RADIOACTIVITY INDUCED IN BUILDING MATERIALS

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GENEVA
1967
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FOREWORD

It is interesting to study the modern radioactivity of building materials, for several reasons.

Firstly, they cannot be removed when they get hot, as may be possible in the case of equipment.

Secondly, the choice is limited, so that one has to be careful, particularly in places where the irradiating particle fluxes are high.

Thirdly, these materials are also widely used in the form of removable blocks, for shielding purposes in very hot areas. These blocks may have to be manipulated rather often.

Fourthly, there is also the problem of low-background laboratories, for which the natural radioactivity of the material is decisive.

The present work is an attempt to give some orders of magnitude of physical facts relevant to the question.

A first part is devoted to the description of the various particle fluxes which can activate, and to computations of the effects which are to be expected on the basis of known cross-sections.

A second part describes experiments which were carried out at both the CERN Proton Synchrotron and the Synchro-cyclotron in the particular conditions of fluxes of activating particles met in the vicinity of the machines.

A third part refers briefly to measurements of the natural radioactivity of some building materials.

General conclusions are drawn at the end.
PART I
DESCRIPTION OF THE ACTIVATING FLUXES
AND CALCULATION OF THE RESULTING ACTIVITY

1. ACTIVATION OF STRUCTURAL MATERIALS BY
THERMAL, EPITHERMAL, AND FAST NEUTRONS

Besides fission products, activated structural materials are a source of hazard in facilities where a large number of fission or evaporation neutrons are released. This is the case not only with reactors, but also with accelerators, since here we find that as well as the very high energy cascade particles generated in the spallation process there is an almost comparable number of so-called evaporation neutrons, produced by the unstable nucleus remaining when the cascade particles have left.

We shall first consider the activation produced by thermal, epithermal, and fission neutron spectra usually found in reactors, as there is some information available on this matter, and we shall then examine the case of evaporation neutrons from compound or excited nuclei, about which much less is known.

The problem here is to take the excitation function of the target element for the particular reaction (cross-section as a function of neutron energy) to multiply it by the expected neutron energy spectrum normalized to unit flux and to integrate the product over the relevant energy interval in order to obtain a so-called average cross-section. This can then be multiplied by the total flux in the energy intervals considered in order to find the induced activity with the usual formulae. Such calculations are long, and have been made by various authors for specific neutron spectra which they encountered in their work. Here we shall use the data of Culp and Page, as reviewed and presented by Layman and Thornton.

The spectrum of reactor neutrons used by these authors represents a close fit to the neutron energy spectrum which had been measured at a given position in a water shield surrounding a particular water-moderated reactor (the so-called Ground Test Reactor) by Romanko and Dungan. This spectrum was divided into three energy ranges:

i) the thermal region, with $0 < E_n < 0.4 \times 10^{-6}$ MeV,
ii) the epithermal region, with $0.4 \times 10^{-6} \text{ MeV} < E_n < 0.1 \text{ MeV},$
iii) the fast region, with $0.1 \text{ MeV} < E_n < 9 \text{ MeV}.$

In each of these energy ranges the form of the energy spectrum was different. In the first range (thermal region) the measurements showed that the energy distribution was practically Maxwellian. This means that the neutrons are in thermal equilibrium with the surrounding atoms. As a consequence of this, when a neutron with a given energy leaves a given volume-element, it is immediately replaced in steady-state conditions by another of the same energy which flies in from the outside of this volume-element. The Maxwellian or thermal energy distribution gives for the density or number of neutrons with an energy between $E$ and $dE$ present in some given volume an expression of the form $E^{1/2} \exp (-\text{const} \times E) \, dE.$ That which activates is, in fact, the flux of particles through the volume, which is obtained by multiplying the density of the particles by their velocity, which is proportional to $E^{1/2}.$
One thus arrives at the expression for the flux which was used for the calculations, and reads as follows:

$$\phi(E) = 1.46 \times 10^{15} \frac{\text{E exp} (-3.82 \times 10^7 \text{E})}{\text{cm}^2 \text{ sec MeV}}$$

for the energy interval (i) where \(\phi(E)\) has been normalized so that the integral of \(\phi(E)\) between 0 and \(0.4 \times 10^{-6}\) MeV will give 1. The energy is always given in MeV.

