

## Development of high-contrast developing for nuclear emulsion film for GRAINE experiment

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The GRAINE experiment is the balloon flight experiment with emulsion chamber telescope for observing of gamma rays that have energy range of 10 MeV to 100 GeV. We launched the emulsion chamber telescope in Australia in the April, 2023 (GRAINE2023). GRAINE2023 have the larger aperture area than previous experiments, and the aperture area increase from 0.38 square meters to 2.5 square meters. The nuclear emulsion film is the kind of photographic film, and we can observe emulsion films on the microscope after the development. The scale of experiments with the nuclear emulsion film increases, the Hyper Track Selector (THS) was developed for convert track in the nuclear emulsion film into data at high speed, and have been producing success. Additionally, the HTS2 was developed for improvement of the scanning speed over THS, by lowering the magnification and widening the field of view of the captured image. On the other hand, the THS2 is concerns about the deterioration of the detection performance of developed silver. As one way, to improve the detection performance of developed silver, we changed the developer, which makes enlarge developed silver grains. So far, we have used XAA development, but we made development possible to adjust the size of developed silver grains by mixing chemicals with ourselves. We study and develop of high-contrast developing for the nuclear emulsion film and expand of development scale ,and report the result of developed emulsion films of GRAINE2023.

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## 1. Introduction

Gamma-ray astronomical observations in the GeV region have made great progress with the Fermi-LAT launched by NASA in 2008, and the angular resolution has improved year by year by observing and collecting statistics for several years. Fermi gamma-ray observations over eight years have reported 5064 gamma-ray objects in the range from 50 MeV to 1 TeV. On the other hand, it is also reported that 26% of these observed gamma-ray objects are unidentified objects[1].

Observations in this energy band lag behind observations in other wavelengths of electromagnetic waves, and recent advances in observational techniques are expected to lead to further understanding of gamma-ray high-energy astronomical phenomena. One of the lagging observation technologies is the improvement of angular resolution.

### 1.1 GRAINE experimet

The GRAINE(Gamma-Ray Astro-Imager with Nuclear Emulsion film) experiment is the joint experiment by using the balloon-borne gamma-ray telescope with high spatial resolution emulsion films. The purpose of the GRAINE experiment is to analyze the gamma-ray imaging of high energy astronomical objects of 10 MeV to 100 GeV band in high detail. We are expected that the nuclear emulsion film telescope used in this study will advance the obsevation of gamma-ray astronomical objects in high angular resolution[2].

The main detector consists of converter, time stamper[3], and three star cameras, and it has 2.5 m<sup>2</sup> aperture area. The converter part consist of twenty blocks that the one block is ninety stacked emulsion films, and the one emulsion film has 250 mm × 500 mm area. The role of the converter part convert gamma-rays into electron-positron pairs, and measures the energy of particles. The time stamper part consist of the multistage shifters, and the three star cameras for evaluates the attitude of telescope by registers the stellar objects.

In April 2023, the emulsion telescope was launched to Longreach from Alice Springs, Australia (GRAINE2023). Flight duration is 24 hour at 36 km altitude. We send back collected detector to Japan, and when detector arrived, we started development at Gifu University.

