

# Carbon Nanotube Coated Piezoelectric Ceramic for Self-Health-Monitoring

Lunwei Zhang, Giulia Lanzara, and Fu-Kuo Chang

*Department of Aeronautics and Astronautics, Stanford University  
Stanford, California 94305, U.S.A.*

*lunwei@stanford.edu  
glanzara@stanford.edu  
fkchang@stanford.edu*

## ABSTRACT

The sensor signal of Lead Zirconate Titanate (PZT) piezoelectric sensors/actuator surface mounted to a structure with a thin adhesive layer is known to be influenced by the bondline quality and integrity. Monitoring the bondline health of sensor/actuators integrated into structures is becoming a major concern to guarantee the success and reliability of Structural Health Management systems. The design of a carbon-nanotube-coated PZT (CPZT) sensor was presented in earlier work shown that the bondline of CPZTs mounted on a structure, can be up to 274% stronger than that of conventional PZTs. A CPZT consists of a standard PZT surface coated with a high-density array of oriented CNT-nanoelectrodes (CNTs-NEA). The CNTs-NEA in the interface plays the role of electrodes and of reinforcing filler material. This paper presents results indicating that CPZTs have better performance than conventional PZTs because CNTs in the interface can additionally allow monitoring the bondline integrity during manufacturing and in-service life of a structure. Tests were performed on CPZTs surface mounted on a metal structure with a thin non-conductive adhesive layer. CNTs in the interface were used to monitor adhesive curing by detecting electrical resistance variations of the interface due to phase changes in the adhesive during curing. Crack and debond formation in the interface were monitored in a similar approach. The CPZT is unique in that it is the only existing PZT that, not only has a stronger interface, but is also capable of self-monitoring the health of its bondline which is essential for accurate and reliable SHM systems.\*

## 1 INTRODUCTION

Guaranteeing safety, performance and reliability of a structure through its entire service life is critical. Current work is approaching this issue with Structural Health Management (SHM) systems which not only have the capability to continuously monitor the health of a structure and detect damage, severity and location (diagnosis), but also can predict the remaining life of the structure and its future performance capability (prognosis). Typically prognosis models use a determined level of structural health or damage to identify life expectancy of the structure, thus SHM solutions require hardware architecture design, the integration of sensors and actuators in structures, computational processors, communication networks as well as diagnostic and prognostic algorithms at the subsystem and at the overall system level. It is evident that an accurate assessment of the effective conditions of the structure during its life-time is crucial to guarantee the reliability and output of prognostic algorithms. Structural health is normally monitored by means of built-in distributed sensor/actuator networks often made of the piezoelectric material Lead Zirconate Titanate (PZT). PZT sensors/actuators are permanently bonded to the structure with an interfacial adhesive layer which has the primary role of coupling the sensors to the structure for stress/strain transfer (Staszewski, Tomlinson and Boller, 2003; Lin et al., 2005). It has been shown that the bondline properties during PZT adhesive curing (Lin and Chang, 2001) and a partial debond of PZT sensors/actuators from the structure (Lanzara and Chang, 2009b) are the cause of phase and amplitude changes in the detected PZT signals. These changes, if not properly detected at the sensor/actuator subsystem level, affect the performance and control capabilities of the underlying monitoring and prognostic systems at the system level. Thus it is extremely necessary to monitor and diagnose the health of sensor/actuators and their bondline properties during manufacturing and during the service life of a structure. The major critical

---

\* Lunwei Zhang et al. This is an open-access article distributed under the terms of the Creative Commons Attribution 3.0 United States License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited.

parameters that affect the sensors output are: (1) adhesive properties which need to be controlled during the adhesive curing and; (2) adhesive integrity during the life of a structure. Currently, the bondline integrity can be detected with NDT techniques, such as X-ray and acoustic scanning. However, these approaches are: (a) extremely time consuming especially in the case of large scale sensor networks, (b) do not allow monitoring of sensors internally embedded within structures, (c) cannot provide any real-time information about the bondline and, (d) cannot monitor the adhesive properties during manufacturing and during the service life of a structure which, as discussed above, is essential for the underlying SHM systems. The ideal solution is to integrate structures with a network of sensors/actuators which have a stronger bondline and that, at the same time, have the capability to monitor, in real-time, the adhesive properties and the interface integrity. This information can then be used to improve the output of prognostic algorithms.

