

Study of the hygroscopic deterioration of ammonium perchlorate/magnesium mixture

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Abstract

A Study of hygroscopic deterioration is necessary because it can improve the safety and quality of pyrotechnics. For effective prevention of hygroscopic deterioration, this paper shows the deterioration mechanism of ammonium perchlorate (AP)/magnesium (Mg) mixture under the presence of moisture. The mixture has been used as Tenmetsuzai (Strobe composition in Japanese) and Mg has been known for its high hygroscopy.

In this paper, the hygroscopic deterioration of strobe composition (AP/Mg mixture) was measured by aging test which we were developed, and the deterioration products were analyzed with X-ray diffraction system and scanning electron microscope. The experiments identified two deterioration products: magnesium hydroxide and magnesium perchlorate. The influence of storage temperature and humidity to the rate of deterioration were investigated, and it revealed that the relative humidity of atmosphere has a significant impact. AP/Mg showed drastic change at a point, and the point was considered to be Critical Relative Humidity. The rate equation which called Jander's equation was applied to the deterioration behavior obtained in the experiments, and its coefficients were obtained which enable to predict the degree of deterioration as function of time.

Keywords: pyrotechnics, ammonium perchlorate, magnesium, aging, humidity

1. Introduction

Pyrotechnics needs careful treatment because there have been fatal explosions caused by pyrotechnics¹⁾. There have been a lot of study on the deterioration of explosive, and some of them focused humidity as a cause. Many humid areas inherently have been suffered from the influence of water molecule on materials, and researchers have investigated the deterioration products, change in sensitivity, heat generated in spontaneous combustion, combustion velocity, and so on. The target explosives were nitrocellulose (used for gunpowder)²⁾⁻⁴⁾, black powder (used for pyrotechnics)⁵⁾⁻⁷⁾, explosive mixtures containing metal (used for fireworks)⁸⁾⁻¹⁶⁾, and other types of explosive mixtures (used for propellant)¹⁷⁾⁻¹⁹⁾.

The behavior of pyrotechnics in moisture have been also a subject of research in this field. A thermal analysis

showed the effect of anti-deterioration agent for pyrotechnics composition by using differential scanning calorimetry¹¹⁾. Boric acid and potassium dichromate were effective for most of the pyrotechnics composition investigated in this study. However, they were ineffective to some pyrotechnics composition and caused reactions. Another study showed minor impurities in water influenced on the reaction behavior of antimony sulfide and Mg. Additionally, the reaction between antimony sulfide/Mg mixture and water exhibited induction period¹³⁾.

Pyrotechnics which contain Mg easily react to water, but Mg were often used for pyrotechnics because of their high performance. For example, Ammonium perchlorate (AP)/magnesium (Mg) mixture is one of the strobe compositions. The mixture is considered to burn in two

types of reactions occurred simultaneously or alternatively, and each reaction emits lights in different luminance. The phenomenon creates strobe effect^{20,21}. AP/Mg mixture exhibits strobe combustion which adds to beautiful effect to the pyrotechnics performance, but it is highly hygroscopic. Therefore the mixture poses a risk of combustion performance change such as abnormal combustion. In previous researches, reactions between AP/Mg mixture in water were investigated^{10,12}. The studies suggested the reaction mechanism and elucidated the effect of potassium dichromate as an anti-deterioration reagent. In those studies, the ion of magnesium and perchlorate were found in liquid phase and magnesium hydroxide in residue.

For prevention of the hygroscopic deterioration of AP/Mg mixture, the reaction mechanism between solid mixture and water vapor needs to be clarified. This paper aims to reveal the mechanism of the hygroscopic deterioration of AP/Mg mixture and the influential factors such as storage temperature and shape of mixture. In contrast to previous studies focusing the reaction in water, the study on the influence of humidity was scarce although the knowledge of the behavior under humidity is more practical than in bulk of water. The investigation leads pyrotechnics to increasing safety and improving quality through their lifecycle, and the result should contribute to the safety of pyrotechnics.