In the second region (that of the so-called epithermal neutrons), the conditions are different. The fission neutrons are being slowed down, mainly by repeated collisions with the atoms of the moderator. One finds experimentally that the neutron flux has roughly a \(1/E\) dependence upon energy. Such a dependence is often encountered when fast neutrons are being slowed down and when there is enough hydrogen around, as this element is known to change the neutron energy in collisions by the largest amounts possible, due to the equality of its mass with that of the neutron. The flux used in this region is:

$$\phi(E) = \frac{1}{12.4} \frac{\text{neutrons}}{\text{cm}^2 \text{ sec MeV}}$$

for the energy interval (ii). This expression integrated between \(0.4 \times 10^{-6}\) and \(10^{-1}\) MeV again gives 1.

In the third region (that of the fast neutrons) the flux measurements showed a rather smooth exponential decrease with energy. The exponential decay resembles the tail of the fission neutron distribution as measured at a distance from a small quantity of fissionable material, the fission of which occurs either spontaneously or by bombardment with a jet of neutrons from an external source (Bowman et al., Leachman). Note that here there is no equilibrium with the surroundings, so that the neutron energy distribution function also gives the flux at a distance, taking into account the appropriate geometrical factors.

The flux energy distribution of the neutrons used for the calculation of the average cross-section in this range (iii) cannot be imitated by a simple formula and is best taken from the graphical representation as presented in Fig. 1 (full line). For comparison the neutron spectrum arising from the thermal fission of \(^{235}\text{U}\) according to Grundl (dashed line) is also shown on this figure, as well as the evaporation neutron spectrum \(N(E)_{\text{evap}}\) from silver bombarded by 190 MeV protons after Gross (line with dots and dashes).

Once these spectral flux distributions were defined, the averaged cross-sections of the nuclear reactions expected in the energy interval were calculated from the formula

$$\sigma = \frac{1}{E_{n_2} - E_{n_1}} \int_{E_{n_1}}^{E_{n_2}} \phi(E)\sigma(E) \, dE,$$

\(E_{n_1}, E_{n_2}\) designating the limits of the neutron energy interval considered.

The nuclear reactions taken into account were the \((n,\gamma)\) reaction in the thermal and epithermal ranges, and the \((n,p)\) and \((n,\alpha)\) reactions in the fast neutron range. This was done for each of the naturally occurring isotopes of the target element considered, and the
corresponding averaged cross-sections were added in proportion to the natural abundance of the particular isotopes. The results for a number of selected structural materials can be found in a table presented in Appendix B of Layman and Thornton's book, and were used by the present author to compute the gamma dose rate decay curves of these materials for an infinitely long irradiation time. These curves, which represent the gamma radiation field in rad/h at 1 metre from a quantity of 1 gram of the particular element irradiated by a flux of 1 neutron/sec cm² in the energy range indicated for an infinite time, are given in Figs. 2, 3, 4. For a finite irradiation time the reader will, as usual, subtract the activity values at the time \( t = t_0 \), \( t = t_1 \) from that at the time \( t = t_0 \), according to the well-known theorem. In cases where this procedure leads to inaccurate readings, especially for short activation times, the reader is referred to Layman and Thornton's book, which gives graphs showing the build-up of radioactivity with irradiation time from 10 to 1000 hours, at various cooling times (1, 10, 100, 1000 hours). The units used by these authors is roentgen/h lb at a distance of 1 foot. The equivalent in our units is

\[ 1 \text{ roentgen/h lb at 1 foot} = 2 \times 10^{-4} \text{ rad/h g at 1 m}, \]

the factor for converting from roentgen to rad (tissue) being 0.965; from lb to g 1/4.54; from feet to metres distance \((0.305)^2\).

