Curies Contaminated Notebook

An analysis of a notebook and papers, originally belonging to Marie Curie, which are now retained by the Wellcome Collection, London.

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Abstract. This paper describes the radiological analysis of a notebook and papers originally belonging to Marie Curie, which were identified as contaminated. The analysis used direct monitoring techniques, wipe samples and gamma spectrometry to identify the type and extent of contamination. The subsequent work to ensure the safe keeping and handling of the notebook illustrated that the UK's legislation for radiation protection and keeping of radioactive materials does not easily cover such items.

KEYWORDS: Curie; radium; legacy; contamination; gamma spectrometry; radium-226; thorium-232.

1 INTRODUCTION

In September 2014 Aurora Health Physics Services Ltd (Aurora) was approached by Health and Safety Advisers at the Wellcome Trust concerning a notebook kept within the Trusts library collection in London. The notebook had originally belonged to the scientist Marie Curie, who had used the notebook to record laboratory results of her experiments on radioactivity. Preliminary monitoring with hand-held contamination instruments had indicated that the item was contaminated with radioactive material; however, the isotopes and activity of the material were unknown. The Wellcome Trust was keen to ensure that staff and visitors were adequately protected from exposure to the notebook, but still wanted to allow public access to view the item if possible. The Trust were also required to have demonstrable evidence of compliance with the relevant regulatory requirements for keeping and using the notebook.

2 BACKGROUND OF THE CURIE NOTEBOOK

2.1 Curies work with Radium

Marie Curie and her husband Pierre Curie are well known for their experiments with radioactive material in the early 20th Century. The Curies are credited with the discovery of both Radium and Polonium in addition to coining the word "radioactivity". Their method to isolate Radium-226 from pitchblende required considerable manual labour to grind and chemically process the mineral in very large quantities. It is now known that the Curies were exposed to significant radiation during this work at their laboratory in Paris.

2.2 The Wellcome Trust and the Wellcome Collection

The Wellcome Trust was established in 1936 after the death of Sir Henry Wellcome. Throughout his life Sir Henry Wellcome amassed an extensive collection of books, journals, manuscripts, prints and drawings on the broad topic of medical history. After his death he also left substantial sums of money to be invested in progressing medical advancements. Since 1936 the Wellcome Trust has financed a number of major biomedical research initiatives. The Wellcome Collection in London has now grown into a library of over 750,000 items, the majority of which can be viewed and handled directly by the general public.

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2.3 'Discovery' of the Curie notebook at the Wellcome Collection

In August 2014 it came to the attention of the Health and Safety Team at the Wellcome Collection that a library user had flagged that workbooks and possessions of Maries Curies, similar to that of the notebook held in the London Collection, were kept at the Bibliotheque Nationale in France. The user mentioned that in France, those who want to view and touch the items must 'sign a waiver and wear protective clothing'. Unsure as to whether their Curie notebook may also be contaminated, the Health and Safety Team in London quickly withdrew the item from the Collection catalogue and sought help from radiation safety professionals.

2.4 The notebook, associated notecards and papers

The notebook is approximately 15cm x10cm in size with a hardbound front and back cover. The label on the front (see Figure 1) has Marie Curie's name and the date, 1920, and is somewhat damaged, but otherwise the notebook is in good condition. The book is handwritten in French and contains experimental method details, results and sketches. Within the notebook were five loose notecards, also with experimental records. Also tested were three sheets of paper: two letters/ notes (see Figure 2) and the other a certificate. Records show that the notebook was originally purchased by the Collection in the early 1960's.

