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Radiological consequences of fly-ash in building materials

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RADIOLOGICAL CONSEQUENCES OF FLY ASH IN BUILDING MATERIALS

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ABSTRACT

The main use of pulverised fuel ash (PFA) from coal fired power stations in building materials is obtained by substituting part of the cement by PFA. A second way is to sinter the PFA to grains, a light weight aggregate (LYTAG) and use it as a filler in concrete. In this case the radiological consequences of gamma radiation in dwellinghouses are much larger than in the first case. It took 7 yaers (from 1982 to 1989) to investigate the total radiological consequences of living in a Lytag-concrete house.
Permisssion of the Ministry of Housing, Spatial Planning and Environment of the Netherlands for the use of Lytag-concrete in dwellings, based on the Stand-Still principle, was recieved in 1989.
The results of the measurements of the concentrations of uranium and thorium and their progeny in fly ash from power stations at the Vasim factory at Nijmegen (NL) are reported.
Radiological consequencies for PFA workers are reported.
The radioactivity of Desulphurisation Gypsum is compaired with the activity of natural gypsum.

HISTORICAL SURVEY

From the beginning of this century in the Netherlands the electricity is produced by burning coal. In the sixties, natural gas took over gradually. In the mid seventies the price of oil and natural gas (in the Netherlands linked to each other) increased to such a degree, that re-introduction of coal became very profitable.
In agreement with the national planning, a 600 MWe coal fired power station was built in Nijmegen (1977-1982). The residues [Bottom Ash and Pulverised Fuel Ash PFA (UK) Fly Ash (USA)] were untill then disposed off or used as landfill. Limited space and environmental consequences led to the decision to search for other destinations of PFA. The sinterprocess of LYTAG Ltd (UK) was chosen to produce a light weight aggregate for the use as a filler in concrete. Twentyfive percent substitution of cement by PFA attributed better properties to concrete. Both applications have been used in the past 15 years in the Netherlands.

LYTAG-PROCES AND PRODUCT

PFA is a fine gray powder with a grain size distribution from 0.3 to 100 μm and with an average of about 20 μm. This powder is mixed and pelletised into grains of 4 to 12 mm on tilted dishes (see fig 1). The "green" pellets are transported to a sinterbed on which after ignition the sintering takes place. The unburned carbon in the ash is used as fuel, eventualy enriched with pulverised coal.

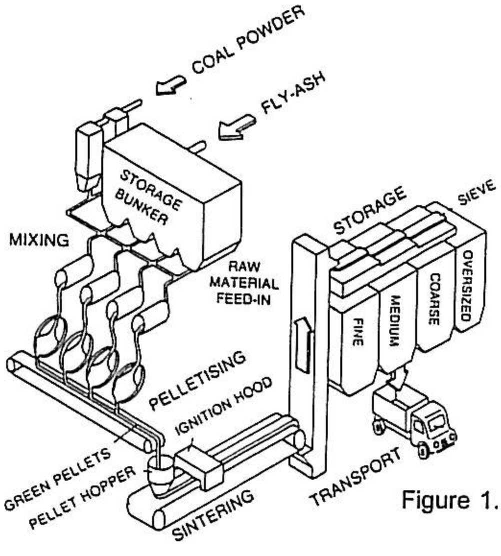

Figure 1. LYTAG-Proces

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After sintering the grains are sieved into fractions 0-4 mm, 4-8 mm and 6-12 mm. The 0-4 mm fraction is recycled. The oversize is crushed and used.
The use of PFA as cement substitute is limited to about 100 kg per m^3 concrete. The use of the Lytag product as a filler in concrete can go up to 800 kg per m^3.
From a radiological point of view it is necessary to investigate the consequences of the enhanced concentrations of uranium (U) and thorium (Th) in PFA. The average concentrations in coal approximately correspond to the average value in the earth crust. By removing the carbon by burning, the concentrations are an order of magnitude higher than in coal (the ash content of coal is about 10 %). All U and Th is concentrated in the ash. The gamma radiation dose from Lytag concrete will be higher than from gravel-concrete.
The radon emanation (responsable for alpha radiation) from PFA was unknown untill 1980. We first consulted the Royal Shell Labaratory at Amsterdam and in 1981 ordered the KEMA (Research institute of the electricity utilities) to measure the radon emanation (the fraction emitted in % of the total amount radon produced in the material) of PFA and Lytag grains.
The results were clear: PFA 0.44% to 1,6%, Lytag 6 mm 0,24%, Lytag
12 mm 0,09%.
Normal gravel-concrete has a radon emanation of 20%. This means, the α-radiaton dose from radon from Lytag concrete will be lower than from gravel concrete despite the higher concentration U+Th.