2. **ACTIVATION OF STRUCTURAL MATERIALS BY HIGH-ENERGY PARTICLES PRODUCING SPALLATIONS**

The phenomena and calculations involved here have been described in a previous paper ('CERN Report 65-34). For our present purpose it will be sufficient to show gamma dose decay curves of some materials activated by 600 MeV protons, to be compared with the dose-levels expected from the activation by neutrons of various energies. This is done with Fig. 5, from which one sees that the curves of spallation-induced activity for the various materials lie very near to each other in the range \(10^{-13}\) to \(10^{-14}\) rad/h g at 1 m, which is higher than most of the activities induced by the neutrons. So, in general, for equal fluxes, the high-energy particles inducing spallation give more radioactivity than evaporation neutrons.

For the evaluation of the radiation fields produced by large solid bodies of radioactive material, it is convenient to use the so-called danger parameter \( D \). \( D \) is the radiation field existing inside a cavity surrounded by an indefinitely extended body homogeneously activated by a unit flux. It depends, of course, on the material, and on both irradiation and cooling times. At a location from which a solid body is viewed under a solid angle \( \Omega \), the radiation field \( R \) is then

\[ R_{\text{rad/h}} = \frac{\Omega}{4\pi} D \phi \]

where \( \phi \) is the irradiating flux. Danger parameter values for specific shielding materials, such as C, O₂, Al, SiO₂, CaO₂Ca, SO₄ Ba are given in Figs. 6 to 11 in graphical form.
1. IRRADIATION IN A PURE 600 MeV PROTON BEAM
FROM THE CERN SYNCHRO-CYCLotron

Two irradiations were made in the extracted proton beam from the CERN 600 MeV Synchrocyclotron. The advantages of using this beam are several: one has a high flux (of the order of \(10^{10}\) protons/sec cm\(^2\)), a well-defined energy, and a pure beam. Neutron fluxes coming directly from the machine are at least three orders of magnitude lower. Also the beam diameter (defining the region where the local flux density is more than 50% of maximum) is above 1 inch, permitting a number of samples in the form of thin slices of reasonable surface (up to 4 cm\(^2\)) to be irradiated at the same time, with their axis either parallel or perpendicular to the beam, depending on the particular requirements.

1.1 Irradiation of various limestones with 600 MeV protons

The first experiment consisted in irradiating samples of various limestones including white and grey Italian marbles and various building stones of French origin with a view to selecting one for lining the walls, ceiling, and floor of a planned irradiation cave. Aluminium and quartz samples were also irradiated for comparison. The gamma counts with the sample in contact with a 3" x 3" NaI(Tl) crystal were registered and are presented in Fig. 12. The figure caption includes the names of the various stones, which for simplicity were marked A to I in the figure. It is seen that all the limestones, with the exception of grey marble, are a factor of at least 3 better than fused silica, in the 15 h cooling-time region (\(^{24}\)Na isotope). The A-rock (yellow Vaurion stone, from the Yonne river region in France) seems to be slightly better than the others, and was chosen for lining the cave. It will also appear as the typical limestone in the subsequent experiments.

Chemical analysis of some of the samples irradiated will be found in Tables 1 and 2, together with the analysis of various other materials used in the following experiments.