In order to investigate deterioration mechanism in humid environment, aging test in controlled environments was conducted and the products were analyzed with powder X-ray diffraction (XRD). The deterioration progress was tracked by monitoring weight change in the function of temperature and humidity as an analogous to chemical reaction which is governed by temperature and the concentration of reactants. In the analysis, the relative humidity is paid attention as well as absolute humidity. Furthermore, the effect of physical contacts between metal and oxidizing agent to the deterioration rate was investigated with regard to moisture absorption. The effect of density was measured by weight variation as well. The deterioration rate was analyzed and assumed that the diffusion of water molecules in deterioration products on the Mg powder is a major factor, and an equation was obtained to predict the degree of deterioration as function of time.

2. Experimental

2.1 Materials and samples

AP (Wako Special Grade: Wako Pure Chemical Industries, Ltd.) and Mg (Mg-100: Kanto Metal Corporation) and magnesium hydroxide, Mg(OH)₂ (99.9%: Wako Pure Chemical Industries, Ltd.) were used without further purification. Three types of sample mixture were prepared since this study focused on the effects of atmosphere (Sample A), contacts between metal particles and oxidizing agent particles (Sample B, D, E), and sample density (Sample C). The sample A and C were composed of AP and Mg with the weight ratio of 60:25, the ratio of which was determined based on the one of mixture which

Table 1 Sample states and storage conditions.

Sample name	Relative humidity [%RH]	Absolute humidity (Vaper pressure) [g m ⁻³] (kPa)	Temperature [°C]	AP [wt.%]	Mg [wt.%]
A1	74.5	61.6 (9.19)	50	70.6	29.4
A2	74.7	38.1 (5.51)	40	70.6	29.4
A3	75.1	22.7 (3.18)	30	70.6	29.4
A4	50.9	43.1 (6.45)	50	70.6	29.4
A5	53.2	28.6 (3.92)	40	70.6	29.4
A6	56.0	17.0 (2.38)	30	70.6	29.4
A7	31.6	16.1 (2.33)	40	70.6	29.4
B	74.7	38.1 (5.51)	40	50.0	50.0
C	74.7	38.1 (5.51)	40	70.6	29.4
D1	74.4	96.4 (14.8)	60	100	–
D2	74.7	38.1 (5.51)	40	100	–
E	74.7	38.1 (5.51)	40	–	100

emits white strobe light²². Equivalent mixture of AP and Mg was named as sample B. Sample C was pelletized AP/Mg mixture with a hand-press. Its density was 1.0–1.6 g cm⁻³ which is similar to those of stars in fireworks (1.6 g cm⁻³)²³.

Aging test was carried out in the plastic container which is in an incubator under the constant temperature and the constant humidity kept with the saturated salt method²⁴. Saturated aqueous solutions keeps equilibrium with moisture in atmosphere at a certain vapor pressure. Half gram of powdery sample A, B, D, and E and pelletized sample C were placed on each hexagonal balance tray. The trays were placed in a sealed plastic container in which the humidity was controlled with saturated salt. The salts generate their characteristic humidity depending on temperature. In this study, humidity was kept at about 75%RH (Sodium chloride), 55%RH (Sodium bromide) and 30%RH (Magnesium Chloride), and temperature was kept at 30 °C, 40 °C, 50 °C and 60 °C. Sample A which stored under about 75%RH of humidity was named A1–A3. Sample A4–A6 was kept under about 55%RH of humidity. Sample A7 was stored under about 30%RH. As the figure increases, the ambient temperature was low. Sample D1 was stored in the same condition as that of sample D2, apart from temperature. All sample mixtures and the storage conditions are shown in Table 1.

2.2 Deterioration products analysis

The deterioration product was investigated with Powder XRD (RINT-2000: Rigaku) which used CuK α 1 radiation (1.5405 Å). The optics were a fixed divergence slit (1°), a fixed scatter slit (1°), and a fixed receiving slit (0.3mm). The X-ray tube worked at 40 kV and 200 mA. The components in the products were identified with integrated X-ray powder diffraction software (PDXL). The deterioration surface was studied by the images obtained with Scanning Electron Microscope (VE-8800: keyence). The accelerating voltage was 2 kV.

2.3 Measurement of hygroscopic deterioration behavior

In this study, the progress of hygroscopic deterioration

was expressed with weight variation. Each sample was weighed regularly every 24 hours for 6 days with an electronic balancer (PA64JP : Ohaus).

The weight variation rate, ΔW was defined as follows :

$$\Delta W = (w_t/w_0 - 1) \times 100 \quad (1)$$

Where w_t is the sample weight after t hours storage, and w_0 is the initial weight of the sample.