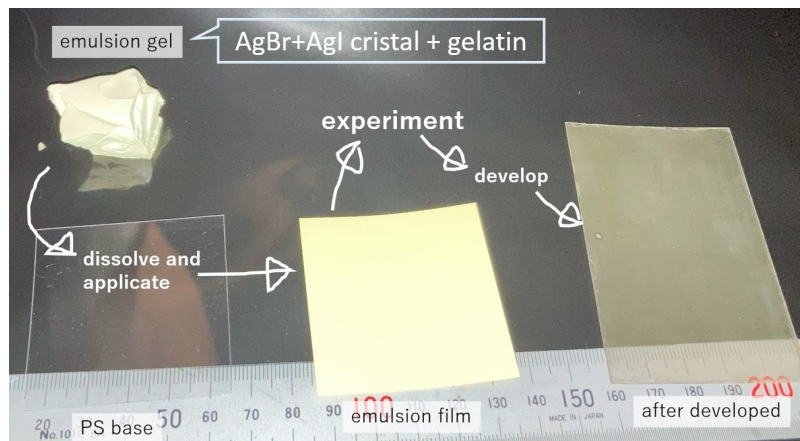


Figure 1: GRAINE2023

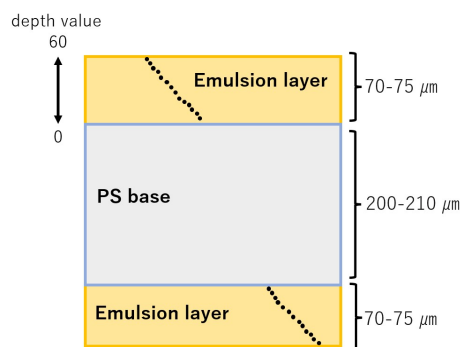
### 1.2 Nuclear emulsion film

The nuclear emulsion film is a track detector type of the photographic film coated nuclear emulsion gel, which is mixed AgBr in gelatin, on a plastic sheet base or on a glass base. The

photosensitive nucleus in nuclear emulsion film is sensitized when charged particles pass through the nuclear emulsion film, and is recorded as developed silver via develop the sensitized emulsion film. Figure. 2 shows procedure of emulsion film production and process. The emulsion film in the middle of the Figure. 2 is used for observation. And the emulsion film becomes transparent in the translucent of development. The developed silver can be observed using a microscope. By taking micrographs, we can analyze the track of charged particles. Track detectors include fog chambers and bubble chambers in addition to the nuclear emulsion film. A feature of the nuclear emulsion film is that it has a high spatial resolution of less than  $0.1 \mu\text{m}$ . The nuclear emulsion film used in this study and GRAINE2023 is a plastic sheet base coated with nuclear emulsion gel on both sides to a thickness of  $70 \mu\text{m}$  to  $75 \mu\text{m}$  (Figure. 3).



**Figure 2:** The procedure of emulsion film production and process. The first procedure is production of nuclear emulsion gel. In the middle of figure is emulsion film that can be used for observation, and emulsion film in the middle of figure is produced gel applied on a plastic sheet base. The emulsion film can be observed using microscope via developing after experiment.



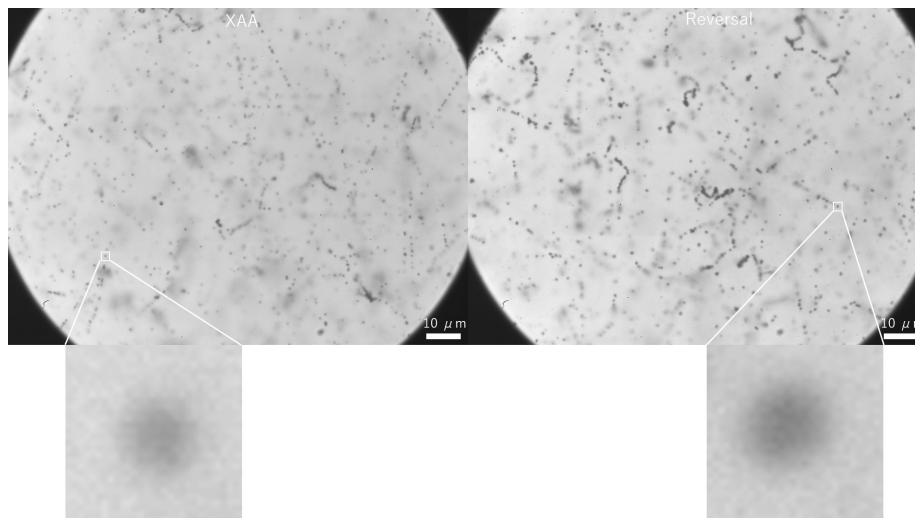
**Figure 3:** Side view of one film. The black points is developed silver particle. A series of silver particles is a track of charged particle.

