Playing hard to catch

Seeing more with less (radiation)

BY JOOST VAN KASTEREN

X-ray and gamma radiation are widely used in hospitals and laboratories, and in the manufacturing and mining industries. Several methods exist for making the radiation «visible», including scintillators, films, and semiconductor materials. Scintillators are crystals that convert ionising radiation into visible light. This is a field in which there is some room for improvement. Some crystals are very difficult to produce, or they have a very long response time. In his search for better crystals, graduate student Hans van ’t Spijker found himself looking at bromides and chlorides. These proved to be pretty efficient, opening the way to seeing more with less radiation.

Radioactive radiation is widely used in many applications. X-ray radiation for example is used for medical diagnostics and therapy as well as for nondestructive research of materials, for example to see if a welding joint will pass muster. Gamma radiation is used in medical diagnostics, but also to explore well bores. To use radiation for diagnostic purposes you need a radiation source and some means of detecting the radiation it produces. Dr. Ir. Hans van ’t Spijker was awarded his doctorate for his research into new materials that can be used for detecting gamma radiation.

Dentists use photographic plates to make an x-ray

The L3 detector at CERN, the European institute at Geneva where particle accelerators are used to find out more about the properties of elementary particles, measures 16 x 16 metres, and weighs 8500 tonnes. Inside the LEP accelerator (with a length of 27 km), collision experiments are conducted with electrons and positrons to discover new particles (quarks). The red magnet contains various types of radiation detectors, including approximately 10,000 crystals (scintillators) that emit light when struck by elementary particles.
picture. Simply hold an envelope against a tooth, expose it to x-rays for a second, and within a few minutes you can see any drilling and filling that needs to be done. However, for many applications, the photographic plate is not very handy, according to Dr. Pieter Dorenbos, lecturer at the Department of Radiation Research Instrumentation of the Interfaculty Reactor Institute. He was Van ’t Spijker’s tutor and for many years has been conducting research into new detectors, not only for gamma and x-ray radiation, but also for neutron radiation. The problem is that photography involves the use of chemicals, whereas these days people universally prefer their data in digital form so they can process it using a computer.

**Metabolic processes**

Large-scale data processing occurs in such diagnostic techniques as computer tomography (CT) and positron emission tomography (PET). Computer tomography involves exposing part of the body to x-rays from different angles to obtain a sectional view of the body part. PET is used in the medical profession to visualise metabolic processes taking place within the human body. Contrary to x-ray diagnostics, PET does not use radiation from the outside, but rather radiation produced inside the body, using radioactive materials with a short decay time. The principle is as follows: the patient is given a drug that plays a role in a certain metabolic process, e.g. inside the brain. One of the atoms in the compound has been replaced by a radioactive isotope, which upon decaying produces a positron, a positively charged particle with a charge equal, though opposite, to that of an electron. Upon its release, the positron practically immediately recombines with an electron from the surrounding tissue. This releases two quanta (photons) of gamma radiation, which can be detected outside the body. The location within the organ from which these quanta originated can also be determined. If sufficient numbers of radioactive nuclei decay, a time-lapse image of the progress of the metabolic process within a certain organ can be obtained.

**Sparkler**

The accurate detection of the gamma radiation produced in PET systems is essential for the diagnostic side of the process. In the first place, a sharp image is required, i.e. with a plate resolution of two millimetres or less. The measuring time is also important. To obtain an image of the human brain, some 500 million positron-electron annihilations have to be observed. To be able to do so within a reasonable stretch of time, detectors are needed that can efficiently detect the photons. Detection takes place by means of scintillation. The gamma quanta are captured (absorbed) in a crystal in which they are converted into light, which is subsequently detected by, e.g., a light-sensitive diode. The diode then produces an electric signal that can be processed. The light pulse generated by the gamma radiation is referred to as a scintillation, after the Latin for spark, scintilla. Consequently, the material that converts the energy of the gamma photon into light is called a scintillator, i.e. a sparkler.