The samples stored for 1, 2, 3, and 6 days were taken out of the container and dried to remove water to separate the reversible water absorption from the irreversible reaction caused by humidity. In the drying process, the sample was heated to 100 °C and maintained for 30 minutes. The specific weight variation ΔW_r which expresses irreversible process by humidity was calculated by the following equation :

$$\Delta W_r = ((\Delta W + 100) \times w_{\text{after drying}}/w_{\text{before drying}}) - 100 \quad (2)$$

Where $w_{\text{after drying}}$ is the sample weight after drying, and $w_{\text{before drying}}$ is the sample weight measured immediately after it was taken out of the container. The ΔW_r also can be expressed with the following equation by incorporating the Equation (2).

$$\Delta W_r = (w_{\text{after drying}}/w_0 - 1) \times 100 \quad (3)$$

However, the Equation (2) was used for the data collection to remove the experimental errors due to the replacement of a hexagonal plastic tray with a heat-proof dish for drying.

For sample C, the change of sample volume was measured with a vernier micrometer. The density was calculated at each measuring point with the change in volume and weight during the storage.

3. Results and discussion

3.1 Deterioration products analysis

The powder XRD data of the deterioration products were obtained from Sample A2, A5 and D1 (Figure 1). A2 was chosen for the analysis because it represented the deterioration in summer in Japan. A5 was analyzed as a reference because it was kept under the same temperature as A2. For the behavior of Mg, D1 was analyzed as well. The result shows some change in the m from the initial state. Peaks of $\text{Mg}(\text{OH})_2$ were found in the diffraction of sample A2 (75%RH, 40°C) after the three, six, and twenty-four days of storage. On the other hand, the peak of magnesium perchlorate, $\text{Mg}(\text{ClO}_4)_2$ was identified only in the sample stored for 24 days. In the result of A5 (55%RH, 40°C), only magnesium hydroxide was identified and its peak intensities were weak. The spectrum of D1 showed the presence of $\text{Mg}(\text{OH})_2$ which was the deterioration product of Mg. The SEM image (Figure 2) of sample B after 9 days storage shows needle-like substance as the deterioration products which was collected on the Mg surface.

In bulk of water, the reaction of AP and Mg was explained with the following ionic formulas in the previous study¹⁰,

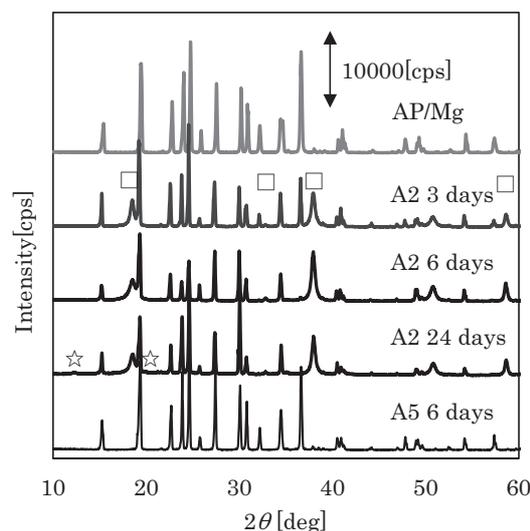


Figure 1 XRD peak of aged AP / Mg mixture by hygroscopic deterioration.

[□ : Magnesium hydroxide, ☆ : Magnesium perchlorate]

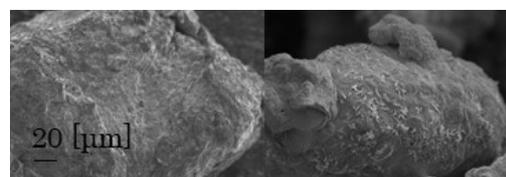
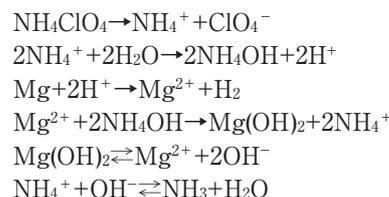


Figure 2 Picture of SEM.

