

# A mechanolectrical property-controllable smart paint and its application to the structural integrity monitoring of spent fuel dry storage canisters

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## 1. Introduction

### 1.1 Background and objective

Regulation for the independent storage of spent nuclear fuel (i.e., 10 CFR 72.122(h)(4) [1]) requires that a spent fuel storage system should be continuously monitored with routine surveillance programs or active instrumentations. The continuous monitoring system during storage especially enables us to determine the status of a dry storage system and to verify the continued efficacy of the system on the basis of measurements of specified parameters including temperature, radiation, confinement, and functionality and/or characteristics of components of the system. Storage confinement systems must have the capability for continuous monitoring in a manner such that the licensee will be able to determine when corrective action needs to be taken to maintain safe storage conditions. Many countries have been trying to develop structural integrity monitoring (SIM) technologies for securing the confinement of dry storage canisters by measuring temperature/pressure change or using robot-assisted nondestructive method. These methods are, however, very limited for detecting an early fracture sign in the storage canister and applying to narrow and niche location between concrete overpack and canister. Thus, there is a high-priority technology need for the SIM of dry storage canisters that can achieve real-time, off-site monitoring for dry storage canisters. This study has been performed focused on developing smart paint-based structural integrity monitoring sensors for detecting the fracture sign prior to CISCC in dry storage canisters. Here, we develop a structure integrity monitoring sensor made of at least 3 kinds of smart paints having differently controlled fracture strains and electrical resistances at fracture. When a dry storage canister is deformed, the smart paints painted on the dry storage canister (especially, its outermost surface) are sequentially fractured according to their fracture strains. A change in the electrical resistance of the structure integrity monitoring sensor induced by each smart paint's fracture is used to detect the fracture sign of a dry storage canister prior to CISCC generation therein.

### 1.2 Conceptual design and working principle

The sensing unit consists of N unit sensing elements, each of them is made of smart paints with differently controlled mechanolectrical properties. When the crack

is generated on the dry storage canisters, the unit sensing elements are sequentially fractured which accompanies a stepwise increase in the electrical resistance of the sensing unit. The fracture order of the unit sensing elements is consistent with the fracture strain order of the smart paints. Compared to usual crack propagation sensors, smart paint-based SIM sensor has a noticeable difference. All sensing elements in the usual crack propagation sensors are made of the same material having identical fracture strain, whereas the sensing elements in this sensor are made of smart paints having differently controlled fracture strains and electrical resistances at fracture. The structure integrity monitoring sensor can therefore the fracture sign of a dry storage canister (prior to CISCC generation) and even the existence of fracture. The sensor is painted in a zig-zag shape, instead of a line shape, around the welding zones to cope with the crack growth in an unpredictable direction. In respect to de-cohesion of the SIM sensor from the SUS 304 specimen of interest, this sensor was measured to have no de-cohesion from the specimen up to a strain of 200%. Figure 1 and 2 show a conceptual schematic design and working principle of the smart paint-based SIM sensor respectively.