The design of a carbon nanotube (CNTs) coated PZT (CPZT) was recently developed and was shown to allow a reduction of the risk of bondline failure or degradation (Lanzara and Chang, 2009a). In this design the silver paste electrode of standard PZTs (weak link) is replaced with a high density array of CNTs nanoelectrodes (CNTs-NEA) that simultaneously play the role of reinforcing the adhesive and of conductive path.

In this paper an investigation is performed to use the CPZT design to determine the bondline health over time. Particular focus is given to the monitoring of the adhesive curing and the bondline integrity. These results can be used to modify diagnostic algorithms to account for degradation and in the prognostic models to accurately predict the remaining life of a structure.

## 2 PROBLEM STATEMENT

Interface degradation, adhesive curing and aging may occur over time in the bondline between PZTs and a host structure, compromising the performance and reliability of the Structural Health Management systems. It is necessary to develop PZT sensor/actuators that can be coupled to diagnostic and prognostic algorithms and that have the capability to monitor the health of the PZT bondline. The objective of this study is to investigate the use of carbon nanotube coated PZTs (CPZT) (Lanzara and Chang, 2009a) to monitor the bondline integrity and adhesive curing for PZT actuators bonded to a metal plate.

## 3 METHOD OF APPROACH

In this investigation, it is proposed to evaluate the capability of CPZT sensor/actuators to monitor the health of the bondline between the PZTs and a host metal structure. The response of the CPZT is compared with the response of standard PZTs mounted on the structure by means of a conductive adhesive layer and a non-conductive adhesive layer. Figure 1 shows the three cases studied. The CPZT design in Figure 1a consists of a standard PZT ceramic disc (sandwiched between two silver paste electrodes) surface coated with a high density nano-electrode array of aligned carbon nanotubes (CNTs-

NEA) and bonded to a metal substrate with a thin non-conductive adhesive layer. The resulting bondline is reinforced with oriented CNTs (Lanzara and Chang, 2009a). The CNTs-NEA allows continuous monitoring of the interface by measuring the electrical resistance between the metal plate and the silver paste electrode in contact with the CNTs tips. Figures 1b and 1c show a schematic of a standard PZT surface mounted on a metal plate with a non-conductive (NC) and conductive (CG) adhesive layer respectively.

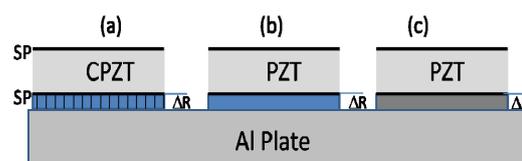


Figure 1. Schematic of the tested bondlines:  
(a) CPZT; (b) PZT with non-conductive adhesive;  
(c) PZT with conductive adhesive

## 4 INTERFACE CURE MONITORING TEST

An investigation is performed to validate the possibility of using the CPZT design to monitor adhesive curing. A Smart Layer manufactured by Acellent Inc. with an integrated  $\frac{1}{4}$ " PZT is used as the baseline sample. The Smart Layer consists of a Kapton HN dielectric substrate that hosts a standard PZT coated with silver paste (SP) electrodes. The SP electrodes are wired by means of copper wires prepatterned on the Kapton layer. The CPZT was fabricated by means of the method described in (Lanzara and Chang, 2009a). To summarize; a forest of aligned CNTs (about  $70\mu\text{m}$  thick) was grown on a silicon substrate with a chemical vapor deposition process using Al/Fe ( $40\text{\AA}/50\text{\AA}$ ) catalyst nanoparticles and Ethylene as the hydrocarbon source. The CNT forest was then transferred on the exposed SP electrode by means of an ultra-thin conductive (CG) adhesive layer. Figure 2a shows a picture of the fabricated sample, the black area in the figure is a macroscopic view of the CNT forest where the CNTs axes are perpendicular to the PZT substrate. The CPZT was then bonded to an Aluminium plate with the NC epoxy Hysol® EA 9396 which wicked into the nanochannels in the CNT forest. The resulting CPZT bondline can be viewed by means of a Scanning Electron Microscope (SEM) image shown in Figure 2b which shows that CNTs are fully embedded and oriented in the thin adhesive layer. With this process the CNTs tips were left in contact with the underlying Al plate establishing electrical contact between the SP electrode and the metal plate.