(Left: Mg reagents, Right: Stored sample B at 9 days)



The hygroscopic deterioration were supposed to occur in the similar manner to the reaction in water since the same non-volatile species ($\text{Mg}(\text{OH})_2$ and $\text{Mg}(\text{ClO}_4)_2$) were observed in hygroscopic deterioration products. However, the reaction in hygroscopic deterioration is not completely the same as the reaction in water. As for the reaction in water, magnesium perchlorate and magnesium hydroxide were observed in the dried residue¹⁰. Meanwhile, the XRD analysis revealed the appearance of magnesium perchlorate lagged behind magnesium hydroxide in the reaction in moisture. Since the peak intensity of $\text{Mg}(\text{ClO}_4)_2$ is almost the same as the one of $\text{Mg}(\text{OH})_2$, the XRD result means $\text{Mg}(\text{OH})_2$ generated faster than $\text{Mg}(\text{ClO}_4)_2$. The phenomenon is considered to be attributed to the difference in the extent of contact among Mg^{2+} , H_2O and ClO_4^- . In the sample mixture under humid condition, water molecules enter into small space created among particles and interact with Mg or AP. The ingress of water leads in the generation of $\text{Mg}(\text{OH})_2$. Also, the water can dissolve AP and serve as reaction field with Mg. However, the contact of AP and Mg shares less area than the area Mg exposes to the water molecule. Therefore, the gas phase - solid phase contact (water - Mg) reaction advances the solid - solid phase contact (AP - Mg) reaction.

3.2 Measurement of hygroscopic deterioration behavior

Weight variation was obtained by aging test (Figure 3–6). In order to grasp the overall picture of weight variation caused by water, samples kept in conditioned atmosphere was weighed regularly (Figure 3). Factors influencing on the behaviors was examined: interaction between oxidants and fuel (Figure 4) and initial density (Figure 5).

Figure 3 clearly shows that the storage atmosphere affected weight variation. The weight variation indicated hygroscopic deterioration. In this experiments, Sample A1 exhibited the largest weight variation, and the extent of weight variation decreased in the order of A2, A3, A4 and A5. In most cases, the reaction rate is increased when the ambient temperature and the concentration of the reactant are high. In this case, high humidity means atmosphere contains high concentration of reactant which is water. A1 and A4 were stored at high absolute humidity, and they would show large weight variation. But A4 showed little weight variation.

The order of the deterioration degree accorded with the order of neither the ambient temperature, nor the concentration of water vapor in the atmosphere: the highest concentration of water vapor was the atmosphere storing A1 (61.6 g m^{-3}), followed in order by A4 (43.1 g m^{-3}), A2 (38.1 g m^{-3}), A3 (22.7 g m^{-3}), and A5 (28.6 g m^{-3}). This result showed that the weight variation did not altogether depend on absolute humidity, which is the concentration of water vapor in air. For example, the temperature and the absolute humidity in air for A3 was lower than those for A4, while the weight variation of A3 was greater than A4.

Figure 4 presents the difference of weight variation led by contacts between Mg and AP. The storage atmosphere of sample B, D2 and E were the same but they showed the different weight variation behavior. As sample B is the equivalent mixture of D2 and E, its weight variation should have been plotted in the middle between D2 and E. However the increase from the initial weight of B reached to about 50% while D2 and E remained at around 1%. It was suggested that the hygroscopic behavior of B was not simply the sum of each component and the result of the interaction between fuel metal and oxidizing agent. Since the interaction promotes hygroscopic deterioration, the components should be separated in storage to avoid hygroscopic deterioration.

Figure 5 shows the significant effect of initial density in hygroscopic deterioration. As the initial density raised, the weight variation after six days storage decreased. The feature can be attributed to the empty spaces between particles and the diffusion of water molecules into the voids because the sample porosity is proportioned to density. The ideal density of the sample, 60 : 25 mixture, is 1.89 g cm^{-3} in this investigation because the density of AP is 1.95 g cm^{-3} and Mg is 1.74 g cm^{-3} . The densities and porosities of samples ranged from 1.2 g cm^{-3} and 40% to 1.6 g cm^{-3} and 20%, respectively. As the porosity increased, the weight variation increased. Thus, this result showed the porosity is important factor for hygroscopic

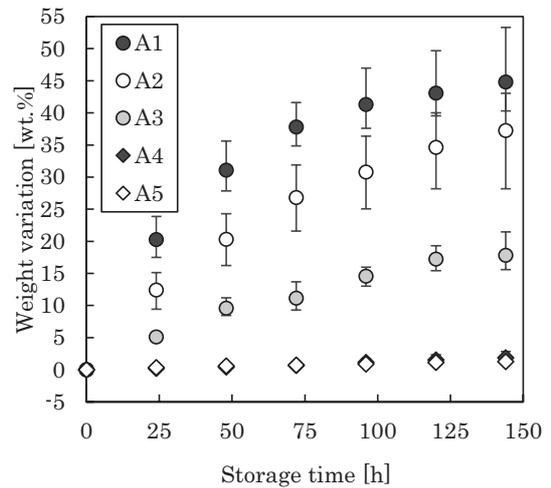


Figure 3 Weight variation by aging test. (Sample A1~A5).

