Ultrasonics 82 (2018) 178–187

Contents lists available at ScienceDirect

Ultrasonics

journal homepage: www.elsevier.com/locate/ultras

Simultaneous sound velocity and thickness measurement by the ultrasonic pitch-catch method for corrosion-layer-forming polymeric materials

Masahiro Kusano [a,*], Shota Takizawa [b], Tetsuya Sakai [c], Yoshihiko Arao [d], Masatoshi Kubouchi [d]

[a] *National Institute for Materials Science, Japan*
[b] *Graduate School of Industrial Technology, Nihon University, Japan*
[c] *College of Industrial Technology, Nihon University, Japan*
[d] *Department of Chemical Science and Engineering, Tokyo Institute of Technology, Japan*

ARTICLE INFO

Article history:
Received 26 February 2017
Received in revised form 8 June 2017
Accepted 6 August 2017
Available online 10 August 2017

Keywords:
Ultrasonic testing
Non-destructive evaluation
Simultaneous sound velocity and thickness measurement
Pitch-catch method
Corrosion-layer formation

ABSTRACT

Since thermosetting resins have excellent resistance to chemicals, fiber reinforced plastics composed of such resins and reinforcement fibers are widely used as construction materials for equipment in chemical plants. Such equipment is usually used for several decades under severe corrosive conditions so that failure due to degradation may result. One of the degradation behaviors in thermosetting resins under chemical solutions is "corrosion-layer-forming" degradation. In this type of degradation, surface resins in contact with a solution corrode, and some of them remain as a corrosion layer on the pristine part. It is difficult to precisely measure the thickness of the pristine part of such degradation type materials by conventional pulse-echo ultrasonic testing, because the sound velocity depends on the degree of corrosion of the polymeric material. In addition, the ultrasonic reflection interface between the pristine part and the corrosion layer is obscure. Thus, we propose a pitch-catch method using a pair of normal and angle probes to measure four parameters: the thicknesses of the pristine part and the corrosion layer, and their respective sound velocities. The validity of the proposed method was confirmed by measuring a two-layer sample and a sample including corroded parts. The results demonstrate that the pitch-catch method can successfully measure the four parameters and evaluate the residual thickness of the pristine part in the corrosion-layer-forming sample.

1. Introduction

Since thermosetting resins such as epoxy resin (EP) and unsaturated polyester (UP) have excellent resistance to chemicals, fiber-reinforced plastics (FRPs) composed of such resins and reinforcement fibers are widely applied to construction materials of equipment, e.g., storage tanks, pipes, and decks in chemical plants. Such equipment is usually used for several decades under severe corrosive conditions so that failure due to degradation may result [1,2]. It is therefore necessary to periodically inspect the equipment, evaluate its failure risk based on the degradation, and maintain it within the acceptable risk levels. Because a corrosion layer is formed on the inner surfaces of tanks and pipes, non-destructive testing (NDT) from outside the equipment is ideal for the practical application. However, there is no non-destructive technique to measure the corrosion depth for polymeric materials.

One of the degradation behaviors in thermosetting resins under chemical solutions is the "corrosion-layer-forming" type [3,4]. In this type of degradation, a corrosive solution degrades surface resins by chemical reactions, and then some degraded resins remain on the surface as a corrosion layer. Here, "corrosion" is defined as degradation due to chemical reactions such as hydrolysis of the polymer. Corroded resins turn into lower molecular weight substances that usually have inferior mechanical properties and chemical resistance than the pristine resin. A solution easily penetrates into such low molecular weight corrosion layers and corrodes the polymer at the interface between the pristine part and the corrosion layer. Thus, while the thickness of the pristine part gradually decreases, the corrosion layer becomes thicker. It is well known that UP under acid or basic aqueous solutions shows this type of degradation because ester bonds in UP are easily cleaved by such solutions because of hydrolysis [3,4,5].

---

* Corresponding author.
E-mail address: KUSANO.Masahiro@nims.go.jp (M. Kusano).

http://dx.doi.org/10.1016/j.ultras.2017.08.001

In order to measure the thickness of the pristine part, pulse-echo ultrasonic testing according to JIS Z 2355 [6] and ASTM E797 [7] is the most promising candidate NDT method. This method uses a piezo-electric transducer (normal probe) to propagate ultrasonic waves into a material and detects the pulse echo reflected from its opposite side. The thickness of the material can be obtained by multiplying half of the round-trip time of the wave and the already-known sound velocity of the material. Furthermore, ultrasonic testing devices for multilayer thickness such as rubber tires, painted and coated surfaces are available in the market. However, the velocity of sound in a material depends on its density and elasticity; the velocity of a polymeric material decreases with absorbed water content [8] and increases with the degree of curing [9]. Therefore, it is impossible to accurately measure the thickness of such a material by the pulse-echo method without evaluating the sound velocity by another method. The sound velocity in the corrosion layer also depends on the degree of corrosion because of the low molecular weight of the resin and also absorption of the aqueous solution [3,4]. Thus, the velocity decreases to an intermediate value between that of the pristine resin (about 2500 m/s) and an aqueous solution (e.g. the velocity of sound in water at room temperature, which is about 1500 m/s [10]). Since corrosion behavior and the degree of corrosion depend on the environment (type of solution, concentration, temperature, etc.), the velocity of sound in the corrosion layer is unpredictable. In addition, although ultrasonic waves reflect at an interface between two materials that have different sound velocities, the pulse echo reflected at the interface between the pristine part and the corrosion layer cannot be detected—or is quite small—in the waveform. The reflectance R depends on the acoustic impedance Z of two materials, which is the product of the density and the velocity;

