Neutron tomography study of a lithium-ion coin battery

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Abstract. Neutron imaging of lithium-ion coin cell battery was obtained using tomography technique at Neutron Scattering Laboratory in Multi-Purpose Research Reactor G. A. Siwabessy (RSG-GAS) Serpong facility. The coin cell battery was CR2032 (20d x 3.2t mm) type consisted of positive and negative cases made of stainless steel, a cathode layer from lithium Ferro phosphate coated on aluminum foil, polyethylene film as separator, lithium hexafluorophosphate solution in ethylene carbonate and diethyl carbonate as electrolyte, an anode layer from graphite coated on copper foil, and spacer as well as spring made of stainless steel. The neutron tomography was used to observe the inside structure of the full charged coin cell. The observation was carried out with a neutron flux of $10^7$ n/s.cm$^2$ at 15 MWatt of reactor power. The image of the object is obtained by a Charge Coupled Detector (CCD) and the reconstruction software based on a filtered back-projection algorithm. The neutron imaging clearly shows the cell structure inside the casing. Even though the cell is not at the center of the casing, but it appears that the cell component consists of a cathode, separator and anode neatly arranged. The cross-cut image shows the important role of spring and spacer in improving the contact between cell and casing. The results showed the ability of neutron tomography techniques at RSG-GAS to investigate in detail the inside structure of a coin battery without disassembly (non-destructive test).

1. Introduction
Lithium-ion battery (LIB) is a type of secondary or rechargeable batteries. LIBs are widely used in various mobile devices such as smartphones, laptops, and so on for more than two decades. LIB, having high power and energy density, is a promising candidate for application in electromobility and stationary energy storage [1], [2], [3], [4] 

LIBs have three main functional components, those are the positive and negative electrodes and electrolyte. Generally, the negative electrode or anode component of LIB is made from carbon. While the positive electrode or cathode is made from a metal oxide, and the electrolyte is a lithium salt in an organic solvent [5]. LIB can be found in the form of a coin or button, cylindrical, and pouch cells.

Coin cells are in the form of small discs, very light, great for small, low-power devices. One of the most popular coin cells in use right now is the CR2032 which is 20mm diameter x 3.2mm thick. Coin cells are also called micro-batteries, often lithium cells are used (3V) but alkaline, zinc-air, and manganese are also used (1.5V). Lithium cell batteries must be distinguished from LIBs. Lithium cell batteries are referred to as non-rechargeable batteries in which metal lithium is used as the anode. While LIB is rechargeable batteries which generally using lithium compounds as cathode and carbon as the
anode. Some examples of the lithium compounds are lithium cobalt oxide (LiCoO$_2$), lithium ferrophosphate (LiFePO$_4$), lithium-ion manganese oxide battery (LiMn$_2$O$_4$, Li$_2$MnO$_3$, or LMO), and lithium nickel manganese cobalt oxide (LiNiMnCoO$_2$ or NMC) [6, 7, 8, 9].

In this study a rechargeable battery in form coin cell has been assembled. The coin cell battery was CR2032 type consisted of positive and negative cases with spacer and spring made of stainless steel. The cell inside was constructed from a cathode layer from lithium ferrophosphate (LiFePO$_4$) coated on aluminum foil, polyethylene film as the separator, lithium hexafluorophosphate (LiPF$_6$) solution in ethylene carbonate and diethyl carbonate as the electrolyte, and an anode layer from graphite coated on copper foil. To study the inside structure of the assembled coin, a neutron imaging experiment has been done using the neutron tomography technique.

Neutron tomography is a form of computed tomography to acquire three-dimensional information related to the structures inside a sample. It uses radiographic projection images from many views to reconstruct the distribution of materials in the sample. By this method, the inner structure of the sample can be visualized. This technique usually used to observe most of the industrial products particularly to investigate the internal structure of materials made of a mixture of metals and non-metals [10]. Neutron Imaging uses a rotate table that can move from 0 to 360 degrees with a data recording every 1° movements [11]. Although basic principles are known for decades, this technique became more used in recent times, due to the availability of digital neutron area detectors and increased power of computers [12].