Figure 1: Front cover of the Marie Curie notebook

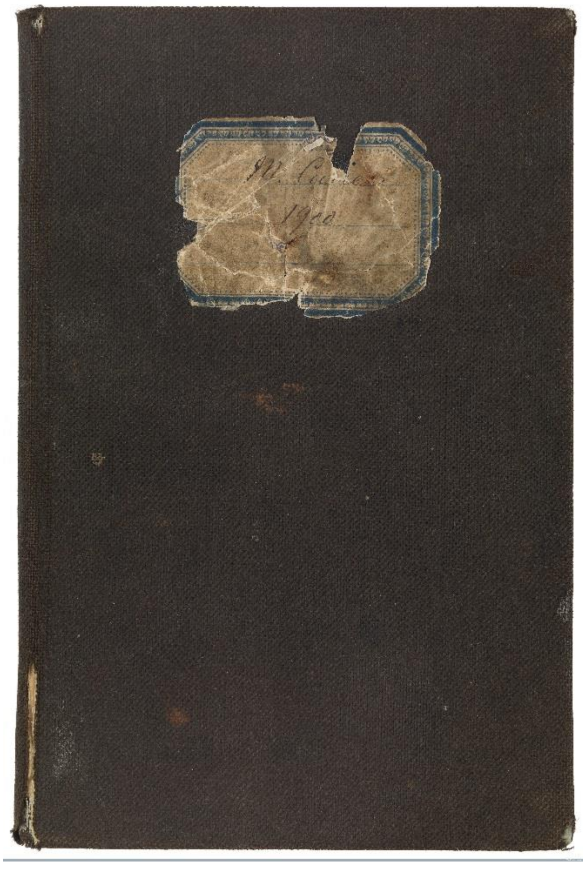
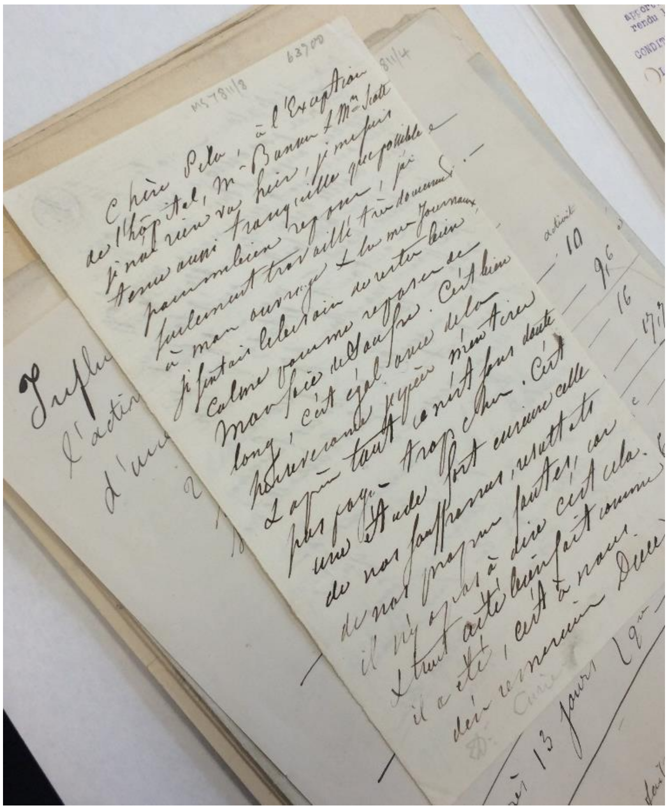


Figure 2: Marie Curies writing on separate sheets of paper



3 RELEVANT LEGISLATION IN THE UNITED KINGDOM

3.1 Ionising Radiations Regulations 1999 (IRR99)

IRR99 are enforced by the Health and Safety Executive. The aim of IRR99 is to ensure that doses to radiation workers and members of the public are kept as low as reasonably practicable and that dose limits are not exceeded.

3.2 Environmental Permitting (England and Wales) Regulations 2010 (EPR10)

EPR10 are enforced by the Environment Agency (EA). These regulations aim to establish and maintain control over the keeping, use and security of radioactive materials and also to ensure that the accumulation and disposal of radioactive wastes are managed effectively to limit radiological impact on the general public and the environment.

3.3 Environmental Permitting (England and Wales) Regulations (Amendment) 2011 (EPR11)

EPR11 outline the conditions under which radioactive materials should be managed in circumstances where EPR10 does not apply. This essentially exempts minor practices; however, a number of conditions must still be met for exempt items.

4 SEARCH FOR OTHER POTENTIALLY CONTAMINATED LIBRARY ITEMS

Before the initial survey of the notebook, library curators were also asked to consider if the Collection included any other items on the theme of ‘radioactivity’ or ‘radium’, or work from other scientists of the time (such as William Crookes), which may also be contaminated with radioactive material. This search resulted in fifteen bulk boxes of books and documents which were taken from storage for analysis by Aurora, in addition to the Curie notebook.

