# Potential Applications of Prompt Gamma Neutron Activation (PGNA) in the Nondestructive Testing of Concrete Structures

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## ABSTRACT

Prompt Gamma Neutron Activation (PGNA) is a spectroscopic technique for the elemental analysis of materials. The elements are identified by characteristic gamma-rays emitted from the target material while it is being bombarded with neutrons. In nondestructive (NDT) of concrete structures, the PGNA technique is distinguished from other methods by the fact that it can measure the chemical aspects of the concrete's condition, as opposed to its mechanical or structural condition. The basic method consists of irradiating a given part of a concrete structure with neutrons from a small portable californium neutron source. The neutrons penetrate within the structure and interact with the elements present to produce characteristic gamma-rays. These gamma-rays are then detected and counted. The data can then be interpreted to yield information about the spatial distribution of characteristic elements at the sampled location. By moving the apparatus from point to point along the structure, it is possible to map the spatial distribution of the elements. These maps, in turn, can be interpreted to yield valuable insights about the condition of the concrete structure. Several aspects of concrete composition and condition can be investigated with PGNA. Probably the most significant application is in the detection of chlorides. Chlorides are easily detected by PGNA owing to the very large thermal neutron capture cross section of chlorine. It has been established that chlorides do accelerate damage to concrete by promoting the rusting of reinforcements. It should be possible to use PGNA to map chloride distributions in reinforced concrete nondestructively.

## KEYWORDS

Prompt gamma-rays; neutron activation; nondestructive testing; concrete structures, moisture; chlorides.

## INTRODUCTION

The degradation of building materials such as concrete is a major problem in the Arabian Gulf region. Such degradation often arises from the presence of contaminants in the constituent materials. A knowledge of the presence and distribution of these contaminants might be needed for effective treatment and preservation. Existing methods for the evaluation of building conditions are generally limited to visual inspection and destructive testing of small samples. Some nondestructive methods can identify cracks and voids, but cannot provide information on the possible causes of these defects. Other nondestructive methods, such as electrical conductivity and neutron thermalization, can provide semiquantitative information about moisture, but only at the surface. There is a need in civil engineering for methods that can measure nondestructively the condition of existing reinforced concrete structures, particularly behind surface finishes. The technique of Prompt Gamma Neutron Activation could help to meet this need. This technique could be used for the in-depth nondestructive testing of concrete structures to determine the distribution of contaminants in building walls, particularly water and soluble salts. Water is a major cause for deterioration, both by acting as a solvent for various building materials and by exerting stresses on the structure during freeze-thaw cycles. The soluble salts, notably chlorides, damage the structure either by periodically dissolving and crystallizing within the pores or by promoting corrosion. The technique of PGNA is particularly sensitive to major elements in water and chlorides, and it could measure them nondestructively to depths of 10-25 cm in typical construction materials.

## INSTRUMENTATION

Neutrons can interact with nuclei of materials in two ways to yield characteristic prompt gamma-rays. First, fast neutrons ( Energy $\geq$ 1 MeV) can scatter inelastically from a nucleus, losing energy in the process and exciting the nucleus to a higher energy state. Immediate de-excitation of the nucleus produces gamma-rays characteristic of the excited nucleus. Second, neutrons with energy around the thermal value (0.025 eV) can be captured by a nucleus. A compound nucleus is formed in an excited state and the prompt de-excitation of this excited nucleus causes the emission of characteristic capture gamma-rays. These two prompt gamma-ray processes provide immediate results during the irradiation and require relatively low strength neutron sources which leave negligible residual radioactivity in the material after the measurement. The presence of a given gamma-ray line with a particular energy is used for qualitative analysis, and the intensity of the line is used for quantitative analysis. For the purpose of nondestructive testing of extended building structures, the technique of PGNA is comprised essentially of three parts: a source of neutrons of suitable energy and intensity, a sensitive radiation detector with associated electronics, and a concrete structure to be analyzed. A schematic diagram showing such a system is shown in Fig. 1.