1.2 Irradiation of other samples in the 600 MeV proton beam

The second experiment, also in the extracted 600 MeV proton beam of the synchrocyclotron, included still more specifically used building and shielding materials. The fused silica was replaced by the actual sand and cement mixture used to make ordinary (or "light") concrete at CERN (without water). This is labelled simply "concrete". Vaurion yellow rock and white Carrara marble were selected to represent the natural limestones. A mortar with high calcium content and inorganic binding, labelled Chemtree 20, recently developed by Chemtree Corporation, Central Valley, New York, was also included as an example (in powder form without water). Finally, aluminium, used as before as a monitor in pure proton beams, and carbon were added to the list. The results of the various measurements taken on the samples are presented in Figs. 13 to 16. For concrete, rock, marble, and Chemtree 20, both thin (1.5 cm x 1.5 cm x 0.2 cm) and thick (1.5 cm x 1.5 cm x 15 cm) samples were prepared, and irradiated at the same time in the beam, the thin ones in front of the thick ones. The latter were placed with their longitudinal axes along the beam line.
### Table 1
Chemical analysis of some building materials

<table>
<thead>
<tr>
<th></th>
<th>CBM ordinary concrete</th>
<th>Cement-sand mixture</th>
<th>CBM barite concrete</th>
<th>White Carrara marble</th>
<th>Porphyrite marble</th>
<th>Jura stone</th>
<th>Yserion yellow</th>
</tr>
</thead>
<tbody>
<tr>
<td>All %</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Losses by fire (1000°C)</td>
<td>15.42</td>
<td>2.50</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO₂</td>
<td>4.29</td>
<td></td>
<td>43.40</td>
<td>43.05</td>
<td>43.71</td>
<td>43.57</td>
<td></td>
</tr>
<tr>
<td>H₂O</td>
<td>4.29</td>
<td></td>
<td>0.28</td>
<td>0.97</td>
<td>0.30</td>
<td>0.07</td>
<td></td>
</tr>
<tr>
<td>SiO₂</td>
<td>46.72</td>
<td>47.59</td>
<td>8.69</td>
<td>0.37</td>
<td>0.09</td>
<td>0.96</td>
<td>0.67</td>
</tr>
<tr>
<td>All sesquioxydes</td>
<td>8.79</td>
<td>3.40</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>1.92</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.51</td>
</tr>
<tr>
<td>MnO</td>
<td>0.06</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Al₂O₃</td>
<td>6.29</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td>0.06</td>
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<tr>
<td>CaO</td>
<td>25.38</td>
<td>30.53</td>
<td>7.59</td>
<td>54.95</td>
<td>54.85</td>
<td>50.54</td>
<td>55.18</td>
</tr>
<tr>
<td>BaO</td>
<td>49.93</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MgO</td>
<td>1.43</td>
<td>1.30</td>
<td>0.09</td>
<td>0.51</td>
<td>0.34</td>
<td>3.80</td>
<td>0.23</td>
</tr>
<tr>
<td>K₂O</td>
<td>1.00</td>
<td>1.16</td>
<td>0.75</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Na₂O</td>
<td>1.07</td>
<td>1.20</td>
<td>0.35</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>SO₃</td>
<td>0.25</td>
<td>0.71</td>
<td>26.60</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe,Al</td>
<td>0.4</td>
<td>0.32</td>
<td>0.56</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total %</td>
<td>100.06</td>
<td>99.97</td>
<td>99.90</td>
<td>99.91</td>
<td>99.60</td>
<td>99.87</td>
<td>100.09</td>
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### Table 2
Chemical composition of Chemtree 20

<table>
<thead>
<tr>
<th>Element</th>
<th>Percentage</th>
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</thead>
<tbody>
<tr>
<td>Al</td>
<td>1.6</td>
</tr>
<tr>
<td>Ca</td>
<td>36</td>
</tr>
<tr>
<td>C</td>
<td>7.5</td>
</tr>
<tr>
<td>H</td>
<td>1</td>
</tr>
<tr>
<td>O</td>
<td>50.5</td>
</tr>
<tr>
<td>Si</td>
<td>2.4</td>
</tr>
<tr>
<td>Na</td>
<td>0.2</td>
</tr>
<tr>
<td>Others</td>
<td>0.8</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>100</strong></td>
</tr>
</tbody>
</table>
Figure 13 shows, as before, the gamma counts registered with the thin samples in contact with the 3" x 3" NaI(Tl) crystal.