In this study, the neutron imaging experiment of the assembled CR2032 type lithium-ion coin cell battery was done using the tomography technique at the Multi-Purpose Research Reactor G. A. Siwabessy (RSG-GAS) Serpong. The aim of this study mainly is to obtain the inside structure of the coin cell battery, the component as well as how space and spring are playing a role.

2. Experimental

2.1. Coin battery assembly

A commercialized electrode sheet (MTI Corporation) was used as the battery component. Aluminum foil single side coated by LiFePO$_4$ (LFP) and copper foil single side coated by CMS Graphite were used as cathode and anode, respectively. The cathode sheet was consisting 15 microns aluminum foil as a current collector and lithium ferrous phosphate as an active material with a material density of 120 g/m$^2$, in which the active material weight was 4.566g with 91.00% proportion in powder. While the anode counterpart was consisting of 9-micron copper foil as a current collector and graphite composite as the active material. SBR+CMC was used as the binder with active material proportion in powder was 94.5% or 120 g/m$^2$ in density. The electrode sheet was kept in a vacuum oven at 80°C for at least 3 hours before assembling. The electrolyte was 1 M LiPF$_6$ dissolved in an ethylene carbonate/dimethyl carbonate/ethyl methyl carbonate (EC/DMC/EMC) solution. The electrodes and separator were assembled into a standard coin cell (CR2032), and the liquid electrolyte LiPF$_6$ was put into the cell and then sealed using the hydraulic crimping machine in a moisture-controlled glove box. The structure of the coin battery components are shown in Figure 1

![Figure 1. Structure of the coin battery components.](image)

2.2. Battery performance characterization
The electrochemical measurements were carried out using BST8-3, 8 Channel Battery Analyzer with cut-off voltages of 2.5-4.2 V was used to examine the charge/discharge performance. The testing was done by using a constant current-constant voltage (CC-CV) charging system: in the first five cycles, the cells were charged to 4.2 V at 0.1 C (1 C= 170 mAh g⁻¹), kept at 4.2 V until the current dropped to 0.01 C, then discharged to 2.5 V at 0.1 C; repeating, the cells were charged again to 4.2 V at 0.1 C, kept at 4.2 V until the current dropped to 0.01 C, then discharged to 2.5 V. Note that the electrochemical capacity of the sample was calculated based on the amount of pure active materials excluding the coated carbon. The battery performance characterizations were conducted at the Integrated Battery Laboratory, National Nuclear Energy Agency (BATAN), Serpong, Indonesia.

2.3. Neutron tomography measurement
In order to observe the assembly quality, the obtained coin battery was analyzed by tomography at Neutron Scattering Laboratory, PSTBM, BATAN. A neutron tomography facility has been completely installed at the S2 tangential beam port of the G. A. Siwabessy Multipurpose Reactor (RSG-GAS). The neutron tomography uses a filter-back projection for reconstruction and analyzes it based on the density of the detected sample. The data were collected from 0 to 3600 position of the sample. The data recorded in a 2-dimensional image in 1024x1024 pixel resolution and reconstructed by neutron tomography software to get the virtual 3D data. An Octopus VGStudio 8.5 Software and 0.0735 pixel / mm optic lens resolution with a 7.5 Cm point of view (POV) were used [13].

3. Results and discussion

3.1. Coin battery assembly
Figure 2 shows the components used in the lithium-ion coin battery assembly. Anode/ cathode pair material can not only be made from lithium Ferro phosphate/ graphite but can also be made from lithium cobalt phosphate/ graphite, lithium manganese phosphate/ graphite, lithium Ferro phosphate/ graphite, lithium Ferro phosphate/ LTO, lithium cobalt phosphate/ LTO, lithium manganese phosphate / LTO, and others.

