# Henri Becquerel, Winner of the 1903 Nobel Prize in Physics: A Change of Hypothesis

**«« By Bryan White, Canadian Nuclear Society**
Bryan White is co-chair of the Education and Communication Committee of the Canadian Nuclear Society. He graduated with an Engineering Physics degree from the University of Saskatchewan in 1970, and a MSc. (plasma physics) in 1972. He worked for Environment Canada, and Fisheries and Oceans before joining Atomic Energy of Canada in 1984. Bryan retired in 2004. He is one of the authors of the CNS Ionising Radiation Workshop that has been presented to science teachers from Halifax to Banff.

***Curriculum Connection: Senior Physics.***

## *Radioactivity Observed*

The Nobel Prize website (www.nobelprize.org) records all the prizes awarded and includes background information as well as copies of the lectures presented by the prize winners associated with the Award Ceremony. In his lecture entitled, "On Radioactivity, A New Property of Matter" Becquerel modestly describes his work from 1896 as having been inspired by the experiments conducted by Wilhelm Conrad Röntgen. Röntgen won the first Nobel Prize in Physics in 1901 for his work with electron beams incident on phosphors in an evacuated glass tube producing invisible rays that were detected some distance from the tube. He called this X-radiation. Becquerel set out to determine whether all phosphorescent material emitted similar rays.

Bequerel chose uranium salts because of the structure of their absorption and phosphorescence (light) spectra. His method was to place sheets of the double sulfate of uranium and potassium on photographic plates that were enveloped in black paper or protected by a sheet of aluminum or glass. The phosphorescent material was exposed to light for several hours. On developing the plates he found that the uranium salt had emitted rays which produced silhouettes of the crystalline sheets through the black paper and the various absorbing layers.

The images were consistent with his hypothesis of solar light having energized the phosphors. However, he soon realized that the emission was independent of any familiar source of excitation energy such as light, electricity or heat. This work continued using different uranium salts to confirm that the emissions were derived from the element uranium. Moreover he observed that metallic uranium was ~3.5 times as active as the salt used in the first set of experiments. He made shadow images of metallic objects placed between uranium-bearing sheets and the photographic plates.

Bequerel reported the observation that the emissions from uranium hastened the discharge of an electrostatically charged object nearby. This facilitated making numerical observations that showed the level of the emissions from the uranium source remained constant. Electrometers became an important tool in the monitoring of such emissions.

It was also observed that an electrostatically charged sphere of uranium in an evacuated vessel did not discharge the electrometer. This led to the conclusion that the emissions ionised the air, increasing the conductivity of the air facilitating the discharge.

Bequerel and his team continued to explore other aspects of the emissions by radioactive materials. Pierre and Marie Curie presented Nobel lectures on their work with radium, and subsequently polonium.

To honour Becquerel's contribution to nuclear science, the SI unit of radioactivity is the becquerel. One becquerel of a radioactive isotope corresponds to the amount of material required to produce a time average of one disintegration per second.

### *A Simple Experiment*

Students of wet chemistry photography have experience with exposing and developing photographic films and print papers to light — and developing the latent image in each case. This technology was once the mainstay of student year books and has now been eclipsed by modern digital photography.

A sheet of sensitive photographic black/white film would be the closest analogue to the plates used by Bequerel. As I do not have access to a real darkroom, my ersatz facility in a basement workshop was more compatible with photographic print paper provided I took advantage of the dark evenings of winter. Using paper has the advantage that the configuration of the exposure geometry and the subsequent development can be performed using "safe lights" — an advantage appropriate to my skill level as it is many decades since I was a yearbook photographer in high school.

Photographic film has been used for personnel dosimeters to monitor the ionizing radiation dose received by workers in nuclear facilities, including hospitals. This too has been replaced by other technologies in most applications. The important point is that sensitive films will register exposures to ionizing radiation intensities greater than background.

Using photographic paper requires much higher intensities and/or much longer exposure times to produce a latent change (image). With such low interaction rates, the photosensitive material is less effective… a phenomenon termed "reciprocity failure."

Attempts to develop a "source self-portrait" using weak sources such as common Naturally Occurring Radioactive Materials and consumer products were disappointing. Ultimately, I resorted to the most intense alpha/beta/gamma source at hand, a vintage lens from a single lens reflex camera. The techniques are compared in the table below.

| Aspect of the Experiment | Bequerel | White |
|---|---|---|
| Photosensitive material | Glass plates with photographic emulsion | Black and white photographic print paper |
| Absorber | Opaque paper | None, paper, aluminum foil, paper and foil |
| Source | Natural uranium salts, uranium metal | Thorium oxide in glass lens |
| Exposure times | Not stated, but seems to be days | Many months (~10) |

### *Thorium Source*

The use of thoriated glass lenses as a convenient source of alpha, beta and gamma is described in the CNS Ionising Radiation Workshop Notes (Appendix B). A brief summary follows:
(**http://www.cns-snc.ca/media/uploads/teachers/Ionising_Radiation_Workshop_notes_R11.pdf**)