Marble and rock appear to be a factor of 3 more favourable than concrete and a factor of 10 more than aluminium. Cheethree 20 is twice as favourable as concrete. These comparison values hold for the number of counts registered; they will be slightly altered when measuring dose-rates (see next figures).

Figure 14 shows the measured dose-rates. For this measurement, the large samples were used and placed at a distance of about 50 cm from the radiation monitor used, with their longitudinal axis perpendicular to the line going from the sample to the detector, so that absorption in the sample itself was small and could be ignored. The curves are in millirad/hour per gram at 50 cm. The same situation is observed as with the gamma-count numbers.

Figure 15 is an attempt to obtain the general "danger parameter" from another dose-rate measurement with the thick samples. This time, the radiation field was measured at a distance from these samples, but along their longitudinal instead of transverse axes. The gamma rays had thus to traverse the sample itself before reaching its surface and passing the detector. On the assumption that the sample has been uniformly irradiated and that the sample thickness (15 cm, or about 40 g/cm²) can be considered as infinite for the gamma rays, one arrives easily at the danger parameter D by solid-angle considerations. It had been defined that D was the radiation field existing inside a cavity embedded in an infinite volume of uniformly active material, i.e. at a point viewing the active body under a solid angle 4π. Here the thick active material slab was viewed from the detector under the solid angle 8π/x₀², where S is the slab section and x₀ the distance to the detector. Thus the measured millirad/h values were multiplied by 4πx₀²/8 in order to arrive at the D values presented in Fig. 15. By this criterion, rock marble and Cheethree 20 are roughly equivalent, and about a factor of 3 more favourable than the CERN ordinary concrete which shows radiation dose values higher by this factor than those from the other materials.

Figure 16 shows the decay of the beta activity, which was measured for only one sample (Cheethree 20). The counter arrangement used is that described in CERN report 65-54. The counts have been corrected both for distance to the counter end-window and for thickness of the sample, according to the procedures described in the above-mentioned report. What is presented in Fig. 16 is the net number of beta particles of either sign leaving one square centimetre of the surface of an infinitely thick block of material after activation as described. This is a significant feature of the material and the activating flux. It is highly probable that this figure is roughly the same for the rock and marble samples irradiated at the same time.

2. IRRADIATION IN A MIXED NEUTRAL FLUX IN THE VICINITY OF THE CERN SYNCHRO-CYCLotron

As was pointed out in the first part of this report, the high-energy charged particles are not the only activating agent around accelerators. In the vicinity of these machines one finds also large amounts of neutral particles, with energies ranging from thermal to the maximum machine energy. With this in mind, samples were placed in the forward direction from the usual synchro-cyclotron internal target, at about 6 m from the target location.
The charged particles can be assumed to be swept away by the stray magnetic field of the machine to a large extent, so that one will have to deal almost only with a neutral flux. From various measurements the neutral fluxes are estimated to be the following:

i) above 20 MeV, inducing $^{11}$C in carbon: $3.4 \times 10^2$ sec cm$^{-2}$;

ii) thermal neutrons: $7 \times 10^2$ sec cm$^{-2}$;

iii) neutrons with energies up to 10 MeV: $2.5 \times 10^7$ sec cm$^{-2}$.

The irradiation time was 5 days.

The purpose of the experiment was also to check whether the $^{23}$Na content of the samples exposed is prohibitive, as $^{24}$Na will then be formed by thermal neutrons with a high cross-section.

Small samples of Na, Al, CERN cement-sand mixture, Chemtree 20 powder, Vaurion rock, white Carrara marble and carbon were exposed. In these, even in the absence of $^{23}$Na, the isotope $^{22}$Na can also be formed via spallation by high-energy neutrons, but with a much smaller cross-section. The results are presented in Fig. 17, where the registered gamma counts are shown. The induced radioactivity in pure sodium is more than two orders of magnitude above that in marble, rock, and Chemtree 20, showing that the Na content of these products, if any, is not relevant. Concrete appears to be a factor of 3 to 4 above the products mentioned under the conditions of the experiment.