After developed, we can get track data for analyze using Track Selector via swelling. Track Selector was developed for convert track in the nuclear emulsion film into data. The scale of experiments with the nuclear emulsion film increases, Track Selector was developed with increased speed, Hyper Track Selector[4] (HTS) and HTS2. The HTS2 was developed for improvement of

the scanning speed over HTS, by lowering the magnification and widening the field of view of the captured image. On the other hand, as a result of lowering the magnification, the HTS2 is concerns about the deterioration of the detection performance of developed silver. To improve the detection performance of developed silver, we changed the developer, witch makes enlarge developed silver grains.

## 2. Developer changes and improvements of developed silver

We changed developer for developing of high-contrast development for the nuclear emulsion film. The developer we have used so far is a developer developed for OPERA experiment, called XAA developer. The developing agent of XAA developer is 3.0 g/L Phenidone and 15 g/L ascorbic acid. And the developing agent of the newly developer for high-contrast development, called reversal developer, is 1.5 g/L Phenidone and 20 g/L Hydroquinone. This difference is mechanism of development, and there calls chemical development or physical development. The mainly developing mechanism of XAA developer is chemical development, and of reversal developer is physical development. Another difference is developing temperature. The XAA developer is 20 °C and Reversal developer is 15 °C. Developed time is 25 minute at both. Figure. 4 shows the photomicrograph of different developer, XAA developer (left picture) and Reversal developer (right picture). The bottom pictures are enlarged image of developed silver called grain. The grain of Reversal developer is easier to see and dark than XAA developer.



**Figure 4:** The photomicrograph of different developer. Left picture is XAA developer, and right picture is reversal developer. Bottom pictures are enlarged image of grain.

After each development for comparing XAA developer and Reversal developer, we scanned counts, position and brightness of grain using track selector. Figure. 5 shows relationship between max brightness and depth of grain, and color map is the number of grain. The high value of depth is defined as surface and the low value as deep (show Figure. 3). The max brightness is darkness point of one grain, and the high value of max brightness is dark and the low value is bright. The left figure is XAA developer, and the right figure is Reversal developer. The number of grain is decrease

with depth in XAA developer, but is not change with depth in Reversal developer. The decrease in the max brightness of the XAA developer indicates a decrease in grain contrast with increasing depth. The cause is size of grain. Bright grain indicate smaller grain. Figure. 6 is histogram of max brightness in XAA developer (left graph) and Reversal developer (right graph). The arrows in the graph indicate approximate peak value each other. The peak values differ by about 20, and grains appear darker in Reversal development. The low value of max brightness is noise. For the XAA developer the signal is also mixed into the low values, but can be ruled out for the reversal developer.

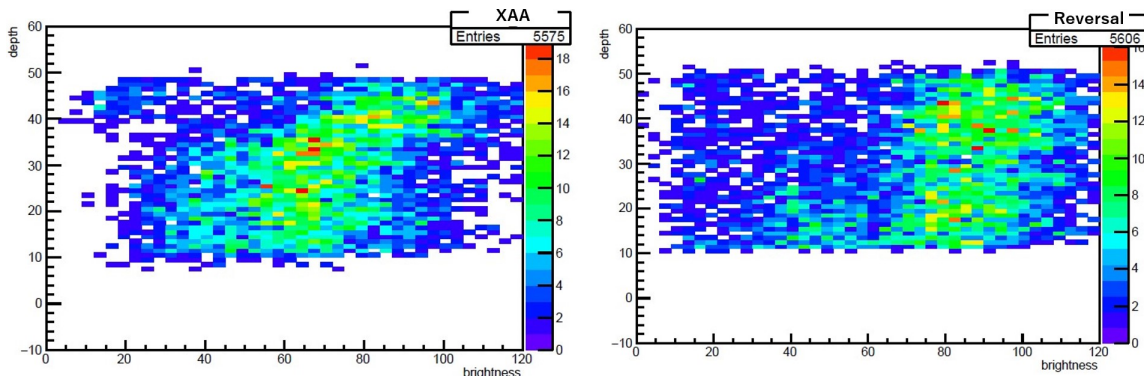


Figure 5: Relationship between max brightness and depth of grain. Color map is the number of grain.