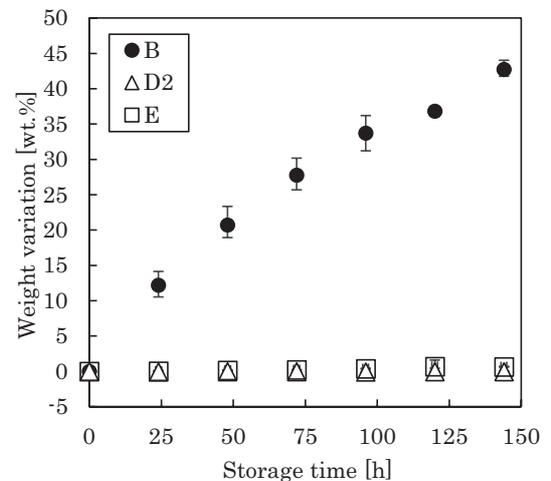


Figure 4 Weight variation by aging test. (Sample B, D, E) [75%RH, 40 °C].

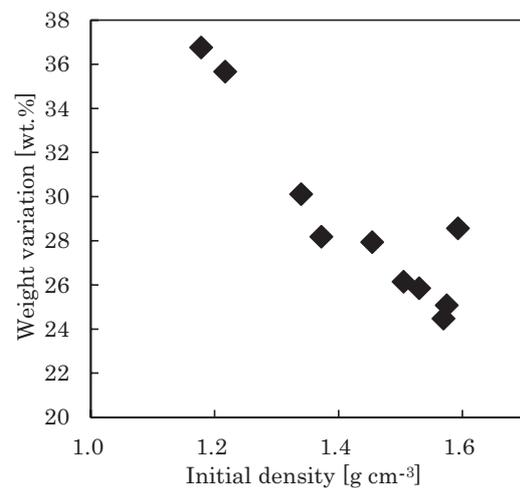


Figure 5 Effect of initial density. (Sample C) [75%RH, 40 °C, 6 days].

deterioration.

Figure 6 indicates the amount of water removed from the stored sample and presents that the weight variation can be separated into moisture absorption and reaction by the drying process. All sample contained water less than 10%.

A reaction rate equation was applied to the rate of

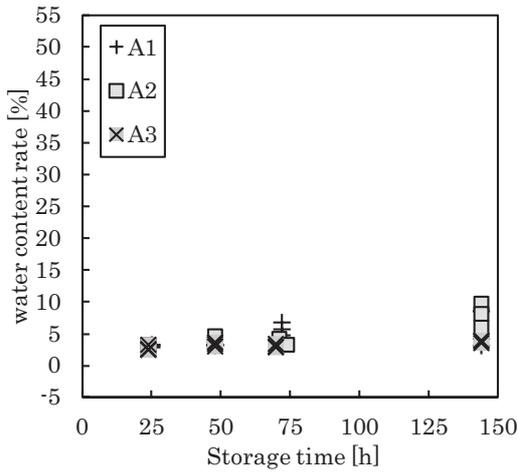


Figure 6 Water content of A1~A3 in 6 days.