Gamma-ray spectra were taken from the irradiated products both during this experiment and after the irradiation in the pure 600 MeV proton beam. As an example, the spectra of Chemtree powder, at about 11 hours cooling time and for both irradiations, are compared in Fig. 18. The $^{24}$Na peak from the neutral irradiation experiment is not markedly bigger than the one from the high-energy proton irradiation when compared to those from other peaks (such as that of the $^{24}$Na isotope, for instance), showing that the thermal neutron production of $^{24}$Na is not dangerous in this material.

3. IRRADIATIONS IN A MIXED CHARGED AND NEUTRAL FLUX NEAR A TARGET OF THE CERN PROTON SYNCHROTRON OPERATED AT 19 GEV

As another example of activation by high-energy particles, we will now examine the decay curves of several similar groups of carbon, marble (white Carrara) and silica samples exposed during 5 months (from 28 May to 20 October, 1964) in the neighbourhood of four different targets located at units 1, 6, 60, and 64 of the CERN Proton Synchrotron. The internal beam current intensity of the synchrotron was about $3 \times 10^{11}$ protons per second during this period, and the machine was in operation about 70% of the time. The distance from the samples to the targets was between 1.2 and 2.7 metres, and the angle to the forward direction 1° to 2°. The decay curves for the same material at the various locations are presented on the same graph, in order to illustrate the differences in specific activity and slope of decay which can be found in practice, and are caused by the irregular operation of each target with respect to irradiation time and fraction of beam allowed to hit the particular target. Figure 19 shows the decay of the carbon samples, and Fig. 20 that of marble and silica. For comparison, Figs. 21 and 22 show the corresponding decay curves of aluminium, lead, iron, and copper. One can conclude from inspection of these figures that differences in the specific activity of the same material can
easily cover a factor of 10 due only to different targeting conditions in the same machine, although the samples are in all cases at about the same angular position and distance from the target. The differences are especially apparent during the first days of the decay, reflecting the different targeting conditions. In contrast, the activities of radio-nuclides with longer half-lives, which represent an average of the machine conditions, show less dispersion. Under the circumstances it is seen that marble is on the whole superior to silica by a factor of 3 to 4 and comparable to carbon for specific activity. Figure 23 shows a typical marble gamma-energy spectrum taken 56 hours after this irradiation, with the main activities noticeable: $^{24}$Na, $^{26}$Na, $^{47}$K, $^{48}$Sc, and $^{47}$Ca (weak).
The natural radioactivity of building materials has always been a matter of interest, both for estimating the level of radiation in houses and for selecting suitable materials for low-background laboratories.

A remarkable study on the subject was done by Penko, Byziek and Duczynski (1963). The results obtained were as follows. The highest gamma activity is to be ascribed to boiler slag, blast-furnace slag, and fly ashes (23-35 gamma photons/min · g). Slightly less radioactive are carbonaceous shale, bricks and brick clays, blast-furnace cements (22 gamma photons/min · g). Still less gamma-radioactive are gravel, ordinary cement, concrete, sand, and mortar (5-10 gamma photons/min · g). The lowest radioactivity is displayed by gypsum, lime, and chalk (average below 5 gamma photons/min · g). The radionuclides found are $^{40}$K, $^{226}$Ra (and decay products $^{214}$Pb, $^{214}$Bi), $^{232}$Th (and decay products $^{212}$Bi, $^{228}$Ac, $^{208}$Tl) and a series of gamma-energy spectra of various materials are presented. The work includes quantitative estimates on yearly radiation doses received by inhabitants of houses made with the various building materials.