- Heavy metals are added to glass to make high quality lenses for photographic cameras and television cameras. This increases the electron density in the glass and hence the index of refraction. Compact lenses with high performance and low dispersion are obtained.
- Over the period between 1950 and 1980 (approximately) lens makers used thorium oxide for this purpose as it is more economical than using other metals such as lanthanum. The US Nuclear Regulatory Commission allowed the use of up to 30% thorium oxide in glass lenses.
- As the lenses are ground, the thorium oxide is present throughout the lens and at the surface apart from very thin coatings applied to reduce reflections. This makes it an effective alpha source.
- Thorium-232 is the most dominant thorium isotope. It has 10 steps in the decay series to lead-208. (There are two branches at the penultimate decay from bismuth-212.)
- When the lens makers prepared the thorium oxide, their chemical procedures would have reduced the concentrations of the members of the decay chain.
- The Th-232 chain is interesting as Th-232 has a half-life of $1.4 \times 10^{10}$ years, and all but two members have half-lives of days or less, the exceptions being radium-228 at 5.8 years, and thorium-228 at 1.9 years. As a consequence, after ~ 6 half-lives of the longest member, the decay series approaches **Secular Equilibrium**. In this case after 35 years all the members of the decay chain (apart from the branching) reach the same level of activity as the Th-232. This acts as a "multiplier" of the Th-232 activity.
- As it is over 40 years since I purchased my thoriated SLR lens new, it has reached this condition (Mamiya/Sekor f1.4 55 mm purchased in 1970).
- Tests with a Geiger counter demonstrated that the rear element of the lens provide the highest activity.

### *Preparation of the Test Assembly*

1. After my initial experiments I assembled a holder for a piece of print paper 8 x 6.5 cm. This provided the four exposure conditions described above.
2. The photographic print paper was inserted in the holder (emulsion side up) under safe lights.
3. The holder was placed on the inside-up (dark brown) plastic lid of an empty hot chocolate drink mix container (aluminized cardboard with metal end) and the camera lens was placed on the assembly centred over the exposure absorbers.
4. The empty container was lowered over the assembly and secured into its lid.
5. This container was then placed on the inverted dark brown plastic lid of an empty metal coffee can.
6. The coffee can was then lowered over the assembly and secured to its lid.
7. The plan was this would be left undisturbed for months.
8. Unfortunately, things happen and I had to move the assembly. Now I wish that I had used a scheme to hold the lens in place.

### Results

When winter returned and my dark room was again useful, I opened the assembly under safe lights and developed the image. Alas the image reveals that the lens occupied three overlapping positions over the exposure period. It is fortunate that the durations for the three positions are not similar as it is possible to resolve some of the more subtle features.

The images shown below include a result that may be related to the foil-scattering experiments described in the Workshop Notes (Geiger Experiment 3 Part I). The notes describe observations with a thorium source using aluminum foils and paper as absorbers. Alpha particles incident on an aluminum foil are shown to forward-scatter electrons from a foil. Adding a paper absorber in front of the aluminum foil absorbs the alpha radiation without scattering electrons.

As the alpha radiation detected is emitted only from the surface layer of the lens, it is apparent that the alpha radiation is more effective at producing an image than the beta or gamma radiation which provide the majority of the counts observed with a Geiger detector as demonstrated with a paper absorber.

### Remarks

The print paper is a poor choice compared to using more sensitive photographic film for similar experiments if a suitable dark room is available. The roll film used in "large format" cameras ("120" or "620") could be used with a suitable film holder rather than flat sheet film.

As Bequerel shielded his photographic plates from light with opaque paper one may conclude that his images were produced by beta and gamma radiation — the alpha radiation being absorbed by the paper. It seems likely that his source was more intense than this camera lens. Note that Bequerel's subsequent experiment with an electroscope in a bell jar would not have included the paper shield and the alpha radiation may have made a significant contribution to the ionization of the air.

**Rear element of Mamiya/Sekor f1.4 55 mm lens**

Note the brown tint due to radiation damage creating colour centres in the glass. Apparently this may be reduced by "bleaching with intense ultraviolet light" or by disassembling the lens and baking the optical elements in an oven.

Developed image with print partially removed from the absorber assembly to show the images.

The image shows the lens was in three positions during the exposure period.

The darkest regions correspond to no absorber between the lens and the print paper. The alpha radiation contribution dominates the exposure in these regions.

The areas with the paper or aluminum foil absorbers show comparable intensity with the foil possibly associated with a lighter shade and thus providing greater absorption than the paper.

Below the white paper the aluminum foil is cut at an angle. For the lightest of the three images, the paper plus foil provides more absorption than the aluminum foil alone.

**Rear element of Mamiya/Sekor f1.4 55 mm lens**