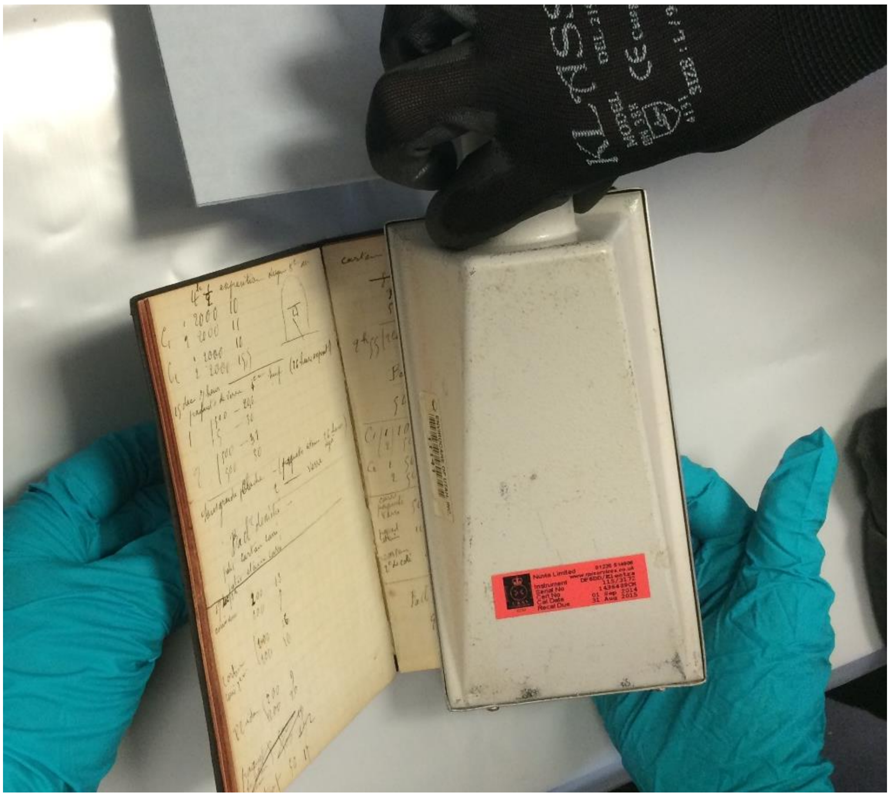
5 ANALYSIS

Analysis of the identified items was carried out using a range of methods. Direct monitoring was initially used to search for items contaminated with beta-gamma emitters, where the contaminated items were found. Wipe tests were used to test for loose contamination. Finally, gamma spectrometry analysis was carried out on all bulk boxes of items and on the Curie notebook in isolation.

5.1 Direct Monitoring

The notebook and other items were monitored using an Electra Ratemeter with a DP6 dual phosphor probe (Figure 3). The highest levels of contamination were found on the outside cover of the notebook up to 2,500 cps alpha and up to 3,500 cps beta corresponding to an estimated 200 Bq/cm^2 Radium-226 for a porous surface. The inside pages of the notebook were less contaminated with count rates up to 60 cps alpha and 2,000 cps beta. The other items in the collection had much lower levels typically less than 10 cps alpha.

Figure 3: Direct monitoring of the Marie Curie notebook using DP6 probe



5.2 Wipe Tests

Items, where the presence of radioactive material was confirmed by direct monitoring, were also subject to wipe testing. This consisted of a filter paper being wiped across the surface of the item in order to pick up any loose contamination. The wipe samples were screened initially with a hand held probe and then were later analysed in Aurora's laboratory using two high sensitivity alpha and beta drawer counters. The results of the wipe tests are provided below in Table 1. Loose contamination was present but was found to be below the typical action levels for establishing a contamination controlled area (0.4 Bq/cm^2 for loose alpha contamination and 4 Bq/cm^2 for loose beta contamination).

Table 1: Wipe test results

Description	Loose Alpha (Bq/cm^2)	Loose Beta (Bq/cm^2)
Letter 1	0.024	0.000
Letter 2	0.018	0.108
Certificate	0.003	0.051
Notebook Front Cover	0.033	0.379
Notebook Back Cover	0.042	0.740

5.3 Gamma Spectrometry

The Aurora High Resolution Gamma Spectroscopy (HRGS) system uses an Ortec trans-Spec-DX-100 detector with a 67 mm × 52 mm high purity germanium P-type crystal. The detector is electrically cooled meaning that liquid nitrogen is not required. Although the detector takes 12 hours to cool from room temperature, Aurora routinely maintains the detector in a constantly cooled state so that it can be deployed for immediate measurement.