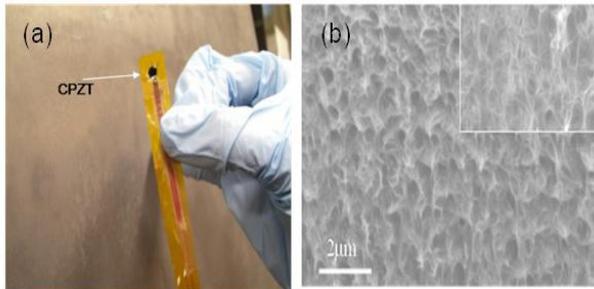


Figure 2. (a) Smart Layer with CPZT; (b) SEM image of a CPZT bondline, side view

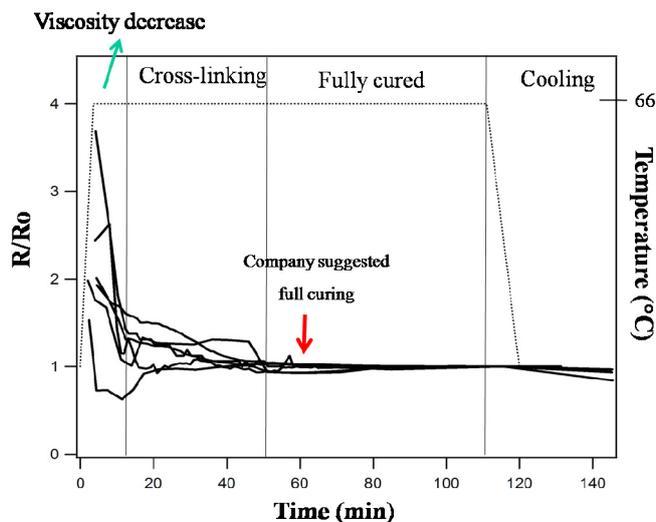


Figure 3. Curing monitoring over time

The principle behind this design is that CNTs in the interface work as sensors to monitor the epoxy curing. This is done by measuring the electrical resistance between the SP electrode and the Al plate during the curing process (as schematically shown in Figure 1) with a Fluke 77-IV digital multimeter. The as-prepared samples were inserted in an oven for 110 minutes at 66°C. The CPZT bondline sensing capability was compared with that of a CG interface and of a NC interface

The test results clearly show that adhesive curing can only be monitored with the CPZT samples. No significant resistance change was in fact observed for the CG interface (Figure 1c) where the electrical resistance was found to constantly have an approximate value of 0.4 Ohm throughout the entire curing process. Standard PZTs bonded to the Al plate with a NC adhesive (Figure 1b), did not allow any detectable resistance value during the curing process. This expected result is in agreement with the insulating nature of the NC adhesive. On the contrary, the CPZT interface was found to be extremely sensitive to the epoxy curing. Figure 3 shows the curing test results for four CPZT samples that were tested with the procedure described above. The graph represents the electrical resistance normalized by the final resistance of the CNTs ( $R/R_o$ ) versus time as well as the trend of the sample temperature over time. The sample temperature was first increased to reach the 66°C pre-set temperature of the