Figure 2. Components of a coin battery.

Figure 3 shows the photograph of the assembled CR2032 coin cell battery. Battery assembly began with a cathode made of aluminum foil printed and coated with lithium Ferro phosphate. It was done by using a disc cutter outside the glove box, the printed result in the electrodes form with 16 mm diameter. A Separator made of polyethylene was cut by a disc cutter to produce a coin separator in 18 mm diameter, while the anode material has a 15 mm in diameter. Before assembly, the cathode, anode, and
separator were dropped by three electrolyte LiPF$_6$ liquid. Battery assembly began by placing the positive case as a container and compiled them with cathode, separator, anode, spring, and spacer. Those steps were considered for the installation of battery components, especially for placing the separator step in order to avoid a short circuit between the anode and cathode.

![Battery components](image)

(a) (b)

**Figure 3.** Lithium-ion coin cell battery, positive (a) and negative (b) side.

### 3.2. Battery performance

The electrochemical performance of the coin cell battery is shown in Figure 4. It is clear that the battery was working properly as energy storage with capacity and voltage of about 1.1 mAh and 3V, respectively. However, the capacity decreased with the cycle number. The battery performance may be affected by the component arrangement inside the cell. The contact between the cell component and the casing may also play an important role. Bad contact between cell components and casing will make the battery working improperly. To observe the inside structure of the coin battery without disassembly, the neutron imaging has been conducted.

![Charge-discharge performance](image)

**Figure 4.** Charge-discharge performance of the coin cell battery 21-30 cycles.

### 3.3. Neutron tomography image

Non-destructive testing performed by the tomographic method that is on file Technology Division facility Neutron-BATAN. This method shows that the results of the physical condition of the coin...
battery, especially to show the state of the components in the coin battery after crimping as shown in Figure 4.

**Figure 5.** Tomography test results on coin battery samples as an outer side (a) longitude slice (b) cross-cut slices (c) and vertical arrangement image (d)

Figure 5 (a) shows that a visible of an outer side of the coin battery in good condition. Fig. 5 (b) shows the longitude slice of the coin battery lithium anode position on proper assembly position, ie the middle case and separated from the cathode to the separator polyaniline. This display also showed that the lithium anode did not contact with the positive case, thus mean the battery assembly was correct. Figure 5 (c) shows the image slices of the coin battery. The image showed that the vertical arrangement of the components of the coin battery has appeared and the spacer at the position in the middle of the case. The neutron imaging clearly shows the cell structure inside the casing. Even though the cell is not at the center of the casing as shown in Figure 5 (b) and (d), it appears that the cell component consists of a cathode, separator and anode neatly arranged (Figure 5 (d)). However, the cathode and anode layer cannot be distinguished from the image due to a lack of resolution. The cross-cut image shows the important role of spring and spacer in improving the contact between cell and casing (Figure 5 (c)).

**Figure 6.** Image dimensional tomography test results coin battery

Figure 6 shows a 2-dimensional image of the coin battery that can demonstrate precision measurements of the components of the coin battery. This information was very useful in addition to
knowing the battery real size. It can also determine the position of damage point base on the battery components assembly, especially for the battery inside.

Neutron tomography was a very useful technique for measuring the coin battery quality, particularly for electrical properties or mechanical test. In this case, neutron tomography showed that there was no contact between the active material and the battery casing. Contact with a case was not expected to avoid battery damage.

4. Conclusion
The assembled coin cell battery was working properly as energy storage. Neutron imaging clearly shows the cell structure inside the casing. Even though the cell is not at the center of the casing, but it appears that the cell component consists of a cathode, separator and anode neatly arranged. However, the cathode and anode layer cannot be distinguished from the image due to a lack of resolution. The cross-cut image shows the important role of spring and spacer in improving the contact between cell and casing.

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