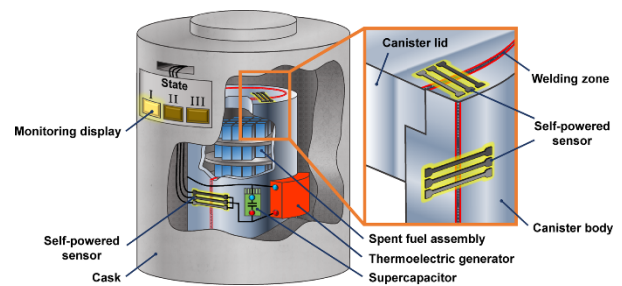


Fig 1. Conceptual schematic design of the smart paint-based SIM sensor

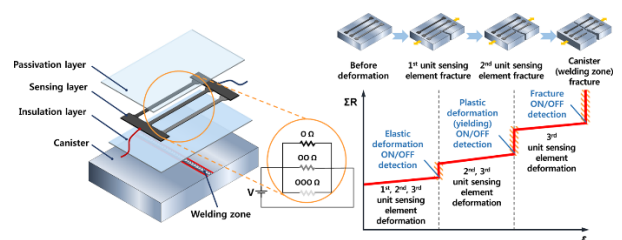


Fig 2. Working principle of the smart paint-based SIM sensor

## 2. Experimental Methods

### 2.1 Sensor Materials

A GnF/PDMS composite is developed as a new functional polymer-matrix composite (PMC) by blending graphite nanoflakes (GnFs) with polydimethylsiloxane (PDMS) where GnF and PDMS are used as reinforcing/conductive filler and an elastic host matrix, respectively [2]. The GnF/PDMS composite, composed of GnFs bound together by PDMS, is a functional PMC with controllable mechano-electrical properties such as elastic modulus,  $E$ , fracture strain,  $\epsilon_f$ , electrical conductivity,  $\sigma$ , and gauge factor, GF. Fig 3 shows changes in the mechano-electrical properties of a GnF/PDMS composite caused by variations in GnF aspect ratio and GnF concentration. The mechano-electrical properties (i.e.,  $E$ ,  $\epsilon_f$ ,  $\sigma$ , and GF) of the functional PMC are significantly changed in a controllable manner by adjusting GnF aspect ratio and GnF concentration. This makes the GnF/PDMS composite as a promising material for the development of smart sensors in SIM. The GnF/PDMS composite was prepared by 3 different methods of casting, brush painting, and spray paint to verify the effect of fabrication method on the mechano-electrical properties of the smart paint, as shown in Fig. 4.

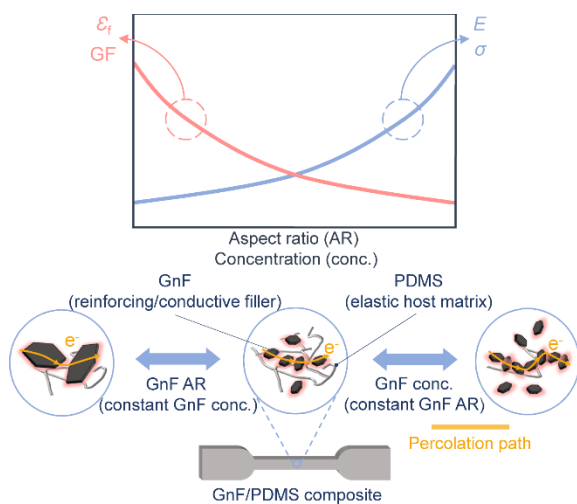


Fig 3. Changes in the mechano-electrical properties of a GnF/PDMS composite (reproduced from Ref [3, 4]).

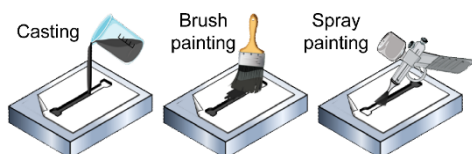


Fig. 4. 3 kinds of fabrication methods for smart paint.

### 2.2 Tensile Tests

For the dumbbell-shaped and circumferentially welded SUS-304 test specimens with a diameter and thickness of 38.1 and 0.9 mm respectively, the post-crosslinking elastic modulus was quantified with tensile test. The load-displacement curve obtained from tensile tests was used to calculate the post-crosslinking elastic modulus. A customized tensile testing system (Instron-5569) was used to measure the mechano-electrical properties of the GnF/PDMS composite concurrently. The system consisted of a z-axis moving stage equipped with a load cell, a multimeter (34401A, Keysight Technologies Ltd., UK), and an acquisition system for force-time (F-t) and resistance-time (R-t) data (Fig 4). The z-axis moving stage moved at a constant crosshead speed of 1 mm/s and the load cell had a capacity of 50 kN; the data acquisition rate was 0.1 kS/s. Each specimen was mounted to the z-axis moving stage using acrylic holder on each of which a Cu electrode was patterned; the specimen and acrylic holds (with Cu electrodes) were pneumatically gripped at a constant operating pressure to secure firm contact between specimen and electrode keeping the same contact area. The F-t and R-t data from tensile tests were used to create mechanical stress vs. strain ( $\sigma_m$  vs.  $\epsilon$ ) and resistance vs. strain (R vs.  $\epsilon$ ) graphs. The mechano-electrical properties (elastic modulus, fracture strain, conductivity, and gauge factor) of the GnF/PDMS composite were estimated from the graphs.

