relative efficiency HPGe detector (DSG model DGC3019), with a system resolution of l.9 keV at 1332 keV, was coupled to a portable multichannel analyzer and a notebook computer.

The detector efficiencies for each peak of interest, in terms of counts/gammas-$m^3$ or cps per Bq/$m^3$, can be determined from the known concentration of the rare gaseous radioisotopes. The measurements were extended to 10 days after irradiation in order to acquire the weak gamma-ray peaks originating from the longer-lived $^{131,133m}$Xe radioisotopes, while the leakage from the chamber (accounted for 35%) was corrected. The efficiency curve is fitted from the calibration data (Figure 1), under the condition that the rare gas radionuclides are dispersed homogeneously in the infinite open air and in equilibrium state. The detecting efficiencies for other rare gaseous radioisotopes, in particular those neutron-rich fission products, can be interpolated from the efficiency curve.

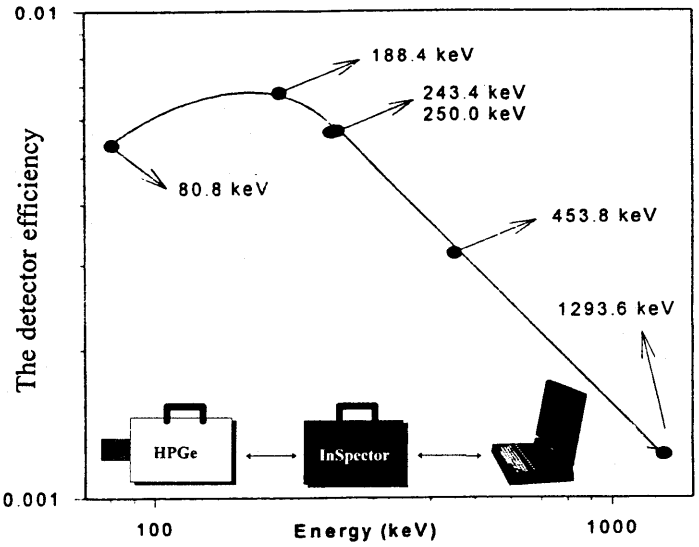


**Figure 1** The detector efficiency curve for the portable HPGe detector(DSG model DGC3019) immersed in open air with homogeneously dispersed rare gas Ar and Xe radionuclides.

The dose rate conversion factors for external exposure to photons emitted from radionuclides dispersed in air have been evaluated (Kocher, 1983) in terms of nSv/h per Bq/$m^3$. Hence, the detection sensitivity (DS) for each radionuclide (nSv/h per cps) can be calculated as dividing the dose rate conversion factor by the detector efficiency and the gamma-ray branching ratio. The DS values for the gaseous fission products frequently observed around a nuclear power station are listed (Table 1). They range from 0.054 nSv/h per cps for fission product $^{131m}$Xe to 11.86 nSv/h per cps for $^{89}$Kr.

**Table 1** Detection sensitivity(DS) for gaseous fission products released from a nuclear power station by use of a portable DSG model DGC3019 HPGe detecting unit.

| Radio-nuclide[1] | Fission Yield(%)[2] | Decay properties[3] | | Detection sensitivity DS(nSv/h/cps) |
|---|---|---|---|---|
| | | Half-life | Major γ−ray(keV) | |
| $^{85m}$Kr | 0.285 | 4.48 hr | 304.9 | 0.956±0.031 |
| $^{85}$Kr | 1.33 | 10.7 yr | 514.0 | 0.031±0.002 |
| $^{87}$Kr | 2.53 | 1.27 hr | 402.6 | 5.292±0.218 |
| $^{88}$Kr | 3.67 | 2.86 hr | 2392.1 | 1.131±0.041 |
| $^{89}$Kr | 4.99 | 3.18 min | 220.9 | 11.858±0.539 |
| | | | | |
| $^{131m}$Xe | 0.031 | 11.77 dy | 163.9 | 0.054±0.002 |
| $^{133m}$Xe | 0.071 | 2.19 dy | 233.2 | 0.191±0.010 |
| $^{133}$Xe | 6.44 | 5.245 dy | 81.0 | 0.682±0.022 |
| $^{135m}$Xe | 0.678 | 15.65 min | 526.6 | 2.483±0.126 |
| $^{135}$Xe | 6.45[4] | 9.10 hr | 249.8 | 1.453±0.048 |
| $^{137}$Xe | 6.21 | 3.82 min | 455.5 | 1.404±0.072 |
| $^{138}$Xe | 5.89 | 14.17 min | 258.3 | 6.989±0.281 |

Note: (1) Rare gaseous radionuclides with half-life less than one minute are excluded.
(2) Fission yields are taken from the thermal neutron induced fission on $^{235}$U.(Crouch, 1977).
(3) Decay properties are taken from (Lederer and Shirley, 1978).
(4) Actual yield is lower, depending upon the operation period because $^{135}$Xe has huge thermal neutron absorption cross section.

## FIELD APPLICATION

Although the portable HPGe gamma-ray detecting unit is calibrated under the assumption that the immersed radionuclide in air is homogeneously dispersed, the converted dose rate sensitivity, however, represents both original and scattered photons impinging on the HPGe detector regardless of their spatial-time distribution. To demonstrate its capability, the portable HPGe detecting unit was taken to the surroundings of all three nuclear power stations in Taiwan for rapid *in situ* monitoring of dispersed gaseous fission products during their full power operation. Typical counting period is set for four-hour at the position 100 meters away from the offgas exit at the reactor containment. The gamma-ray spectra taken from each nuclear power station are illustrated (Figure 2).

