



## The effect of ozone on polymer degradation in Polymer Core Composite Conductors

J. Middleton<sup>a</sup>, B. Burks<sup>a</sup>, T. Wells<sup>b</sup>, A.M. Setters<sup>c</sup>, I. Jasiuk<sup>c</sup>, P. Predecki<sup>a</sup>, J. Hoffman<sup>a</sup>, M. Kumosa<sup>a,\*</sup>

<sup>a</sup>Department of Mechanical and Materials Engineering, University of Denver, 2390 S. York St., Denver, CO 80208, USA

<sup>b</sup>Department of Chemistry and Biochemistry, University of Denver, 2190 E. Iliff Ave., Denver, CO 80208, USA

<sup>c</sup>Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, 1206 West Green Street, Urbana, IL 61801, USA

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### ABSTRACT

The matrix material of high voltage Polymer Core Composite Conductors used in the next generation power transmission is generally a moderately high temperature epoxy, which has the potential of experiencing extreme aging effects in-service, including severe ozone degradation. It is well known that polymers are susceptible to oxidation by ozone, due to chain scission and the breaking of crosslinks. It is also known that ozone is present around high voltage transmission systems and its concentration rapidly increases close to energized transmission lines. Therefore, the primary goals of this study were to investigate (1) the effects of aging in ozone at room temperature of a high temperature epoxy, which could be used in the polymer composite conductors and (2) the effect of ozone exposure on a hybrid polymer matrix composite rod, presently used in-service. It was found using nanoindentation that the ozone oxidation depth on the surface of neat resin specimens was limited to about 120  $\mu\text{m}$ . It was also observed that exposure to 1% ozone for up to three months at room temperature did not negatively affect the flexural performance of either the neat resin epoxy, or the composite. Comparatively small variations in the bending strength and deformations of the specimens were associated with an initial increase in crosslinking of the epoxy followed by a small increase in chain scission and bond breaking. These effects also demonstrated themselves in the type of the viscoelastic behavior of the polymer after aging. The scission of the polymer chains in the neat resin and rod specimens was further evidenced by FTIR spectroscopy, where the aged neat resin specimens showed an increase in absorption around the 3500  $\text{cm}^{-1}$  wavenumber associated with the formation of carbonyl groups and carboxylic acids.

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### 1. Introduction

Transmission line High Voltage (HV) Polymer Core Composite Conductors (PCCC) can experience excessive bending [1], static and dynamic mechanical stresses in-service [2–5], various electrical stresses, pollution, and degradation caused by High Temperature (HT) [2,3]. They can also be damaged by prolonged ozone exposure. In the case of unidirectional glass fiber and carbon fiber/polymer composites, Fig. 1, the matrix dominated properties are especially vulnerable to the effects of aging [6–9]. It is therefore important to study aging effects of a high quality HT epoxy, which can be potentially used as the matrix material in the load bearing composite core, to better understand the in-service behavior of the PCC conductors.

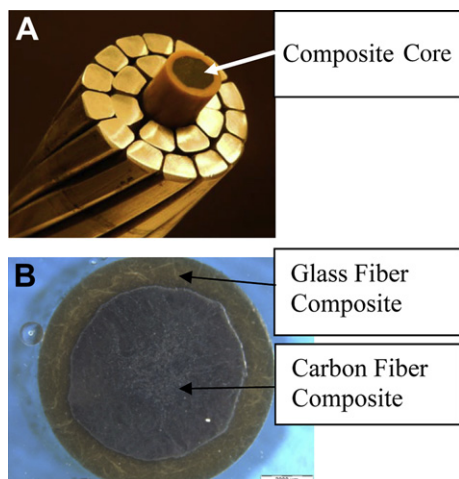
In the case of PCCCs there are two major sources of aging that are of primary concern. The first is thermal aging [2,10–12] and the

second one is exposure to ozone. Heating a polymer in air can degrade it due to thermo-oxidative effects, especially when aging near the glass transition temperature ( $T_g$ ) [2,10,11]. Both thermoset and thermoplastic polymers are susceptible to oxidation which breaks down polymer chains and crosslinks. This can damage the polymer, weakening its mechanical and electrical properties. Epoxies with high  $T_g$  are more resilient to this effect, albeit not much aging research has been done in this area. Oxidation occurs due to various sources, e.g. thermo-oxidation [13–15], UV oxidation [16], or ozone. Ozone is highly reactive and in the case of transmission lines, is constantly present due to the fact that ozone can be created from air and an electrical discharge, albeit dissociates at elevated temperatures (above  $\sim 100$  °C) into  $\text{O}_2$  and atomic oxygen [17].

Ozone has been investigated as a mechanism for polymer degradation [18–23], and as a functionalizer [24,25]. However, the degradation of epoxy due to ozone has received less attention [26]. It has been shown in [26] that for a Bisphenol A epoxy cured with methyl anhydride and 2-methyl-4-ethyl imidazole, degradation by

\* Corresponding author.

E-mail address: [mkumosa@du.edu](mailto:mkumosa@du.edu) (M. Kumosa).