**Neutron Sources**

The two types of neutrons sources which are of practical value for the nondestructive testing of concrete structures are particle accelerators (neutron generators) and radioisotope neutron sources. Particle accelerators use a beam of deuterons accelerated to about 150 kV onto a tritium or deuterium target providing relatively high fluxes of fast monoenergetic neutrons. The tritium target reaction produces neutrons of about 14 MeV, and intensities of about $10^{11}$ n/s. These yields of fast neutrons are satisfactory to accomplish a broad range of elemental determinations with high sensitivities. Accelerator neutron sources are the automatic choice for high-threshold-energy reactions such as the $^{16}O(n,p)^{16}N$ reaction. The main advantage of the accelerator source, other than the higher intensities, is that it can be turned off. The radiation shields required, however, are relatively massive, and personnel protection from exposed high voltages of some accelerators is an additional cause for concern. . The combined application of nanosecond timing techniques with PGNA using the inelastic scattering of 14.6 MeV neutrons was successfully applied to the determination of all major elements ( abundance > 1%) in a bulk sample of concrete [Hlavac and Oblozinsky (1987)]. It was observed in this work that about 90% of γ-rays of interest for prompt γ-ray analysis are emitted from a 12 cm thick surface layer in less than 10 ns after the neutrons hit the sample.

On the other hand, in thermal-neutron activation analysis, the accelerator source is not needed, since the equivalent low-energy neutron flux can usually be provided by a radioisotope neutron source. The main advantages of radioisotope neutron sources in nondestructive testing of extended structures are: physically small size and rugged construction, flux stability, independence of external power, long useful life, and flexibility in source dimensions and shape to afford optimum design in the activation geometry to suit the application. Although the neutron emission cannot generally be turned off, shielding is easily achieved owing to the compactness of the source and low energy and intensity of the radiation. Radioisotope neutron sources are available in a variety of chemical forms, offering a choice of average neutron energies and intensities to best fit the intended application. Some properties of the more commonly used sources are given in Table 1 [Knoll (1983)].

For those application requiring only low-energy neutrons, i.e., thermal-neutron activation analysis, the photon-neutron source may be used. These sources comprise a high-energy γ-ray emitting radioisotope which is surrounded by a beryllium target. The neutrons are produced in the reaction:

$$^{9}Be_4 + \gamma \text{ ---------> } 2\alpha + n - 1.67 \text{ MeV} \qquad (1)$$

A unique feature of these sources is that they can be " turned-off " by separating the γ-ray source from the target. These sources can provide thermal neutron fluxes in excess of $10^8$ n/cm$^2$-s. Two major drawbacks are the short half-life of the source (60.4 days), which necessitates frequent replacement, and the substantial amount of heavy shielding material required to reduce the γ-radiation to permissible levels.

Table 1. Properties of some radioisotope neutron sources

| Source | Half-life | Source strength n/s-Ci, | n/s-g | Average neutron Energy [MeV] |
|---|---|---|---|---|
| (γ,n) | | | | |
| $^{124}$Sb-Be | 60.4 days | $1.6 \times 10^6$ | $1 \times 10^{10}$ | 0.024 |
| (α,n) | | | | |
| $^{210}$Po-Be | 138.4 days | $2.5 \times 10^6$ | $1 \times 10^{10}$ | 4 |
| $^{238}$Pu-Be | 86.4 years | $2.5 \times 10^6$ | $5 \times 10^7$ | 4 |
| $^{241}$Am-Be | 458 years | $2.2 \times 10^6$ | $7 \times 10^6$ | 4 |
| $^{226}$Ra-Be | 1602 years | $1 \times 10^7$ | $1 \times 10^7$ | 4 |
| Spontaneous fission | | | | |
| $^{252}$Cf | 2.646 years | $4.4 \times 10^9$ | $2 \times 10^{12}$ | 2 |

A second class of radioisotope neutron source which is used in many applications is the alpha-initiated (α,n) neutron source. These sources are constructed as an intimate mixture of an alpha-emitting radioisotope and a target material such as beryllium (Be). The neutrons are generated following an interaction between the alpha particle and the Be nucleus, and possess energies up to about 10 MeV, with an average value of about 4 MeV:

$$^{9}Be_4 + \alpha \text{ ----------> } ^{12}C_6^* + n + 5.71 \text{ MeV} \quad (2)$$

The alpha particle emitters most commonly employed in neutron sources are $^{210}$Po, $^{239}$Pu, and $^{241}$Am. Other target materials, such as lithium (Li), boron (B), or fluorine (F), may be used instead of Be to give lower average neutron energies. The main advantage of the (α,n) source is its ability to activate those elements (e.g. silicon and fluorine) whose reaction thresholds occur in the few MeV region. At the same time, the neutron energy is low enough so that a useful thermal-neutron flux can be obtained by moderating the neutrons in and around the building structure. Neutron outputs from these sources can reach $10^8$ n/s. This is largely dictated by economic and shielding considerations. Shielding for this type of sources is relatively less massive than for (γ,n) sources, since the associated γ-ray emission is low relative to the neutron output. In addition to biological considerations, shielding designs are also dictated by the stringent requirements of the

radiation detector, especially from the 4.43 MeV γ-ray line associated with the decay of the excited $^{12}C$ nucleus.