During reactor full power operation, air is normally not in contact with reactor core neutrons. Likewise, it is unlikely that failed fuels be utilized in the nuclear power reactors. Hence, activated rare gas radioisotopes and gaseous fission products are not expected to show up in the gamma-ray spectrum, except perhaps those peaks associated with natural radionuclides (such as 1461 keV of $^{40}$K) and with nuclear waste (such as 1332 keV of $^{60}$Co) in the figure. Although the gaseous fission products are unlikely to leak and be detected during full power operation, it is necessary to assess the limit of detection using the current portable unit in case of nuclear accident.

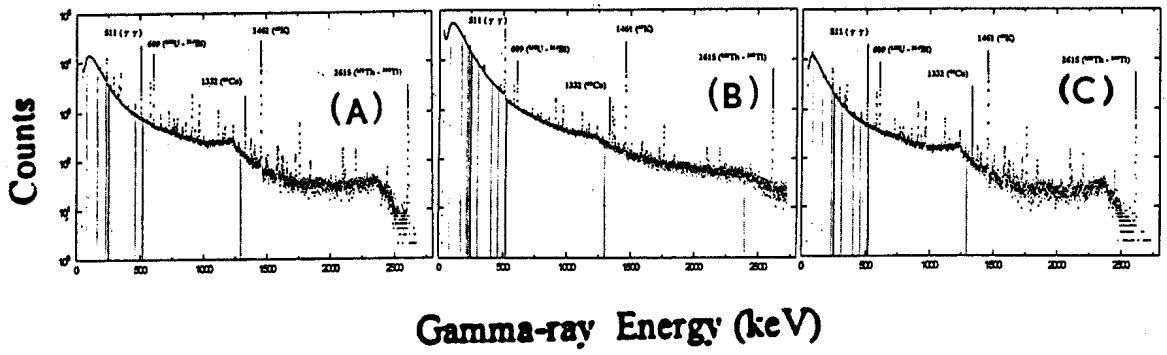


**Figure 2** *In situ* gamma-ray spectra in (A) the first nuclear power station with two 636 MWe BWR units, (B) the second nuclear power station with two 985 MWe BWR units, and (C) the third nuclear power station with two 951 MWe PWR units; a DSG model DGC3019 portable HPGe detecting unit is utilized with four-hour counting period for all field measurements. Shaded areas below spectra are the regions of interest for gaseous fission products.

The detection limit (DL) can be assessed as 4.65 x DS x $(BCR/t)^{1/2}$, where BCR is the background count rate at the full energy photopeak and t is the counting period(Cruie, 1968). In the four-hour monitoring period near the reactor site boundary, it gives DL values for all gaseous fission products and the results are listed (Table 2) for each nuclear power station. These DL values are lower than the dose rates reported (Gogolak, 1984) at 450 meters from a BWR in full power operation with minor leakage by factors varying from 48 (for $^{135m}$Xe) to 9,200 (for $^{88}$Kr). Hence, it is highly feasible to use the *in situ* measurements for rapid determination of dose rate contributed from individual gaseous fission product dispersed in air.

The detection limit of the portable detecting unit used in this work is far less than the dose rate of derived air concentration (DAC), converted from the annual limit of intake assessed for gaseous fission products (ICRP,1993). Hence, the portable HPGe detecting unit can be used for field measurements ranging from normal reactor operation to a nuclear accident. In a typical four-hour measurement, the detection limit for most Kr and Xe gaseous fission products are only 0.0028% ~ 0.98% of those DAC values imposed by the local authority. However, the portable detecting unit has limitations in high count rate environments; it will lose spectrometric characteristics when the system deadtime reaches 30%.

The operation of this portable HPGe gamma-ray detecting unit, with a total weight of 14 kg, requires no supporting equipment and it can operate continuously for two days between liquid nitrogen refills. In an emergency situation, the ability of this portable detecting unit to make rapid *in situ* measurements of the dose rate due to individual airborne gamma-emitters could prove extremely valuable, in particular when the release rates of gaseous fission products to the environment are otherwise uncertain or even unavailable in the case of an accident.

**Table 2 Detection limit for gaseous fission products released from the nuclear power stations (NPS) in Taiwan using a portable DSG model DGC3019 HPGe detecting unit for *in situ* field measurement.**

| Gaseous Fission product | DAC[(1)] (nSv/h) | Detection Limit (nSv/h) | | |
|---|---|---|---|---|
| | | No.1 NPS | No.2 NPS | No.3 NPS |
| $^{85m}$Kr | 1,400 | 0.039 | 0.097 | 0.044 |
| $^{85}$Kr | 19 | 0.00077 | 0.00283 | 0.00101 |
| $^{87}$Kr | 131 | 0.148 | 0.362 | 0.183 |
| $^{88}$Kr | 114 | 0.0047 | 0.0087 | 0.0064 |
| $^{89}$Kr | N.A. | 0.671 | 1.819 | 0.808 |
| $^{131m}$Xe | 68 | 0.0040 | 0.0113 | 0.0048 |
| $^{133m}$Xe | 113 | 0.0102 | 0.0273 | 0.0129 |
| $^{133}$Xe | 102 | 0.0636 | 0.1753 | 0.0943 |
| $^{135m}$Xe | 92 | 0.0528 | 0.1086 | 0.0665 |
| $^{135}$Xe | 91 | 0.0720 | 0.1895 | 0.0870 |
| $^{137}$Xe | N.A. | 0.0345 | 0.0819 | 0.0430 |
| $^{138}$Xe | 90 | 0.322 | 0.878 | 0.387 |

Note: (1)Derived Air Concentration (DAC) for gaseous fission products is derived from the dose conversion factor and annual limit of intake (ICRP, 1993).

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