To find out whether a well contains oil and/or gas, different types of probes containing various measuring instruments are lowered into the well including a gamma radiation source and scintillation detector.

(image anadrill schlumberger)
Salt
Scintillators were being used shortly after the discovery x-rays in 1895. Wilhelm C. Röntgen (1845-1923) himself used a scintillator to detect the radiation that was to bear his name. When he irradiated a sheet of glass coated with barium platinocyanide, the irradiated sections of the glass became fluorescent. A few months after the discovery by Röntgen, Thomas Alva Edison discovered that calcium tungstenate worked even better. Since then, many more scintillators have been discovered and tested, including various oxides, sulphides, and halides of cadmium, cesium, bismuth, and gadolinium (one of the rare earth elements). Halides are compounds of fluorine, bromine, chlorine, or iodine. One of the most common scintillator materials is sodium iodide (a halide) with thallium, which has the same crystal structure as ordinary table salt. Generally speaking, scintillators are clear crystals. Dorenbos shows a piece of bismuth germanate. It is clear like glass, but a lot heavier. It is made by melting a powder inside a furnace, and then pulling the crystal very slowly from the molten mass. Manufacturing a large crystal (of say, 50 cm) can take many weeks. During the cooling process, more or less regular crystal lattices are formed. More or less, since a lattice like this will always contain a few defects. Moreover, extra impurities may be added to the material to ensure that the scintillator emits light.

Photo multiplier
‘At this moment, there is a real need for more efficient scintillators’, says Van ’t Spijker. ‘If you can improve the detection efficiency of the scintillator, you need less gamma radiation to obtain the same result. And, scan times can also be reduced. Both would be beneficial to patients.’

According to Dorenbos another reason is that we are currently seeing a shift in the method used to measure scintillation light. In the past, and today still, the process involves the use of photo multiplier tubes, rather like a tv set that works the other way around. In these tubes, light strikes a photo-sensitive cathode, which releases an electron that flies across the tube to land on the anode, where it causes a minute current to flow. Photo multiplier tubes take up quite a bit of space, and for medical applications in particular, this is a serious drawback. Photodiodes are more compact. By replacing the photo multipliers with diodes, more scintillation crystals can be fitted so the resolution can be improved. ‘In addition,’ Dorenbos adds, ‘the tubes are relatively expensive. All in all, photodiodes are the preferred solution, but the problem is that they work best with red light, whereas most scintillators emit light with wavelengths in the blue and ultraviolet range. This is another reason why new types of scintillators are needed.’

Apart from these practical considerations, the search for new, improved scintillators has also been inspired by scientific curiosity, Dorenbos says. ‘Scintillation is a fairly complicated process, in which various phenomena play a role at atomic levels. Since we are not entirely
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certain how the process works, research on scintillation also forms a scientific challenge.’

**Binding energy**
The scintillation mechanism comprises four stages, Van ’t Spijker explains. It starts with the absorption of the gamma quanta. Absorption occurs through interaction with the atoms of the scintillator’s crystal lattice. Due to the high energy of the quanta – of the order of magnitude of 1 megaelectronvolt (MeV) – it takes a pretty heavy material to halt the quanta. Heavy in this case refers to the atomic nucleus. The heavier the nucleus, the greater the binding energy of the electrons circling it. The greater the binding energy, the higher the chance of interaction.

Dorenbos: ‘This is why lead is generally used as a shield against radiation. Lighter materials will allow a gamma quantum to pass straight through it, but lead will bring it to a halt after a few millimetres.’

**Collision**
The gamma quantum contains so much energy that it can easily shoot an electron from one of the atoms in the crystal lattice out of its trajectory around the nucleus. ‘We’re talking about the electrons in one of the inner shells of the atom. As a result of the collision, the electron will contain almost as much energy as the original gamma quantum. Kicked out of its orbit, the electron slices through the crystal, starting a deluge of ionisation reactions. Electrons will be ejected in large numbers from the outer shell, the valency band, into the conduction band. Depending on the type of crystal, as many as 50,000 to 100,000 can be moved in this way’, Dorenbos says. ‘The distance across which the ionisations occur is in the order of magnitude of 1 millimetre. The time in which they occur is less than 1 billionth of a second.’