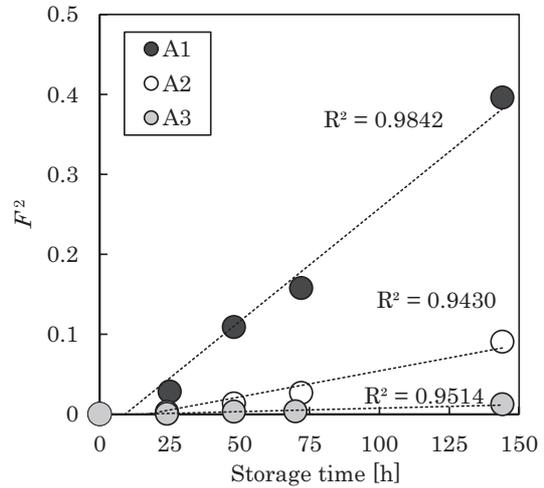


Figure 7 Applied Jander's equation.

deterioration. Based on figure 6, moisture absorption as reversible weight change least contributes to the overall weight variation and the influence of moisture absorption can be negligible. Therefore, the rate of diffusion should be directly applied to the rate of reaction. Some equations have been proposed which relate a rate of diffusion, and Jander's equation has been mostly adopted for powder^{25), 26)}. In Jander's equation, particles of sample are assumed as sphere, and the reaction rate can be expressed with the following equation.

$$F^2 = (1 - (1 - \alpha)^{1/3})^2 = k_1 t \quad (4)$$

Where α is the rate of reaction, k_1 is the reaction rate constant. The time of reaction t is proportional to F^2 when the rate determination step is controlled by diffusion process. In this study, the rate of reaction was calculated according to the Equation (4).

$$\alpha = \Delta W / ((M_{Mg(OH)_2} / M_{Mg} - 1) \times \text{Wt. \%}_{Mg_0}) \quad (5)$$

Where M is molecular weight $g \text{ mol}^{-1}$, Wt. \%_{Mg_0} is initial rate of Mg weight. Figure 7 is Jander's equation plot, and it showed that the weight variation behaviors were good fit to Jander's model. The slope on Figure 7 lead to k reaction rate constant. Figure 8 shows the Arrhenius equation plot and its can be formed Equation (6).

$$k = \exp(-1.7 \times 10^4 / T + 38) \quad (6)$$

Where T is the temperature.

By modifying the Equation (4) and (5), weight variation can be predicted as following equation :

$$\Delta W = (M_{Mg(OH)_2} / M_{Mg} \times \text{Wt. \%}_{Mg_0} - 100) (1 - (1 - (kt)^{0.5})^3) \quad (7)$$

3.3 Critical relative humidity of AP / Mg deterioration

The experimental results of AP/Mg mixture shows the parameter which was effective to the deterioration of the mixtures is relative humidity. Relative humidity is the ratio of the water vapor pressure to the equilibrium vapor pressure at a temperature. Figure 9 plotted the weight variation after 6 days storage as the function of absolute humidity (left) and relative humidity (right) and the left figure shows that weight variation seems irrespective of

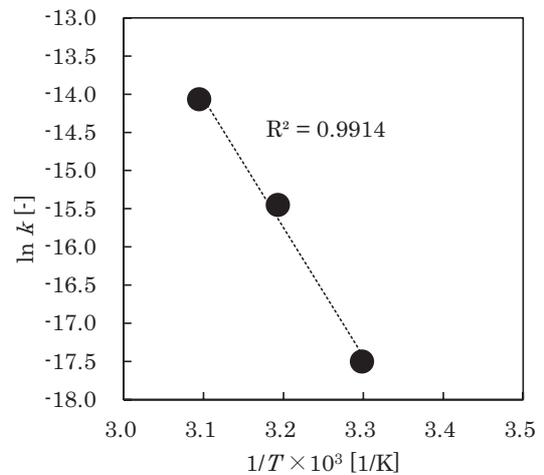


Figure 8 Arrhenius equation.

absolute humidity. From the viewpoint of relative humidity, the samples which were kept at 75%RH showed the temperature dependence of weight variation while the samples stored at 55%RH represented little influence of temperature. Contrary to the region of low humidity (55% RH), temperature played an important role under high humidity condition (75%RH).

In other words, samples at low relative humid region hardly increase weight while samples absorbed water at 75%RH and the increment depends on the ambient temperature. The gap between the behaviors of high and low relative humidity can be due to the increase of the absorption site. During the storage, sample gradually lost smoothness as samples absorbed water from the environment. Needle-like substance was found from the surface and XRD indicated it was the product of deterioration, $Mg(OH)_2$. The generation of $Mg(OH)_2$ expands the surface, which means the increase of moisture absorption sites. However, its contribution to the weight variation behavior should be less significant because the weight variation of moisture absorption is lower than the weight variation of reaction (Figure 6) As far as the weight variation is originated to hygroscopic deterioration, the surface expansion was less influential on the weight variation behavior.