In CERN, measurements of the natural radioactivity of four of the materials irradiated in the 600 MeV proton beam (see Part II, 1.2) were performed by the Health Physics Group. The materials selected were the sand and cement mixture composing the CERN concrete, the white Carrara marble, the Vaurnon yellow stone, and Chemtree 20. The results are presented in Table 3. The first two columns indicate the specific activity of the $^{40}$K alone, expressed in both picocuries/g, and in photons/g · min as measured from the peak at 1.46 MeV in the gamma-energy spectrum. In the picocuries/g calculation, account was taken of the fact that $^{40}$K emits a photon only in 11% of the disintegrations. The third column gives the total gamma photons ($^{40}$K and all others) emitted per gram and minute for each material. This information has been obtained as follows. The gamma counts in each channel of the energy spectrum were multiplied by the increase in efficiency of the counting equipment, and integrated up to and including the $^{40}$K peak. Above the $^{40}$K peak, no significant activity was observed, and the remaining few counts were neglected.

<table>
<thead>
<tr>
<th>Material</th>
<th>$^{40}$K activity only</th>
<th>Total activity (incl. $^{40}$K) up to 1.5 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>picocuries g</td>
<td>photons min · g</td>
</tr>
<tr>
<td>CERN concrete</td>
<td>6.6</td>
<td>1.6</td>
</tr>
<tr>
<td>Limestone (Vaurnon yellow)</td>
<td>1.53</td>
<td>0.37</td>
</tr>
<tr>
<td>Marble (Carrara white)</td>
<td>0.99</td>
<td>0.24</td>
</tr>
<tr>
<td>Chemtree 20</td>
<td>4.79</td>
<td>1.17</td>
</tr>
</tbody>
</table>
The counts other than $^{40}$K are mainly due, according to the form of the energy spectrum, to $^{226}$Ra in these materials. The values observed of total photons emitted per gram and minute are roughly the same as the average values given by Pasko, Ryszko and Dusznyski for marble and limestone, and 1.5 times higher for concrete and Chemtree 20. The agreement is thus very good. Thus it would appear that marble is the best natural stone for building low-background laboratories.

* * *

CONCLUSIONS

In high-energy accelerator installations, two kinds of activating fluxes are found: the very high energy particles producing spallation, and the evaporation neutrons emitted by excited spallation products. These evaporation neutrons, subsequently moderated and thermalized, give rise to a neutron atmosphere which has an energy spectrum roughly similar to a fission spectrum or to a reactor spectrum measured behind some shielding.

Computations of specific radiation fields in rad/h per gram at 1 metre were made by various workers and by the author for both the spallation part (very high energy particles) and the evaporation part (thermal epithermal, fast neutrons). The results vary widely from target to target for the evaporation neutrons, and lie between $10^{-11}$ and $10^{-20}$ rad/h per gram at 1 metre. In contrast they are grouped together for nearly all materials around $10^{-14}$ rad/h per gram at 1 metre in the spallation case, except for materials with mass numbers below 20 and for calcium, which are 10 times less active after one day.

For building materials proper, it is thus interesting to compare experimental data on concrete and limestones, and to compare these with aluminium and iron, often used as structural or as shielding materials. For the experiments we have taken aluminium instead of iron, as the radiation levels in at least the first 15 hours are comparable both in the spallation and in the fast neutron region. The results are roughly as follows. Calcite $CaCO_3$ (either as marble or relatively pure rock) is three times less active than concrete or silica, and about ten times less than aluminium or iron. Carbon is usually still better than calcite.

For low-level laboratories, the natural radioactivity of pure white marble is about the best that can be found in natural rocks.

* * *

Acknowledgments

It is a pleasure to thank Mr. F. Hoffmann for having helped in performing the experiments, done all the counting, and drawn the graphs. We acknowledge the help of the CERN Health Physics Group, and especially that of Mr. Renaud and Miss Dubosson, in doing for us the measurement of the natural radioactivity of four of the samples.


J.A. Grundl, Study of fission neutron spectra with high-energy activation detectors, Report LANL 2883, Los Alamos Scientific Laboratory, New Mexico, 20 May, 1963.