**Luminescence centre**
After a while, the electrons knocked from the conduction band will drop back to their original energy level, and continue to complete their laps around the atom’s nucleus. Some of them however, will have migrated through the crystal lattice, to end up around a different type of atom within the crystal lattice, referred to as the luminescence centre.

Van ’t Spijker: ‘The crystal contains about one half of a percent of these centres, as a result of which completely different energy levels will occur locally. If an electron in these spots drops back to the outer shell, a photon will be emitted in the process. In other words, light will be produced, which can be observed.’

The efficiency of the scintillator is determined by a number of different factors. In the first place, it is determined by the efficiency with which the gamma quanta are captured and converted into electrons ejected from the valency band. As mentioned, this requires the presence of heavy atoms. Another important factor is the efficiency with which the electrons migrate to the luminescence centres that have to produce the photons.

Dorenbos: ‘To some degree this is linked to defects in the material. These can be present in the material from

The x-ray source and the detector are positioned at right angles to each other, and revolve around the patient in steps. A 2-dimensional section of the patient is obtained by measuring the transmission level of the x-rays.

Sectional view of the brain obtained using a ct scanner.
the start, but they can also be produced during the scintillation process. Defects in the crystal lattice for example, tend to capture electrons. As a result, the energy of the electron will not be transferred to the luminescence centre. No photon will be produced, so there will be nothing to observe.’

As it happens, not all defects are the same. The chlorides and bromides, the group on which Van ‘t Spijker focused his research, also contain defects in the crystal lattice. These can be caused by the ionising radiation, and contribute to the transfer of energy to the luminescence centre. What’s more, if an electron is ejected from the valency band to the conduction band, this also results in a «hole» in the ion.

In chlorides and bromides, two ions can combine into a «hole trap», a place in the crystal lattice with a positive charge. Van ‘t Spijker and Dorenbos call this a «self-trapped hole». The positive charge, the «hole», can move through the lattice from hole trap to hole trap, i.e. without the ions themselves leaving their place. In addition to the electron current, this results in a hole current through the lattice, as it were. Van ‘t Spijker suspects that the hole trap operates well in the halides, in particularly in chlorides and bromides. The result is that the crystals have high photon yields.

Doping
Van ‘t Spijker focused his attention on crystal lattices containing chlorides or bromides, or lanthanum, gadolinium, and lutetium. These host materials have been artificially doped with slight quantities of trivalent cerium, Ce³⁺, an element from the rare earth range of elements. The scintillators offer high light production, much higher than that of most fluorides doped with cerium. This means that there will be an efficient transfer of energy to the luminescence centres.

However, the decay time is rather long, in the order of magnitude of several microseconds. The decay time is the average time between the moment at which the gamma quantum is captured and the moment at which the crystal emits its photons. It is a measure of the speed at which the energy is transferred. The fact that the speed is so low is a surprise.

Van ‘t Spijker: ‘Actually, we thought that gadolinium in the crystal lattice would act as an efficient way to transfer energy to the cerium, the luminescence centre. We were proved wrong. So, we’re now looking for other elements for the crystal lattice. One of the likely candidates is lutetium (Lu), another rare earth element.’

Patent
Van ‘t Spijker’s research has attracted some interest from commercial circles. The French company Saint Gobain Cristaux et Détecteurs, famous for its glass and crystal, and a main supplier of scintillators, has taken an option on the research results. So far it hasn’t (yet) resulted in a patent, because the new scintillators are still very limited, technically speaking. Even so, the insight gained from the research can form the basis for further development of more efficient scintillators.