Another possible explanation is the critical relative

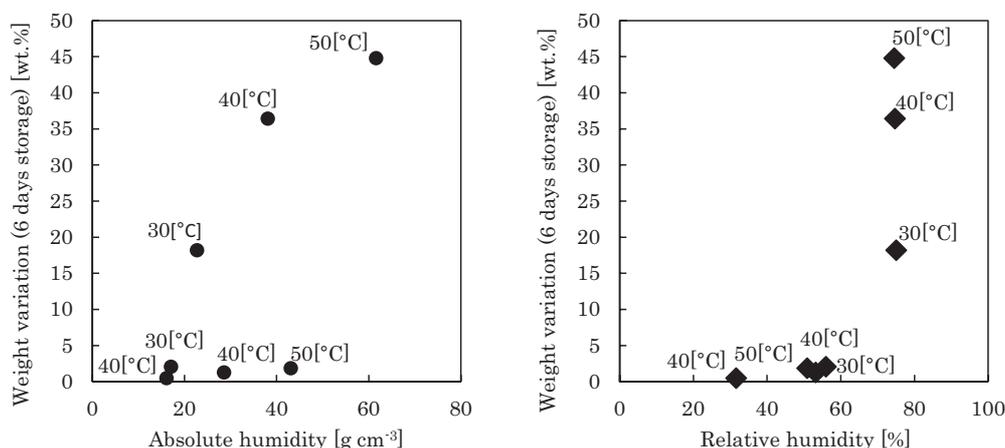


Figure 9 Effect of humidity parameter. (Sample A after 6 days storage)

humidity (CRH)^{(22), (27), (28)}. CRH is characteristic to a substance and represents a specific relative humidity at which the substance starts to absorb moisture in the air. At a humidity under CRH, thin saturated solution of a substance on the solid substance. The water vapor pressure is at equilibrium with the saturated solution. When the ambient humidity exceed CRH, the solid substance starts to dissolve into the solution and the overall substance absorbs water to balance the equilibrium between water vapor pressure and the solution on the solid substance. As CRH is obtained for inorganic salts; CRH of AP is from about 90 to 96%RH (at 30 °C and 40 °C)⁽²⁹⁾. In the mixture of this study, CRH was at some point between 55%RH and 75%RH, and the mixture shows decrease of CRH. The CRH depends on the highest hygroscopic substances in mixture, and in this case, magnesium perchlorate decreased CRH of the mixture. The effect of CRH is significant and the deterioration behavior exhibits drastic change between before and after CRH. Since the treatment without care for CRH will lead to acceleration in the deterioration of the AP/Mg pyrotechnics composition.

4. Conclusion

For effective prevention of hygroscopic deterioration, the mechanism of the hygroscopic deterioration of AP/Mg was studied. The deterioration products were identified as magnesium hydroxide and magnesium perchlorate by XRD. The products were similar to ones generated in the reaction in water, but the generation rate of products was slower in moisture. The most of deterioration products after 6 days storage was magnesium hydroxide. The understanding of the deterioration mechanism can contribute to the improvement of deterioration prevention measure.

The aging test showed that controlling atmosphere without attention to the relative humidity can lead to accelerating deterioration because the relative humidity have a more impact on deterioration than the temperature and absolute humidity. The mixture changed hygroscopic behavior between 55%RH and 75%RH and CRH was supposed to be somewhere in the region. When the temperature was risen over the CRH, the deterioration

rate of the mixture was accelerated. Thus CRH is important parameter in this mixture to consider the deterioration behavior.

Other parameters which effected to the hygroscopic deterioration were porosity and interaction between particles. The initial density of the mixtures influenced on the rate of deterioration and it related to porosity. Mixing AP and Mg which brought interaction between oxidizer and metal advanced its hygroscopic deterioration. The result contribute to deciding the form of the pyrotechnics composition and storage condition.

Jander's equation was applied to the deterioration phenomenon. The result provided the method to predict deterioration in the AP/Mg mixture. In order to decide the storage period, the prediction of the hygroscopic deterioration is necessary. This study proposed the equation based on the deterioration rate. T equation was validated only for AP/Mg deterioration, but the proposal of the deterioration prediction contribute to the safety management for the pyrotechnics composition storage.

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