W.C. Middelkoop and B. de Raad, Induced $^{24}$Na activity in the concrete of the CERN FS Tunnel, CERN internal report AR/Int. 65/13, 11 June, 1965.


Fig. 1: Neutron spectra in ground test reactor (full line) from thermal fission of $^{235}$U (dashed), and from evaporation (dots and dashes).
Fig. 2: Specific gamma dose-rates for selected target elements, unit flux, and infinite irradiation, with activation by thermal neutrons.
Fig. 3: Specific gamma dose-rates for selected target elements, unit flux, and infinite irradiation, with activation by epithermal neutrons.
Fig. 4: Specific gamma dose-rates for selected target elements, unit flux, and infinite irradiation, with activation by fast neutrons.
Fig. 5: Calculated dose decay curves of common construction materials for an irradiation by 600 MeV protons.
Fig. 6: Danger parameter for C.
$O_2$

Irrad. energy $\geq 500\ MeV$
Flux $10^6$ part./sec. cm$^2$

$t_i = 360\ days$
$t_i = 30$
$t_i = 7$
$t_i = 1$

Fig. 7: Danger parameter for $O_2$. 
Fig. 9: Danger parameter for SiO$_2$. 

SiO$_2$
Irrad. energy $\geq 500$ MeV
Flux $10^6$ part./sec.cm$^2$

$t_i=5000$ d

1, 30, 360
Fig. 10: Danger parameter for Co$_3$Ca.
Fig. 11: Danger parameter for BaSO₄.
Fig. 12: Gamma activity of various limestones irradiated in a 600 MeV proton beam:
A - Vaurion roche jaune  F - Grey marble Santo Patrizio
B - Vaurion pointillé     G - White Carrara marble I
C - Bernais Liais rubané  I - White Carrara marble II
D - Tournus veiné         SiO$_2$ - Fused silica, for comparison
E - Malombré Lonchant unie Al - Aluminium, for comparison
(counts/g * min registered with sample in contact with 3" x 3" NaI crystal).
Fig. 13: Gamma activity of carbon, aluminium, concrete, Vaurion rock, Carrara marble, and Chemtree 20, after irradiation by 600 MeV protons (registered counts/g • min with sample in contact with 3" × 3" NaI crystal).
Fig. 14: Gamma radiation field from concrete, Vaurion rock, Carrara marble, and Chemtree 20, after irradiation by 600 MeV protons.
Fig. 15: Gamma danger parameter from concrete, Vaurion rock, Carrara marble, and Chemtree 20, after irradiation by 600 MeV protons.
Fig. 16: Beta activity of Chemtree 20, after irradiation by 600 MeV protons, expressed in betas/cm$^2$ min leaving the surface of an infinitely thick sample.
Fig. 17: Gamma activity of C, Na, Al, concrete, Vaurion, marble, and Chemtree 20, after irradiation by a neutral flux in the vicinity of the 600 MeV synchro-cyclotron target (registered counts/g \cdot min with sample in contact with 3\" \times 3\" NaI crystal).
Fig. 18: Comparison of gamma-ray spectra of Chemtree 20 from 600 MeV protons and from neutral flux irradiations.
Fig. 19: Activity decay of carbon samples irradiated at various locations at the CERN Proton Synchrotron.
Fig. 20: Activity decay of silica and marble samples irradiated at various locations at the CERN Proton Synchrotron.
Fig. 21: Activity decay of aluminium and lead samples irradiated at various locations at the CERN Proton Synchrotron.
Fig. 22: Activity decay of iron and copper samples irradiated at various locations at the CERN Proton Synchrotron.
Gamma energy spectrum of marble
$t_i = 5$ months  $t_c = 54$ hours (PS)

--- efficiency of NaI(Tl) 3''x3''

Fig. 23: Gamma energy spectrum of marble irradiated at the CERN Proton Synchrotron.