Morphology Changes and Fillers Migration in Urethane Composites during Thermal Degradation

Atsuki Tanifuji[1], Takato Ishida[2] and Ryoma Kitagaki[3]

[1] Graduate School of Engineering, Division of Human Environmental Systems, Hokkaido University, Kita 13-Nishi 8, Kita-ku, Sapporo, Hokkaido 060-8628, Japan, atsuki_123@eis.hokudai.ac.jp

[2] Graduate School of Engineering, Division of Human Environmental Systems, Hokkaido University, Kita 13-Nishi 8, Kita-ku, Sapporo, Hokkaido 060-8628, Japan, takato.matphysichem@gmail.com

[3] Graduate School of Engineering, Division of Human Environmental Systems, Hokkaido University, Kita 13-Nishi 8, Kita-ku, Sapporo, Hokkaido 060-8628, Japan, ryoma@eng.hokudai.ac.jp

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1 Introduction

Building material products used for topcoat of constructions contain so many chemical materials and consist of chemically complex systems. However, there are still many unclear points about how the coating material containing many kinds of polymers and inorganic substances to protect the surface layer. Therefore, this paper examines thermal degradation mechanism of coatings based on the result of the chemical analysis of the coating surface by XPS and elemental mappings by SEM-EDS.

2 Methods

In this study, both thermally-degraded and non thermally-degraded samples were prepared. A conventional two-component type urethane coating including TiO_2, $CaCO_3$, and $BaSO_4$ as fillers was applied on a slide glasses with a spin coater to control film thickness. After the surfaces of the samples were sufficiently dried, they were cured for 120 days at 20°C and 60% relative humidity. Subsequently, the samples after cutting were heat-degraded at 30°C, 50°C, and 70°C. The periods of the degradation are 0, 1, 3, and 7 days, and XPS measurement and SEM-EDS measurement were conducted respectively (L.S De Bernarde *et. al.* (1984)). The features of the measurements are briefly shown in Table 1.

Table 1. Survey contents in each method.

XPS	・History of elemental type and chemical bonding state of coating surface layer	
SEM-EDS	SEM image	・Cross section thickness of coating ・Filler particle size
	Elemental mapping	・Distribution of elements in the cross section of coating film and its history

3 Results and Discussion

Figure 1 shows XPS narrow scan spectra of the urethane resin coatings that were thermally degraded at 70°C for 0, 1, 3, and 7 days. The peak of O 1s, N 1s, and Si-O increased as thermal degradation treatment continued, but the Ca peak was hardly detected on the surface layer. This result could be explained that the polymer such as urethane and siloxane was decomposed and appeared on the surface layer of the coatings due to low molecular weight and low density. Figure 2 shows that Ti in the coating, which was homogeneous in the initial state, and the

position sinks to the lower layer on the first day and floats to the surface layer with time by heating at 50°C. On the first day, fillers move to the bottom of the coating as polymers become smaller and less dense due to thermal decomposition. From the third day, the filler could have been pushed up from the bottom due to polymer cross-linking and polymerization reaction (Petrović *et. al.* (2000), and Yasir Nawab, *et. al.* (2017)).

These results indicate that the substance in the coating may move up and down dramatically before the surface of the coating is cracked while the coating is thermally degraded.
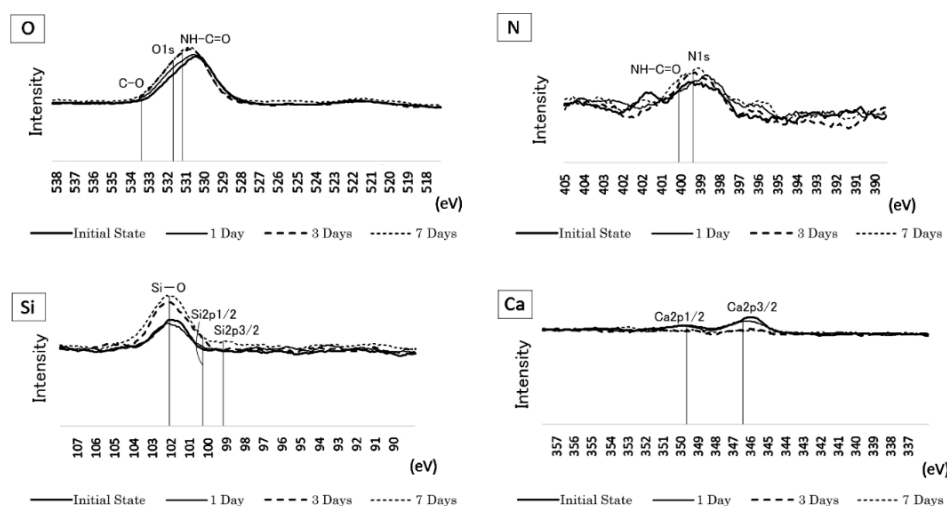

Figure 1. Narrow scan spectra of the urethane resin coatings that were thermally degraded at 70°C.
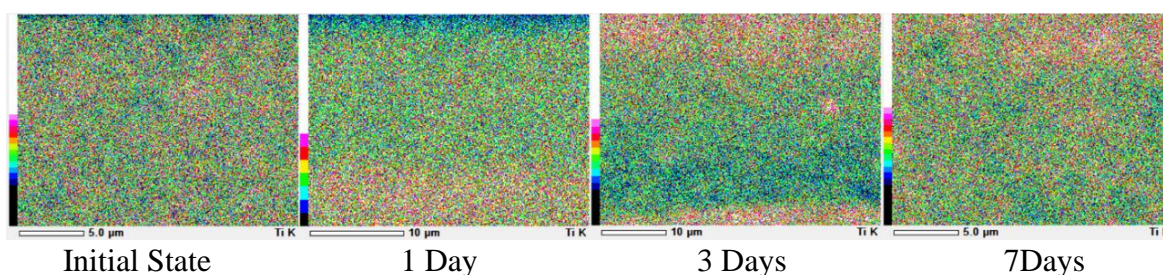

| Initial State | 1 Day | 3 Days | 7Days |

Figure 2. Sectional elemental mapping of Ti in urethane composite coatings at 50°C.

ORCID

Atsuki Tanifuji: http://orcid.org/0000-0003-4363-5834
Takato Ishida: http://orcid.org/0000-0003-3919-2348
Ryoma Kitagaki: http://orcid.org/0000-0001-6001-3688

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