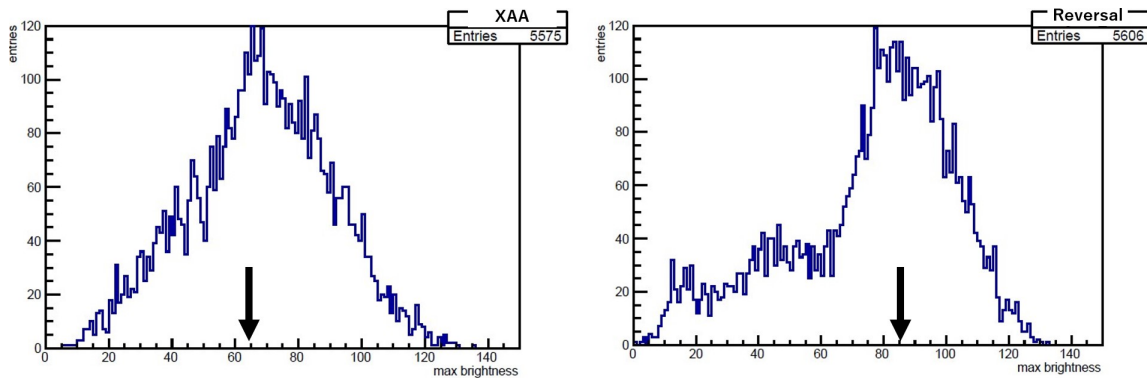


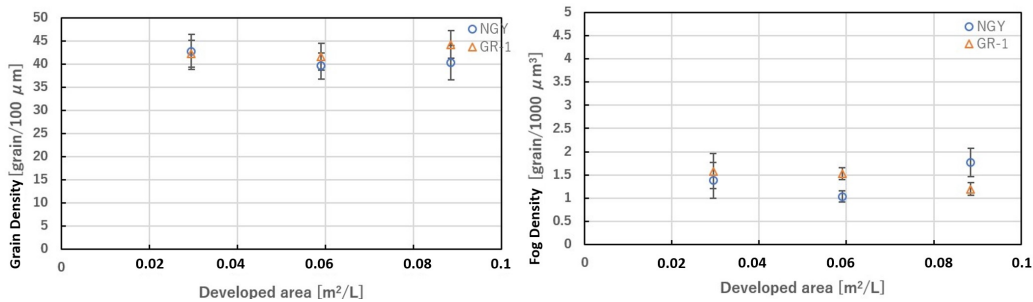
Figure 6: Histogram of max brightness in XAA developer and Reversal developer.

### 3. Development for large scale processing

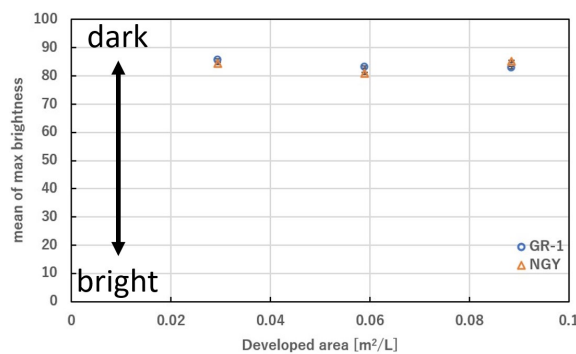
By changing the developer, it became necessary to prepare the developer from powder. However, the failure rate of liquid preparation in 2L is about 20%. Therefore, We decided to outsource the preparation of the developer. In order to reduce the amount of developer to store after purchase, we decided to make a developer concentrate. Precipitation of hydroquinone occurred by making developer the concentrated solution, but to store developer at above 5 °C by raising the pH became possible. We outsourced the raised pH Reversal developer and made to create the developer simply possible by diluting developer with ion-exchanged water. By simplifying the liquid preparation, we eliminated mistakes. Call the outsourced developer GR-1.