RADIOLOGICAL CONSEQUENCES

Thesis: The lower radon emanation could probably lower the α-radiation dose in dwellinghouses built from Lytag-concrete enough to compensate the higher gamma radiation dose.

To prove this thesis, it was necessary to quantify the indoor radiation dose in the average Dutch dwellinghouse.
The first rough radiological estimate was presented on the AshTech '84 conference in London [1].
The Dutch Health Council had requested The Radiological Institute TNO (Arnhem) to set up a computerprogramm to calculate the contribution of building materials to the indoor absorbed dose rates. J.G. Ackers designed the programm.
Values used for radioactivity content, radon emanation power and density of building materials were measured values. Insufficient knowledge about length of diffusion for radon in different materials led to the use of only one value in the calculations.
Other relevant factors are:

- average of a population, adults in relation to susceptible groups such as children,
- ventilation rates (0.5 h^{-1} to 0.7 h^{-1})
- room dimensions, wall thickness,
- resident times for the various groups,
- influence of neighbouring rooms
- wall covering,

During the study the initial conservative setting of values for these factors were, based on growing knowledge, replaced by more complex calculation systems and as a consequence more accurate (lower) values.

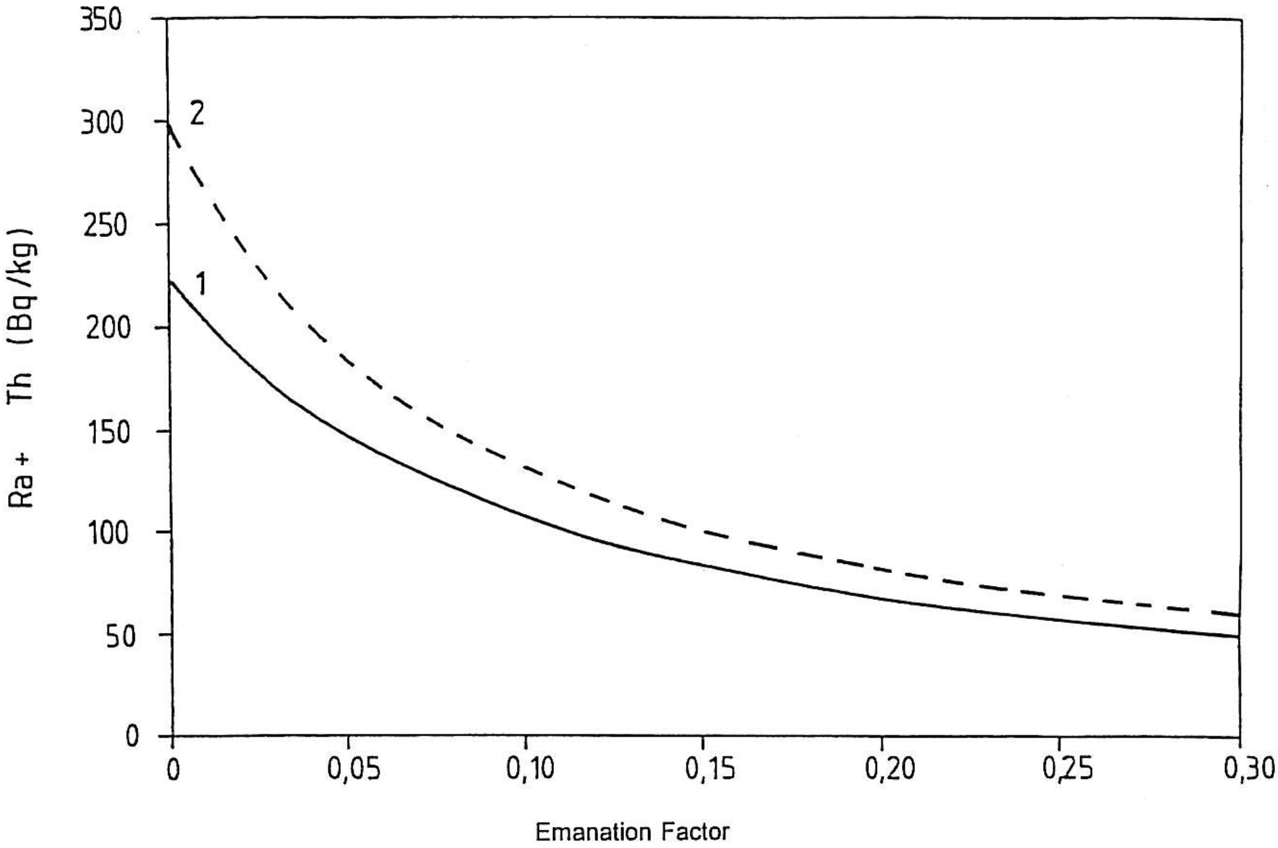
The outcome of this work "Methodology to determine additional radiation dose as a consequence of the application in dwellinghouses of building material with enhanced contents of natural radioactivity" is reported in [2].

