Pyrolysis Mechanism of Triazole Compounds By Molecular Orbital Calculation and Evolved Gas Analysis

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ABSTRACT

The gas generating agents have been industrially used for mold foamed plastics, air-bag system, propellant, and so on. Since demand performance is vary according to use, the development of gas generating agents are accompanied by many experiments. The risk and cost of experiment are concerned with increasing of experimental number. The predictive calculation of pyrolysis mechanism of gas-generating agent is expected reduce the risk and cost of experiment with decreasing of the experimental number.

In order to obtain a better understanding of the pyrolysis mechanism of 1,2,4-triazole-3-one (TO) as triazole compound, molecular orbital (MO) calculations and evolved gas analysis were carried out. The MO calculations were performed using the density functional method (B3LYP) at the 6-311++G(d,p) levels by Gaussian 03 and AM1 method by MOPAC2002. The geometrical structure of TO and its tautomers were examined theoretically. Two-center energies (2CE) for TO was calculated via energy partition analysis. The value of 2CE corresponded to the binding energy, and were used as a standard for ease of bond dissociation in the triazole ring. The minimum value of 2CE in TO was the N1-N2 bonds. To investigate the properties of TO when the bond length N1-N2 bond in the triazole ring is extended, a relaxed scan was carried out using optimized structure of TO as the initial structure. The length of the N-N bond was found to increase with that of C3-N4 bond. The results of MO calculations, initial pyrolysis mechanism of TO was predicted that TO occurs by a ring-opening reaction of the triazole ring and the gases which evolved from TO were HNCO and CH2N2 compounds.

On the other hands, identification of the gases evolved from TO were carried out with thermogravimetry-infrared spectroscopy (TG-IR) and thermogravimetry-mass spectrometry (TG-MS). From the experimental results, it was determined that the gases which evolved from TO were HNCO, N2, CO2, NH3, and N2O. Additionally, HNCO was the major evolved component during the thermal decomposition of TO.

Since MO calculation results were supported the experimental results of evolved gases analysis TO, it was expected that the prediction of pyrolysis mechanism of gas-generating agents.