A third type of neutron source is $^{252}Cf$, a spontaneous fission neutron source. No target material is required with the source, since the radioisotope itself emits the neutrons. This type of source provides a prolific neutron output; 1 μg of $^{252}Cf$ produces about 2 x $10^6$ n/s. The average neutron energy is about 2 MeV, which for thermal-neutron applications may be an advantage because the neutron flux is more easily moderated. $^{252}Cf$ sources used in the analysis of masonry buildings using PGNA have typical masses in the 1-10 μg range.

**Detection System**

Two types of radiation detectors are suitable for use in activation analysis of building structures-the scintillation counter, and the high purity germanium detector. These detectors yield output electronic pulses which are proportional in magnitude to the absorbed energy of the incident gamma radiation. selective gamma-ray detection is thus readily achieved using linear amplifiers and pulse-height discrimination devices. Scintillation detectors with large-volume NaI(Tl) crystals have a high detection efficiency for energetic gamma radiation and have simple operational requirements. They are also less sensitive to neutron damage. The pulse-height energy resolution of the detector, however, is often a limitation for multi-element determinations. HPGe detectors, on the other hand, have a very high intrinsic energy resolution , but are less efficient. Moreover, they must be operated at liquid-nitrogen temperature. These detectors have an energy resolution of about 2 keV at the 1330 keV gamma-ray line of $^{60}Co$.

Irradiation of germanium detectors by neutrons, particularly fast neutrons, can give rise to the displacement of atoms from their lattice positions in the crystal forming an interstitial-vacancy pair. These are particularly effective traps and if germanium detectors are used in neutron fields for some length of time the spectrum will deteriorate because of the increasing proportion of these defects. Table 2 lists the threshold fast neutron dose above which resolution degradation can be expected for a number of different types of detectors [Chao (1993)].

Table 2. Fast neutron radiation damage thresholds

| Detector type | Relative efficiency | Threshold fast neutron dose [n/cm$^2$] |
|---|---|---|
| p-type | 20% | 2 x $10^8$ |
| | 70% | 1 x $10^7$ |
| n-type | 30% | 4 x $10^9$ |
| | 70% | 1 x $10^8$ |

The n-type germanium detector is substantially more tolerant of radiation than the p-type detector and is recommended for use in PGNA. Moreover, large detectors are much more susceptible to damage. When using a detector in PGNA, it may be better to trade off efficiency against a better damage tolerance and longer detector life. Radiation-damaged detectors can be repaired by some manufacturers and some independent companies provide a repair service. Apart from the loss of efficiency in p-type detectors the performance of repaired detectors is likely to be almost as good as new. There appears to be no limit to the number of times a detector can be annealed. If an n-type detector is only slightly impaired, warming to room temperature and then recooling may improve the resolution to some extent. However, the general advice would be to maintain the detector at liquid nitrogen temperature until proper repair can be undertaken. Normally a detector would be returned to the manufacturer for annealing. If the neutron damage is a predictable certainty, as in PGNA, rather than an unfortunate accident, there is now a self-help option. Taking advantage of the demountable detector, format detectors can now be bought in a capsule with a built-in heating element to allow in situ annealing. Repair can be accomplished within a matter of hours.

Cooling is most conveniently provided by liquid nitrogen (boiling point 77 K). For field use, the manufacturers now provide portable $\gamma$-ray spectroscopy systems based on HPGe detectors with over 8 hours of actual operating time. The Ge crystal is cooled by liquid nitrogen contained in a portable dewar connected at the base of the detector. The system comes with a Laptop computer, built-in computer parallel-port interface, 16k and 5-$\mu$s ADC, single-cable operation, and MCA emulator software [EG&G Ortec (1985)].