The trans-Spec-DX-100 was set up in within a location which was chosen with consideration to minimising background radiation readings. A background spectra was collected in order to subtract the background from measurements of items in the collection.

Each item was weighed and analysed for five or fifteen minutes. The collected spectra were post processed using software to provide spectral analysis and nuclide identification and this information, together with the data from the calibration model, provided quantification of the radioactivity present. By carrying out background subtraction and using a geometry calibration file it is possible to calculate the total activity in the measurement volume.

The minimum detectable activity is dependent on the counting geometry, the density of the package and the count time. Typically, the minimum detectable activity during this work varied from 1kBq to 5kBq of Radium or Thorium activity in a box or package.

Table 2: Gamma spectrometry results

Item	Radium-226 Activity (kBq)	Thorium-232 Activity (kBq)
Notebook	120	1
Letters (Combined)	3	1

6 RECOMMENDATIONS TO THE COLLECTION FOLLOWING ANALYSIS

6.1 Public handling of the notebook and papers

The survey and sample analysis results were used to provide a radiation dose assessment for handling and inspection of the items containing radioactivity. Conservators and members of the public could be exposed to contact dose rates of up to 3.5µSv/h although whole body dose rates are only marginally above the background of 0.1µSv/h. Even regular exposure to the identified items is likely to result in annual whole body doses of less than 10µSv (or 0.010mSv) and hand doses of less than 35µSv

(0.035mSv). The dose limit to members of the public from an employer's activity is 1,000μSv (1mSv) whole body and 50,000μSv (50mSv) to the hands.

Loose activity could be removed from handling the notebooks at levels similar to those removed during the wipe test; if the handling of items was not carefully supervised some of this activity could be ingested by hand to mouth contact. Up to 10Bq of activity was removed from the items during the wipe tests. Ingestion of this level of activity would result in a Committed Effective Dose of 22μSv which is again well below the above limits.

6.2 Complying with IRR99

A prior radiological risk assessment was carried for handling of the notebook by staff and members of the public. Many of the existing controls for physical protection of valuable historical documents such as wrapping, gloves and supervision also provided a high degree of protection from the radiological hazard. Additional recommendations on storage and supervision procedures were made.

6.3 Environmental Permit

The Environmental Permitting (England and Wales) Regulations 2010 apply to naturally occurring radioactive materials (NORM) but only to specific industrial practices. The notebook could not be classified in any of the specified practices and after consultation with the Environment Agency the Wellcome Trust applied for and received an Environmental Permit for the keeping and use of open radioactive sources, but with no allowance for radioactive waste to be generated.

7 WIDER CONTEXT

The discovery of the contamination present in the Curie notebook and papers presents a number of questions about other items of a similar nature that could be unknowingly kept across the globe. However, it's difficult to estimate the number of items, their level of possible contamination, or their location. Considering the likely public perception of the discovery of such items in the public domain, the radiation protection community would be wise to be ready to respond to these situations quickly in order to ensure that dose implications are understood and misinformation is prevented from spreading.

It could be argued that the radiation protection community should have some responsibility in helping finding these items, though who provides the resource to search for and then keep such items securely would depend on each individual situation. In some cases, it may transpire that such an item cannot be kept or used safely, in which case those responsible will have to ask themselves at what point is safety more important than preserving historical artefacts.

8 CONCLUSION

Analysis confirmed that the Collections notebook from Marie Curie contains approximately 120kBq of Tadium-226 and 1kBq of Thorium-232. Additional notecards and papers from Marie Curie also contain 3kBq of Radium-226 and 1kBq of Thorium-232, with some evidence that this contamination is loose.

The documents present a very low external radiation hazard, but contamination when touching or the items is possible. The Collection have now implemented a number of controls to ensure compliance with the relevant legislation within the UK.

9 ACKNOWLEDGEMENTS

The author would like to acknowledge the efforts of the Wellcome Collection Conservation team for their work to ensure compliance with all the necessary requirements to keep the contaminated items on site.

Thanks to staff at Imperial College London for their initial help to the Wellcome Trust to identify the notebook as containing radioactive material.

Thanks are also extended to Amber Bannon and Phil Fahey who carried out the site inspection at the Wellcome Collection on behalf of the Environment Agency, shortly after the initial analysis. Their help to understand legal requirements for keeping of the items is much appreciated.