**Fig. 1.** A Polymer Core Composite Conductor; (A) conductor design and (B) composite rod.

ozone at room temperature (RT) with the ozone concentration up to 100 ppm originates from the hydroxyl and methyl groups forming ozonide and carbonyl groups. It was also shown that after exposure to ozone for 500 h the pencil hardness went down from 2H to 1H. Moreover, it was found that by adding 50% of carbonyl iron by weight the degradation process slowed down significantly.

An interesting problem that arises from oxidation is the measurement of the oxidized layer that is produced on the surface of a polymer. A research team from Osaka: Narisawa et al. used Auger spectrum analysis to obtain elemental concentration as a function of depth in the material [27]. Several other groups used scanning tunneling microscopy (STM) to measure the depth of oxidation. Liao et al. used STM along with x-ray photoelectron spectroscopy (XPS) to study the topology of graphene and the oxygen signal [28], while Lee et al. used STM to study the depth of defects in graphite [29]. Lenkor et al. used infrared (IR) spectra to evaluate oxidation layers through absorbance of a common constituent found in each specimen [30], and Tandon et al. used dark-field photomicrographs to measure the oxidation layer [31]. For the current study the extent of oxidation was analyzed using nanoindentation by measuring nanohardness as a function of depth through the thickness of the sample. This data was then

related to the chemical changes determined by Fourier transform infrared (FTIR) spectroscopy and mechanical properties tested using three point bending.

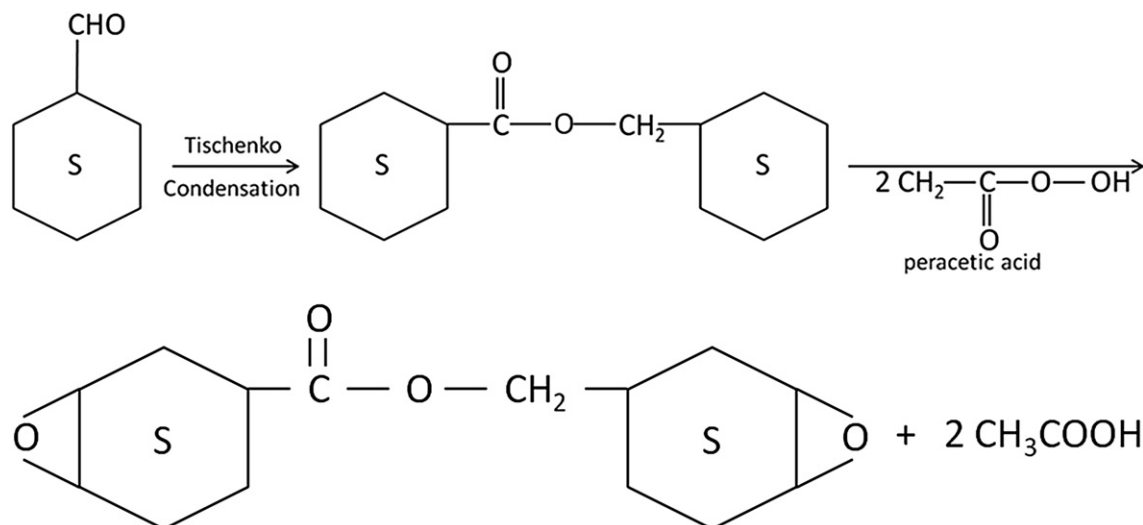
For structural applications in the presence of HV fields, cycloaliphatic epoxy resins are a common choice of matrix material. They have high resistance to ultraviolet degradation and arc-tracking due to their fully saturated molecular structures. This makes them an ideal candidate for applications with such environments [32]. They exhibit low viscosity and can have relatively high glass transition temperature. Such resins are expected to be used in PCC conductors since aging and electrical arc-tracking are major problems. Cycloaliphatic epoxy resins are prepared by converting tetrahydrobenzaldehyde by Tischenko condensation to form an olefin which is epoxidised with a peracid following the reactions shown below in Fig. 2.

Two commercially available cycloaliphatic epoxy resin monomer structures are presented in Fig. 3. They exhibit significantly different  $T_g$ , viscosity as well as mechanical and electrical properties. For outstanding electrical properties in outdoor environments, any of the cycloaliphatic epoxy resins (A and B) can be used if cured with fully saturated acid anhydrides [32]. However, if electrical performance is not a primary requirement but low viscosity coupled with high  $T_g$  is of paramount importance, then the epoxy structure shown in Fig. 3B would be the material of choice. This resin is cured at high temperature by nadic-methyl anhydride and its glass transition temperature can exceed 180 °C.

## 2. Experiment

### 2.1. Materials

The neat epoxy tested in this research was supplied by Lindau Chemicals Inc. (Lindau). According to Lindau, the epoxy is a cycloaliphatic epoxy resin and could have one of the preceding chemical structures shown in Fig. 3. This type of epoxy could be an outstanding candidate for the use in the PCCs. The chemical composition of the epoxy was: 3,4-epoxycyclohexylmethyl-3',4'-epoxycyclohexane-carboxylate, trade name Lindoxy-190 (190) which is the B type structure. The hardener specified as methyl-5-norbornene-2,3-dicarboxylic anhydride, or product name LS-252 (252). Using the cure cycle described below, Lindau reports  $T_g$  values of 222 °C.



**Fig. 2.** Cycloaliphatic epoxy resin reaction.

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