$$R = \frac{(Z_2 - Z_1)^2}{(Z_2 + Z_1)^2} \tag{1}$$

While the impedances of the pristine part and the corrosion layer are the same order of magnitude that of air is four orders of magnitude lower. Thus, the echo at the pristine part/corrosion layer interface cannot be detected. Instead, the echo reflected at the opposite surface (the interface between the corrosion layer and air or solution) can appear clearly. If we try to measure the thickness by the pulse-echo method based on the round-trip time of the pulse echo (including pristine and corroded regions) and the sound velocity of the pristine material, the calculated thickness will be greater than the true thickness. Thus, the conventional pulse-echo method can completely fail to measure the thickness of the undamaged part of the corrosion-layer-forming sample. In fact, a specific example of a case where the pulse-echo method produces totally incorrect results will be presented later in Section 4.3.2. of this paper. It is necessary to develop a method to measure the thicknesses and sound velocities of the corrosion layer and the pristine part simultaneously.

Hsu and Hughes introduced four different configurations of a transmitter and receiver of ultrasonic waves for simultaneous sound velocity and thickness measurement [11]. Gomez also presented a method for determining the sound velocity and the attenuation coefficient of ultrasound in materials, as well as the thickness, using an air-coupled transducer and receiver with analyses of both the phase and amplitude spectra [12]. Their methods produced good results for the simultaneous measurement and were applied to the evaluation of porosity (void content) of composites [11] and thicknesses of composite plates [12]. However, the configurations of the probes (transmitter and receiver) were in a transmission set-up, and it is still difficult for these methods to evaluate layered samples. While the ultrasonic wave path in these methods is normal to the samples, an oblique path by angle probes has some advantages in certain cases, e.g., cracks parallel to the depth direction. An advanced method using angle probes was proposed by Yang et al. for fiber orientation detection in unidirectional carbon fiber reinforced plastic (CFRP) composites [13]. Their map of the ultrasonic amplitude analysis shows a clear fiber direction in the 7-mm-thick CFRP sample because the oblique path of the ultrasonic waves is more sensitive to subtle flaw conditions in the composite than the normal path.

In this study, we investigated a pitch-catch method using a pair of normal and angle probes that simultaneously measures four parameters: the thicknesses of the pristine part and the corrosion layer, and their respective sound velocities. An oblique ultrasonic wave transmitted by an angle probe refracts at the interface between materials with different sound velocities in accordance with Snell's law [14]. Thus, an oblique ultrasonic wave refracts at the interface between the pristine part and the corrosion layer. The unknown parameters can be derived from multiple measurements of time-of-flight data in the oblique path with different probe separations. This method was confirmed by measuring a two-layer sample composed of epoxy resin and silicon rubber which imitates a pristine part and a corrosion layer. Then, the method was also applied to the measurement of a corrosion-layer-forming sample and compared with the true thicknesses and sound velocities. As a result, we demonstrated that the pitch-catch method can successfully measure the four parameters and evaluate the residual thickness of the pristine part degraded in the corrosion-layer-forming sample.

2. The pitch-catch method

2.1. Measurement principle for a one-layer sample

Before explaining the measurement principle for a two-layer sample, we describe the simultaneous measurement of thickness L and sound velocity C of a one-layer material (just a plate-like material) by using a pair of angle and normal probes. Then the method is extended to the case of the two-layer sample.

The pair of probes is set on one side of the material as shown in Fig. 1(a). The angle probe transmits an ultrasonic wave and the

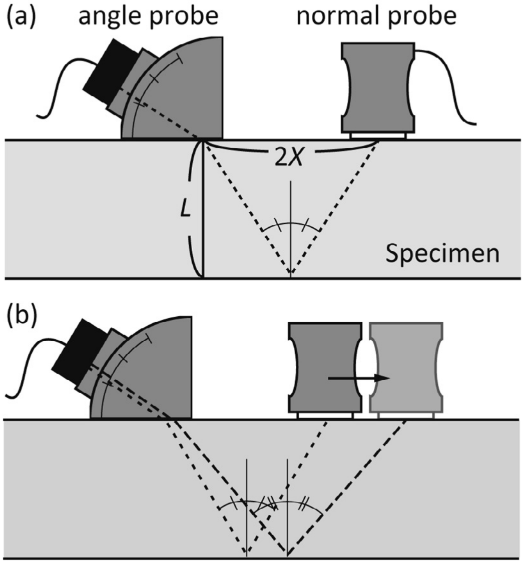

Fig. 1. (a) The schematic image of the pitch-catch method for one-layer material. The path of the ultrasonic wave is illustrated by the broken line. (b) The path varies with the distance between the probes.