‘Anyway, in spite of almost a century of scintillator development, there is still some room for improvement’,
Van 't Spijker reckons. He is now working at the Inorganic Chemistry Group of Prof. Dr Joop Schoonman. There he is doing research into nanostructured solar cells by means of optical techniques and a self-built transient absorption set-up that facilitates the electron movement in such cells upon light absorption. Meanwhile the research on inorganic scintillators at iri continues. Two new scintillators, LaCl₃:Ce³⁺ and LaBr₃:Ce³⁺, were discovered showing properties superior to NaI: Tl⁺, with respect to decay time, energy resolution and light yield. Both materials have been patented.

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A Whole Body Counter is used to measure radioactivity in the body. The patient is placed in lead-screened area to minimize the effect of natural background radiation. The radiation is measured by means of four large cylindrical scintillators.

![Photo multiplier tube](image)

Photo multiplier tube like the ones used for measuring low light levels.

Principle of radiation measurement using a scintillator and a photo multiplier tube. Incoming radiation results in a minute flash of light inside the scintillation crystal. When the light strikes the photosensitive cathode of the multiplier tube, a small number of electrons is released. The electrons are multiplied 100,000-fold as a result of a high voltage applied across the amplifier plates (dynodes). At the output of the photo multiplier tube (the anode), a small electric current is produced, which is a measure of the radiation energy.
The chorus detector at CERN. The detector will be used to find out whether the neutrino has mass or not. For this purpose, a beam of muon-neutrinos will be aimed at the detector. The neutrinos pass through a large quantity of photographic emulsion (800 kg). Every once in a while, a neutrino will interact with an atom’s nucleus within the emulsion, and in some cases this will produce a muon. A number of scintillators is used to detect these muons. The lead calorimeter and the muon spectrometer are used to trace the path of the muons.

Measuring set-up at the Interfaculty Reactor Institute for research on the luminescence properties of scintillators. The crystals are irradiated using an x-ray tube. The light of the scintillation crystals is subsequently analysed using a monochromator and a photo multiplier tube. The monochromator selects light of a certain wavelength.
Schematic diagram of the scintillation process of a LuAlO$_3$: Ce$^{3+}$ crystal, in which the Ce$^{3+}$ has been added as an impurity at a lattice position that would normally contain the Lu$^{3+}$-ion. The gamma quantum enters the crystal. As a result of interaction with the heaviest atom in the crystal lattice (in this case Lu), the gamma quantum is absorbed and the atom becomes ionised. The energy of the gamma quantum is almost completely transferred to the released electron. This electron in turn creates (secondary) electrons and holes, until the resulting electrons and holes have insufficient energy left for further ionisations. The energy is transferred to the luminescence centre (in this case the Ce$^{3+}$-ion), where in the last step, the electrons and holes excite the Ce$^{3+}$-ion, which then emits light.
In many chlorides and bromides, defects in a crystal lattice occur as a result of the absorption of radiation. In common salt, for example, a number of these defects can «walk» through the lattice. When the Cl-ion becomes ionised, a hole is formed in the Cl-ion, or in other words, a Cl\textsuperscript{0} atom is formed. This Cl\textsuperscript{0} atom then bonds with a neighbouring Cl-ion, resulting in a Cl\textsubscript{2} «molecule». This is sometimes referred to as a «self-trapped hole». The molecule can «walk» through the crystal lattice because the hole keeps jumping to neighbouring Cl-ions.

Prototype of a scintillator on a miniaturised photodiode, which is currently being developed at IRI Delft by Ir. Jan Sonsky. The top illustration shows a very small transistor on the photodiode, which acts as an amplifier. The picture below shows the scintillator on the photodiode. This type of radiation detector is much more compact than the current types of scintillator that use amplifier tubes.
Radiation measurement using a scintillator and a light sensitive diode. The light generated after the absorption of radiation strikes the photodiode, which contains a light-sensitive semiconductor material. A number of electrons in the diode will be excited, and will migrate to the cathode. The holes left by the departing electrons will be filled by electrons from the anode. The number of electrons is a measure of the energy of the radiation that struck the scintillator.