We evaluated the performance of the GR-1 in large-scale development. And we were assumed that the emulsion film of maximum area of up to 100 m<sup>2</sup> was developed with 390 L developer, we evaluated developer with 0.5 L developer. We developed 120 m<sup>2</sup> emulsion films three times, and we also developed about 30 m<sup>2</sup> emulsion films for evaluate sensitivity and amount of noise too same times. The developer result are shown in Figure. 7, Figure. 8 and Figure. 9. These figures are compared handmaid developer at laboratory (NGY, plot of blue circle) and outsourcing developer (GR-1, plot of orange triangle). Figure. 7 is grain density against developed area. Grain density is equivalent to sensitivity of emulsion film, and it is the number of grains in 100 μm of an electron track in the emulsion layer. The acceptable range for grain density is above 35 at GRAINE experiment. Figure. 8 is fog density against developed area. Fog density is equivalent to amount of noise of emulsion film, and it is the number of noise grains in 10 μm<sup>3</sup> in emulsion layer, grain of not track of charged particle is noise and call fog. The acceptable range for fog density is under 4 at GRAINE experiment. Looking at the graphs of sensitivity and amount of noise of emulsion film, both satisfied the acceptable range even if the developed area increased. Figure. 9 is mean value of max brightness against developed area. This mean value of max brightness is the mean value when fitting the max brightness form 60 to 120 with a gauss function. It is known that the mean value of max brightness is decrease when developer is exhaust, figure. 9 showed that 0.09 m<sup>2</sup>/L development in GR-1 was not exhaust. Therefore, even if large scale develop of 0.09 m<sup>2</sup>/L with GR-1 developer, there shows no problem with sensitivity, amount of noise and exhaust for GR-1 developer.



**Figure 7:** Grain density against developed area. **Figure 8:** Fog density against developed area.



**Figure 9:** Mean value of max brightness against developed area. This mean value of max brightness is the mean value when fitting the max brightness form 60 to 120 with a gauss function.

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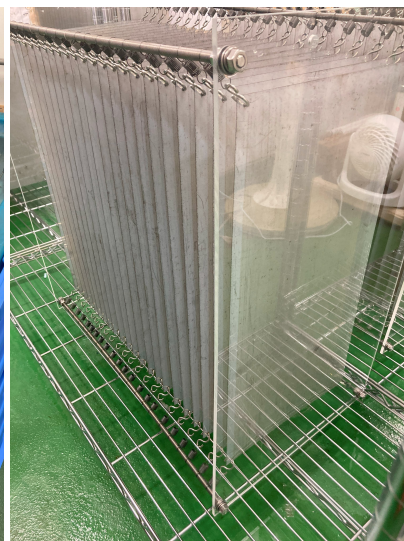
#### 4. Development for balloon flight experiment film of GRAINE2023

After the experiment, emulsion films were returned to Japan and developed at Gifu University. We received 10 liters  $\times$  135 boxes developer. The one box is 10 liters. We prepared 390 liters developer in 400 liters tank (Figure. 10), and developed 33.75 m<sup>2</sup> emulsion films with 390 liter developers. The all developed area of emulsion film is above 670 m<sup>2</sup>. We completed development the balloon experiment films in July 25, 2023.

Figure. 12 is photomicrograph of emulsion film of balloon experiments at GRAINE experiment in 2018 and GRAINE experiment in 2023. The left picture is balloon experiment flight film in 2018, and the right picture is balloon experiment flight film in 2023. Grains are more clearly visible in GRAINE experiment film in 2023 than in GRAINE experiment film in 2018. Therefore, we succeeded in high-contrast development of the GRAINE experiment film.