## APPLICATION

Neutrons are very useful as probes for nondestructive examination of extended structures because they can travel long distances before interacting with the nuclei of the media. Depending on the type of material, neutrons can have a mean free path of tens of centimeters to meters. The number of gamma-rays originating from a unit volume in a structure and counted in a unit solid angle by the detector is given by [Clark et al. (1982)]:

$$I_\gamma = \varepsilon\,(\mathbf{R})\,\kappa\,C(\mathbf{R})\,\phi_{th}(\mathbf{R})\,e^{-\mu R} / 4\pi R^2 \qquad (3)$$

where $\mathbf{R}$ ( R) is the vector (distance) to the volume probed with reference to the detector, $C(\mathbf{R})$ is the mass concentration of the element of interest at $\mathbf{R}$, $\phi_{th}(\mathbf{R})$ is the thermal neutron flux in the unit volume, $\varepsilon\,(\mathbf{R})$ is the efficiency of the detector for that particular gamma-ray energy emanating from $\mathbf{R}$, $\mu$ is the gamma attenuation coefficient, $\kappa$ is a constant and:

$$\kappa\,C(\mathbf{R}) = \sigma\,Y\,n(\mathbf{R})\,A_w/N_A \qquad (4)$$

where $\sigma$ is the thermal-neutron absorption cross section, Y is the gamma yield for the gamma-ray energy of interest, n(**R**) is the number density of the element at **R**, $A_w$ is the atomic weight of the element, and $N_A$ is Avogadro's number. A comprehensive listing of the analytical sensitivity of PGNA to all elements is given in Duffey et al. (1970) and Senftle et al. (1971). Eq. (3) would have to be integrated over the effective volume of the structure probed by the neutrons. In order to carry out such an integral, however, it is necessary to specify the spatial distributions of $\varepsilon$, C, and $\phi_{th}$. Since the latter is generally a complicated function, it is generally not possible to solve for the integral of Eq. (3) analytically. Instead a numerical model is used. Indeed, the computation of this flux from first principles described by the Boltzman transport equations is extremely difficult except for very simple cases. In the case of PGNA of extended concrete structures with the transmission configuration shown in Fig. 1, the thermal neutrons produced by the source itself are largely captured by a few tens of cm within the target material. Thus at larger distances from the source, the thermal neutron flux is essentially a function of the slowing down of neutrons in the fast flux. This slowing down of fast neutrons is dependent largely on the presence of H, Si, O, and Cl in masonry materials. An accurate evaluation of the integral of Eq. (3) could only be accomplished using Monte-Carlo simulation [Clayton (1982), [Clark et al. (1982)]. While the intensity of a gamma-ray line can be used for quantitative analysis, there is no general linear relation between the intensity of a line and the concentration of an element in the material. Other factors affecting the intensity are the bulk density, the amount of hydrogen, and the presence of elements with high neutron capture cross sections. Neutron and gamma-ray transport calculations can be used in some cases to model a particular experiment and to obtain absolute elemental compositions from measured gamma-ray line intensities. However, in many cases of interest for building diagnosis only the relative changes in intensity of a given element are needed for successful interpretation. Compensation for the variation in gamma-ray intensities can be achieved by taking ratios of intensities for each element relative to that of an element, such as silicon, that can be assumed to remain constant in concentration for different measurements.

A major concern about the applicability of PGNA to the analysis of concrete structures will be the influence of the variability of the physical properties of the masonry on the nuclear properties that determine neutron and gamma-ray transport. These major properties are the scattering cross-section and the absorption cross section for thermal neutrons. For gamma-ray transport, it is the attenuation coefficient. Masonry properties can vary in many ways due to the inherent variability in the physical and chemical properties of concrete, the nature of the raw materials, and the process of manufacture. Whether for neutrons or gamma-rays, the characterization of nuclear properties begins with the calculation of appropriate macroscopic cross-sections from the equivalent microscopic cross-sections and the number density of the elements in the material. The general form for a macroscopic cross-section is:

$$\mu = \sigma \rho N_A / A_w \qquad (5)$$

where $\sigma$ is the microscopic cross-section for the interaction, $\rho$ is the density of the element, $N_A$ is Avogadro's number and $A_w$ is the atomic weight of the element. For mixtures of elements in a single phase, the overall mass attenuation coefficient is given by:

$$\mu = \Sigma \rho w_i [\mu / \rho]_i = \Sigma \rho_i [\mu / \rho]_i \quad (6)$$

where $w_i$ is the weight percent of the ith element in the mixture and $\rho_i$ is the partial density.