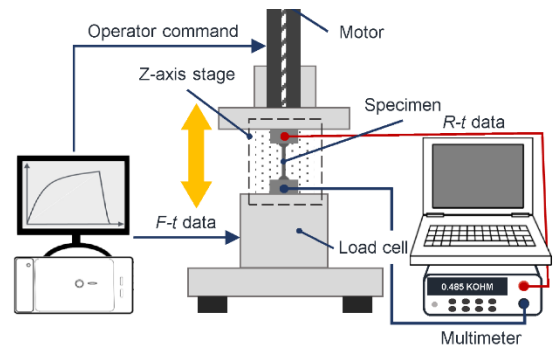


Fig 4. Experimental setup for the characterization of the GnF/PDMS composite as reproduced from Ref [3, 4]

## 3. Results and discussion

We investigated the effects of fabrication method (i.e., casting, brush painting, and spray painting) and GnF concentration on the mechano-electrical properties of GnF/PDMS composites. The elastic modulus and fracture strength of smart paints were measured to be influenced by fabrication method as well as GnF concentration, as shown in Fig. 5. In contrast to the proportionality between elastic modulus and filler concentration in other PMCs, the smart paints showed nonmonotonic behavior in the elastic modulus. This is because pores (not shown here) are generated within the smart paints during fabrication process, especially when a GnF concentration is high (e.g., GnF concentration of higher than 15.0 wt.%). The pores result in a decrease in

the elastic modulus of smart paints. The elastic modulus was also affected by fabrication method. The spray-painted smart paint was measured to have higher elastic modulus than casted- and brush painted-ones. This means a spray painting method is effective to suppress the generation of pores during fabrication process. Fracture strength was in inverse proportion to GnF concentration; the fabrication method had a significant effect on fracture strength. This shows the fracture strength (or fracture strain) of smart paints can be controlled by adjusting GnF concentration and applying a specific fabrication method.

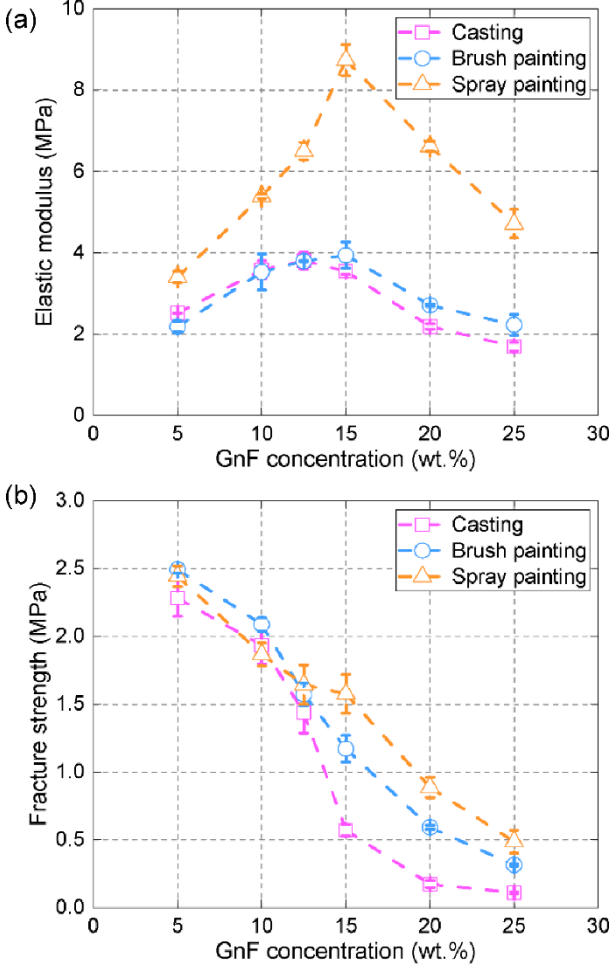


Fig. 5. Elastic modulus and fracture strength as a function of GnF concentration obtained from 3 kinds of smart paints.

Next, the effect of fabrication method on the electrical properties (especially, sheet resistance) of smart paints was explored for 3 kinds of smart paints, as shown in Fig. 6. Their sheet resistance was inversely proportional to GnF concentration for all smart paints. This is a natural consequence because the GnF is an electrically conductive filler in the smart paints. Among casting, brush painting, and spray painting, the smart paint prepared with spray painting were measured to have the lowest sheet resistance. This can be explained by the amount of pores introduced during fabrication process. The amount of pore which was determined by measuring the density of each smart paint were observed to increase

as the GnF concentration; the generation (or amount) of pore was suppressed by applying spray painting instead of casting and brushing painting.