oven, then was kept constant and then was brought back to room temperature. The electrical resistance of the interface was monitored during the entire temperature cycle and for several days after cool down. It was observed that the electrical resistance first showed a rapid decrease, after 40 minutes the resistance decreased at a slower rate until reaching, at 60 minutes, an almost constant value. It is to be noted that 60 minutes is the time suggested by the epoxy manufacturer to reach full cure at this temperature. A further  $R/R_o$  decrease was detected during the cooling step reaching a final constant value at room temperature. It is believed that the electrical resistance variation over time can be correlated to the phase changes thus the viscosity changes that typically characterize epoxies (Shigue et al. 2004). The four different stages described above seem to correspond to: (1) rapid viscosity decrease; (2) adhesive cross-linking; (3) complete crystallization at 60 minutes, and (4) CNTs resistance variation due to temperature decrease. Viscosity changes may affect the electrical contact between the CNTs tips and the metal plate. When the epoxy becomes more fluid (lower viscosity) it starts flowing and thins down the bondline thus, in most cases, improving the contact (resistance decrease) between the CNTs tips and the metal plate. This phenomenon goes on, at different rates, until full crystallization is reached (60 minutes). From this point on, the electrical resistance is nearly constant due to a constant temperature and a rigid CNTs tips/metal configuration. As soon as the sample is cooled down to room temperature, the resistance decreases again mostly because of the lower electrical resistance of CNTs at room temperature. More studies are being performed to validate the assumptions of the physics behind the observed phenomenon. These results suggest that the CPZT has good potential to monitor curing and aging of sensor interfaces and can also be used to optimize the curing condition.

## 5 INTERFACIAL CRACK DETECTION

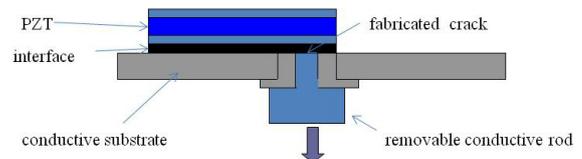


Figure 4. Test setup and crack/debond fabrication

In the previous section the CPZT sensitivity to epoxy curing was discussed. Here an investigation is performed to use the CPZT for debond/crack detection. The concept is that the forest of oriented CNTs is sensitive to debond/cracks that may occur in the interface. In Figure 4 the test setup is presented. A small interfacial debond was artificially generated by fabricating a removable metal rod (diameter of 2 mm) and fixing it with an

Al substrate. A CPZT was mounted and fully cured on top of the rod with the same procedure described in the previous section. The rod was removed to generate a debond/crack in the interface. The electrical resistance between the SP electrode surface coated with the CNTs and the Al plate in contact with the opposite CNTs tips, was measured before and after the crack. The electrical resistance was recorded for 3 days before the crack and then was monitored for 1 day after the crack. Electrical resistance measurements were achieved by means of a milli-ohmmeter. The response of the CPZT interface was compared with that of a standard PZT mounted on the Al plate with a CG adhesive layer. The test results are shown in Figure 5 and Figure 6 where the percentage of electrical resistance variation is plotted, over time, before and after the crack. These results clearly indicate that the CPZT is very sensitive to debond/crack generation. The resistance was in fact found to vary 11.1% after crack. Much lower sensitivity was detected in the CG interface where the resistance changed only of 0.067%. From the test result, the CPZT interface has good potential to detect the interfacial crack/debond with the resistance monitoring. This will offer a new approach for sensor health self-monitoring.