The cross-sections of concrete are likely to be dominated by Si and O which might not vary much and, therefore, these cross-sections will not be very sensitive to variations in the concentrations of the fluxes. Even though on a large geographic scale, the wt percentages of the fluxes can vary by an order of magnitude, the range should be much narrower on an individual structure, where the concrete mix comes from a local supplier. Consequently, the variations in neutron fluxes will be primarily due to water and salt only. It will then be possible to use a single nominal concrete composition for transport calculations. The mean free paths of concrete are likely to be such that the structure can be treated as a homogeneous material. The effect of porosity will simply be a scale factor for the concrete nuclear properties. The other effect of porosity will be to set an upper bound on the salt or moisture that can be found in concrete. Finally, water in concrete will act as a moderator because of its very high scattering cross-section. Its effect on thermal neutron absorption will not be as significant. In contrast, Cl will be important as both moderator and as an absorber.

## CONCLUSION

The favorable nuclear properties of hydrogen and chlorine in prompt-gamma neutron activation analysis make the technique of PGNA potentially useful in the nondestructive testing of concrete structures. The availability of affordable small neutron sources requiring minimum shielding and portable, light-weight, and compact detection systems make the technique suitable for field use. The technique should prove accurate and sensitive when combined with a good mathematical model that helps to calibrate the setup.

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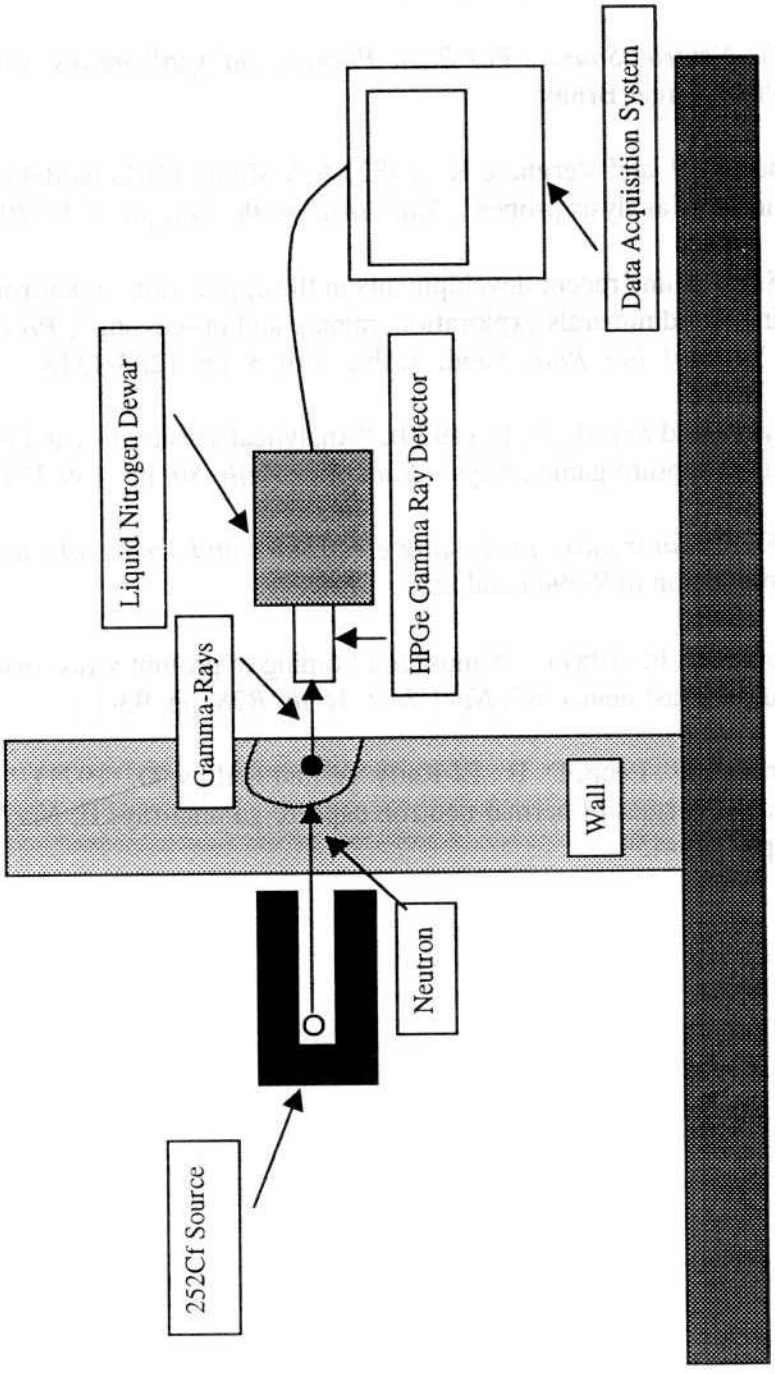


Fig. 1. Schematic setup in transmission mode for the analysis of Concrete structures using PGNA