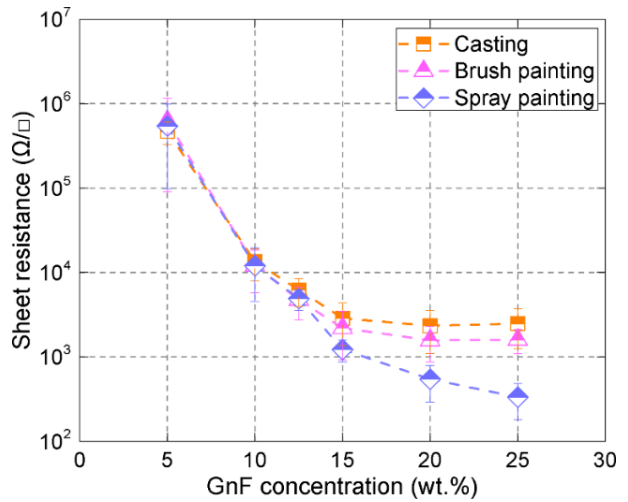


Fig. 6. Sheet resistance as a function of GnF concentration measured from 3 kinds of smart paints.

The mechano-electrical properties were characterized from the SUS-304 tensile test to verify whether this smart material sensor application is appropriate for detecting the fracture sign, in distinction from crack generation, in dry storage canisters. There is a clear indication that the stress increases as the strain increases while the resistance was on a plateau before the strain value reaches sensor breakage point. The fracture signs of the specimen, in other words, three distinct sensor breakages were detected at strain value of 0.009, 0.014 and 0.035 respectively as illustrated in Fig 6. The 3 sensing elements of the sensor painted on the SUS-304 mock-up of a dry storage canister were sequentially fractured at their fracture strains when the mock-up was deformed, which led to successful detection for the fracture sign of the mock-up prior to its physical fracture. The tensile test specimens corresponding to each sensor breakage are shown in Fig 7. The distinct specimen fracture behaviors were well matched with corresponding sensor breakage signals.

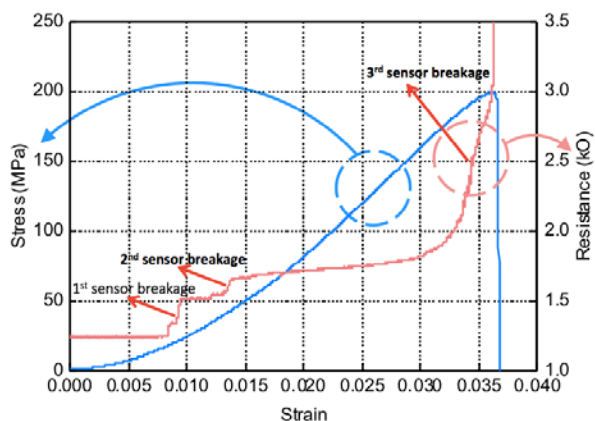
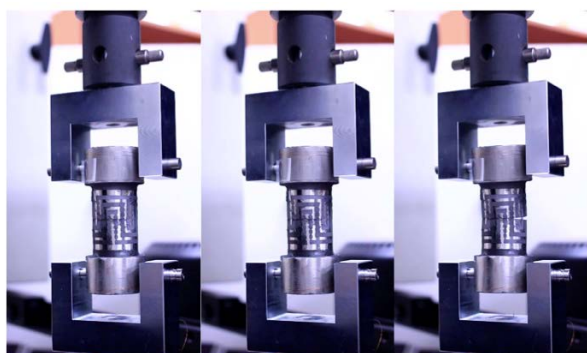


Fig 6. Stress vs. strain and resistance vs. strain graphs obtained from the specimen with SUS-304 tensile test



(a) 1<sup>st</sup> sensor breakage (b) 2<sup>nd</sup> sensor breakage (c) 3<sup>rd</sup> sensor breakage

Fig 7. SUS-304 tensile test specimen corresponding to sensor breakages

#### 4. Conclusions

The characterization study on the mechano-electrical properties of smart material sensors was performed to investigate the potential for application of the smart material to the structural integrity monitoring sensor for detecting the fracture sign, in distinction from crack generation, in dry storage canisters. The analysis demonstrated that the fracture signs of the specimen, in other words, three distinct sensor breakages were detected at certain strain values. The distinct specimen fracture behaviors were also well matched with corresponding sensor breakage signals. Structural integrity monitoring of dry storage canisters by the smart paint-based sensors would contribute to improve the safe operation of dry storage canisters and give an on-time preparation for the licensing of dry storage system and international-level protocol preparation for dry storage canisters. The mechano-electrical properties of smart material sensors at high temperature and irradiation environmental conditions are planned as a future study.

#### 5. References

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- [2] R.-M. Wang, S.-R. Zheng, Y.-P. Zheng, 1 - Introduction to polymer matrix composites, Polymer Matrix Composites and Technology, Woodhead Publishing 2011, pp. 1-548..
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