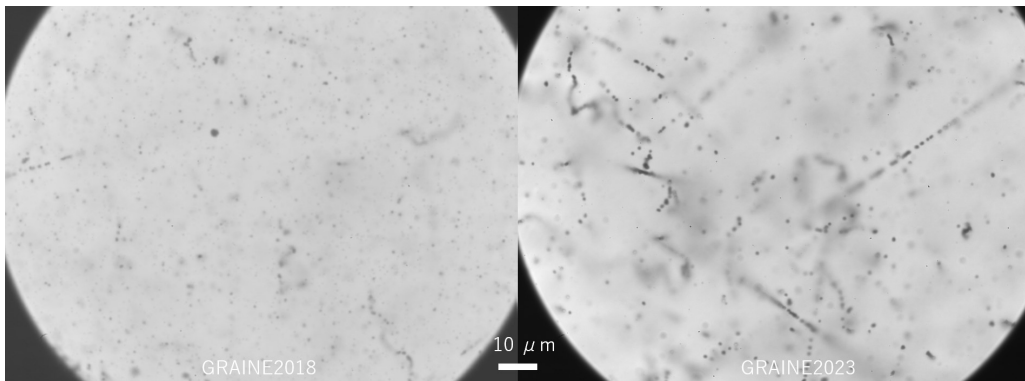
**Figure 10:** The 400 liter tank for de-  
veloper. 390 liter developer is inside  
this tank.



**Figure 11:** The one rack can hang  
23 films. The size of one tank (Fig-  
ure. 10) can develop 2 racks at once,  
and developed 4 racks in one day.  
These films were dry in this time.

#### 5. summary and discussion

The purpose of the GRAINE experiment is to analyze the gamma-ray imaging of high energy astronomical objects of 10 MeV to 100 GeV band in high angular resolution with nuclear emulsion film. The nuclear emulsion film is a track detector type of the photographic film coated nuclear emulsion gel, which is mixed AgBr in gelatin, on a plastic sheet base or on a glass base. We can get the charged particle track data of emulsion film by Track Selector after development. The scanning speed of Track Selector has improved with each upgrade, and the HTS2, the fastest Track Selector now, was developed for improvement of the scanning speed, by lowering the magnification and widening the field of view of the captured image. To compensate for the deterioration of



**Figure 12:** The microscope image of balloon flight film. Left picture is balloon experiment flight film in 2018 (GRAINE2018), and right picture is balloon experiment flight film in 2023 (GRAINE2023).

the detection performance of developed silver by lowering the magnification, we developed high-contrast developer for the nuclear emulsion film by enlarging the developed silver, and we succeed high-contrast development by changing the developer to reversal developer (GR-1) from XAA.

In Figure. 5, When XAA developer is deep, the max brightness value decreases, but the reversal developer is constant. With the XAA developer, the max brightness value was about 20 lower than reversal developer except for the  $10\ \mu\text{m}$  surface. The change in the max brightness value is caused by the size of the grain size, and the change in the grain size is caused by the development speed. The development of the surface progresses easily, because emulsion film takes time for the developer to penetrate deep into the emulsion. The reversal developer could develop the surface and the deep portion to the same degree. We outsourced the developer with chemical composition of reversal developer. In Figure. 7, Figure. 8 and Figure. 9, we researched performance of GR-1 developer, it is no problem with sensitivity, amount of noise and exhaust. These results allowed large scale development to begin. There is no problem in the GRAINE experiment in 2023, but if amount of development area exceeds  $0.09\ \text{m}^2/\text{L}$ , further consideration is required.

In the April 2023, we launched the emulsion chamber telescope to Longreach from Alice Springs, Australia. We completed development of GRAINE2023 films using GR-1. In Figure. 12, Grains are more clearly visible in GRAINE experiment film in 2023 than in GRAINE experiment film in 2018. We succeeded in high-contrast development of the GRAINE experiment film. The next step is a more detailed evaluation, like Figure. 5 and Figure. 6. In addition, track data will be acquired with HTS2 and data analysis will be performed.

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