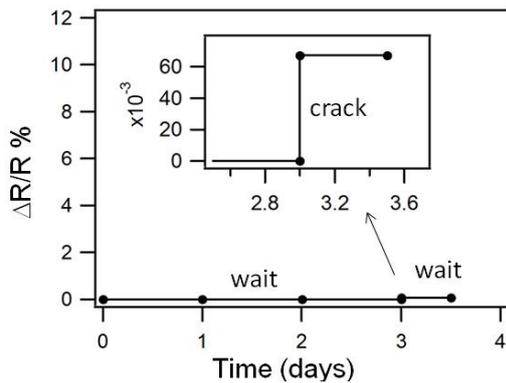


Figure 5 Debond/crack test results for a conductive interface

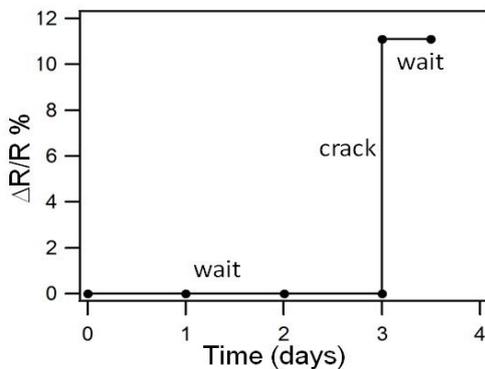


Figure 6. Debond/crack test results for a CPZT interface

## 6 COMPARISON OF THE CPZT DESIGN AND STANDARD PZT AS BONDLINE SENSORS

The results described in the previous sections are summarized in Table 1. It was found that the CPZT design can be used to monitor the bondline integrity, curing and aging. Standard PZTs bonded with a conductive adhesive to an Al plate, are not sensitive to adhesive curing monitoring and are not sufficiently sensitive to debond/crack detection. Standard PZTs bonded to a substrate with a non-conductive adhesive can't be used to monitor the sensors/actuators bondline.

These results show that the CPZT design has a great potential to be implemented in Structural Health Management systems and improve the prediction of life-time of a structure.

Table 1. Comparison of self-sensing capabilities of different interfaces

Tests	Resistance response from CPZT interface	Resistance response from CG interface
Curing	sensitive (10 samples)	no potential (3 samples)
Interfacial crack	Sensitive (6 samples)	no potential (3 sample)

## 7 CONCLUSION

An investigation was performed to validate the use of carbon nanotube coated piezoelectric ceramic discs as interfacial self-health monitoring sensors. The CPZT design is made by coating a standard PZT with a high density array of oriented CNTs nanoelectrodes (CNTs-NEA). The CPZT was surface mounted to a metal substrate by means of a thin non-conductive adhesive layer which resulted in a bondline integrated with the CNTs-NEA which played the role of reinforcing filler material, of electrode as well as self-sensing elements. The CPZT bondline was compared with that of standard PZTs bonded to a structure with a conductive as well as a non-conductive adhesive layer. It was found that CPZTs only could be used to monitor the bondline health. The CPZT was found to be extremely sensitive to adhesive curing and interfacial crack/debond. It is believed that the proposed self-monitoring design has a great potential to be used in diagnostic and prognostic algorithms to predict the remaining life of smart structures.

## ACKNOWLEDGMENT

The authors are grateful to the Air Force Office of Scientific Research (AFOSR) under Contract No: 1085749. The AFOSR program monitor is Dr. Les Lee.

## REFERENCES

- (Lanzara,2009a) G. Lanzara and F.-K. Chang (2009a). Design and Characterization of a carbon nanotube-reinforced adhesive coating for piezoelectric ceramic discs, *Smart Materials and Structures*, in Press.
- (Lanzara,2009b) G. Lanzara, Y. Yoon, Y. Kim, F.-K. Chang (2009b). Influence of Interface Degradation on the Performance of Piezoelectric Actuators, *Journal of Intelligent Material Systems and Structures*, vol. 20, pp 1699-1710.
- (Lin ,2002) M. Lin and F.-K. Chang (2002). The Manufacture of Composite Structures with a Built-in Network of Piezoceramics, *Composites science and Technologies*, vol. 62:7-8, pp 919-939.
- (M. Lin, 2001). M. Lin, et al., SMART Layer and Smart Suitcase for Structural Health Monitoring Applications, in *Proceedings of SPIE on smart structures and material systems*, SPIE Optical Engineering Press.
- (Shigue,2004) C. Y. Shigue, R. G. S. dos Santos, C. A. Baldan, and E. Ruppert-Filho (2004). Monitoring the Epoxy Curing by the Dielectric Thermal Analysis Method, *IEEE Transactions on Applied Superconductivity*, vol. 14: 2, pp1173-1176.
- (Staszewski,2003) W. J. Staszewski, G. Tomlinson, C. Boller (2003). *Health Monitoring of Aerospace*