Using these calculations the dose rate due to the application of Lytag in concrete could be compared with the dose rates from other concrete under equal conditions.

STANDARDS

Based on this work, the Ministry of Housing, Spatial Planning and Environment issued a publication "Radiation from Building material. Limit values for Concrete" in december 1988.
To evaluate limits on radioactivity contents for concrete in the Netherlands, a dwellinghouse in a block of flats was chosen as a reference. The reference material was concrete having a radioactivity content for the sum of Ra-226 + Th-232 of 75 Bq/kg.

The result is the graph as shown in figure 2, for concrete made with gravel (2400 kg/m^3) or Lytag (1900 kg/m^3) [3].



________ 1 Density 2400 kg/m^3 ----------- 2 Density 1900 kg/m^3

Figure 2. Limits for concrete and concrete with light weight aggregates

From this result a practical limit on the Lytag product and PFA is determined by the sum of Ra-226 and Th-232 activity of 400 Bq/kg (Ra-226 is in the decay chain of U-238 and therefore the same activity per unit mass in Bq/kg as U-238).

ACTIVITY MEASUREMENTS OF PFA

The Lytag factory was started up in 1983. The activity measurements have been performed from 1988 till today. In 1984 the sum of the activity of Ra and Th measured in ashes from coal from seven different mines was between 196 and 544 Bq/kg with an average of 352 Bq/kg. These were single coal types, resulting in rather wide spread values. Today, only blends are used.

The number of types of coal in a blend is high, sometimes up to 10 types. The consequence is a much more stable value of radioactivity. For example, in 1996 the number of measurements of Ra+Th in PFA at the Vasim factory was 295. The values had an average of 193 Bq/kg, a minimum of 132 Bq/kg and a maximum of 249 Bq/kg. This illustrates the influence of mixing the input-product as a possibility to stabilise the radioactivity of the product.

Since 1990, the value of 300 Bq/kg is not reached in practice. At the request of the factories which produce unsupported system flooring made of pre-stressed Lytag-concrete, the limit for floorings is lowered to 300 Bq/kg.

The limits for U in the EU for licensing is 1000 Bq/kg as well as for Th.
This limit of 2000 Bq/kg for the sum of U and Th is rather far from the present limit of 400 Bq/kg.

The routine measurement with Vasim for the activity of Ra+Th system, will be presented by L.P.M. van Velzen (presentation 5.2).

RADIOLOGICAL CONSEQUENCES FOR FLY ASH WORKERS

The first evaluation of this problem was carried out by J.A.M.M. Kops (Kema) in 1981 [4].
The results were based on the assumptions:

- working time 2000 hours a year,
- inhalation concentration dust (PFA) 10 mg/m^3,
- U concentration 244 Bq/kg, Th concentration 122 Bq/kg.

The sum of the dose of the isotopes in fly ash inhaled, of two chains of U and Th was 0.63 mSv/a

A more recent calculation appeared in reference [5], based on the practical assumptions for inhalation as follows:

- working time 1700 hours a year,
- inhalation concentration in practice at coal fired power stations,
- U concentration 140 Bq/kg and Th concentration 120 Bq/kg.

The sum of the doses of all isotopes of the two chains of U and Th and of Potassium (K-40) was 0.12 mSv/a.

Here we see again the effect of the first pessimistic calculations with a result of higher values and the more detailed, realistic assumptions and more complex calculations with lower values.

RADIOACTIVITY OF FLUE-GAS-DESULPHURISATION GYPSUM (FGD-GYPSUM)

To reduce acid rain, all coal fired power stations in the Netherlands have a desulphurisation system with the production of gypsum as a consequence. The radioactivity of the gypsum was measured as soon as it was produced.
The results were:

Radium 226	7 Bq/kg
Thorium 232	1 Bq/kg
Potassium 40	8 Bq/kg

Natural gypsum had values of activity of Ra-226 of 4 to 30 Bq/kg.
The use of FGD-gypsum as a building material is acceptable on a radiological point of view.

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