Ultrasonic Measurement of Oxide Layer Thickness and Oxide Influence on Flaw Detection and Sizing in Pressure Tubes

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Abstract

Oxide layer on the pressure tube inside surface in CANDU reactors masks flaws; complicates their detection, characterization and sizing; entails degradation of the material properties and brittle microstructure; and leads to many other different issues. Various ultrasonic methods, techniques, and transducers, used for detection, characterization and sizing of different flaws in Zr-2.5%Nb tubes covered by oxide ZrO$_2$ and also for oxide layer detection and its thickness measurement in pressure tubes are described and analyzed. Obtained experimental results have demonstrated that using ultrasonic waves, it is quite possible to characterize and size flaws covered by oxides and also detect and measure oxide layers in pressure tubes by using ultrasonic signals in the time and frequency domains.

Keywords: ultrasonic inspection of pressure tubes, oxide layer detection and sizing.

1. Introduction

During CANDU reactor operation one of the main components - the pressure tube (PT) - , made from zirconium alloy Zr-2.5%Nb has oxide layer ZrO$_2$ on the inner surface (or inside diameter – ID) of PT, which masks flaws, can cause additional refraction and reflection of ultrasonic (UT) beam complicating flaw detection, characterization and sizing, and leads to many other different issues.

Oxide layer thickness grows up to ~150µm at the end of tube design life. Actual oxide layers on the PT ID are non-uniform with comparatively rough surface. If localized, oxide can mask true depth measurements and will affect repeatability. It can lead to an increased likelihood to oversize depth on recently created flaws, since oxide layer on such flaws is usually thinner than layer on the ID surface of PT. Usually oxide layer ZrO$_2$ is distributed non-uniformly on the ID. The oxidation can also lead to microstructure changes of the base alloy: grain morphology, phase transforms, alpha-Zr grains structure changes, cracks can appear inside of oxide layer, and latter on these cracks propagate in the substrate in a trans-granular mode, etc. For other flaws, which may be oxide filled, such as crevice corrosion flaws, the presence of thick oxide layers (which are generated as part of the flaw formation process) may lead to difficulties in sizing flaw lengths, width, depth and root radius.

Therefore, generically, the oxidation entails the degradation of the material properties, leads to the brittle Zr-2.5%Nb microstructure, and also drastically decreases the heat transfer from coolant to the moderator. That is why UT examination includes characterization and sizing of flaws covered by oxide and also measurement of the ID oxide layer thickness. The motivation of this work was to evaluate the performance, sensitivity, resolution and accuracy of UT measurements of oxide layer thicknesses and also detection, characterization and sizing of different flaws on the ID of test PT samples with various thicknesses of oxide layers implementing different UT techniques and equipment, and determine the optimum ones.
2. Equipment

Calibrated computer-based UT immersion scanning system was employed for experiments. This UT system includes the Winspect™ data acquisition software, a SONIX STR-8100 digitizer card, and a UTEX UT-340 pulser-receiver.

The tests were performed using various UT probes with different center frequencies $f_c$, focal lengths $F_L$, and aperture diameters $D$.

3. Samples (PT coupons)

Test specimens (coupons) had shape of the segments: $120^\circ$ circumferential width and 15mm axial length. Each coupon contained one ID axial flaw. Two of these flaws, simulating debris frets, had shape of V-notches: $45^\circ$ tip angle with depths 0.5mm and 0.25mm. Two other flaws, simulating bearing frets, had shape of flat-bottom notches: 3mm wide with depths 0.5mm and 0.25mm.

Four coupons had as received oxide layers. Besides these four coupons, we used for investigation twelve coupons after oxidation. These twelve coupons had three different oxide layer thicknesses (within ranges 30-40µm, 90-120µm, and 150-190µm) and four various flaws described above. So in total we investigated sixteen coupon samples with four various types of flaws and four different oxide layers. Each coupon was subject to only one oxidation cycle (from start to finish). The oxide layers were quite uniform along the ID surface and along the notches, but on some samples with deep V-notches the thick oxide layers were non-uniform.

4 Inspection Procedure

Using calibrated computer-based UT immersion scanning system, we performed various pulse-echo (PE) and pitch-catch (PC) circumferential 2D-scans and 3D-scans from the ID [1]. Unfortunately, due to small sizes of all coupons in the axial direction and because all flaws were oriented only in the axial direction, it was impossible to perform axial PC and PE scans. We applied various normal beam (NB) and angle UT transducers [1] with different focal lengths, frequencies and diameters, water-paths, and incident angles.

We tried longitudinal and shear waves propagating in circumferential direction, used such signal processing methods as filtering and averaging, and analyzed various types of images.

We experimented with multiple UT reflections within the tube wall sometimes “amplifying” the weak indications, and investigated results presented in time domain and frequency domain.

As result, we determined maximum detectability, identification, and sizing accuracy of flaws covered by oxides and accuracy measurement of the ID oxide layer thickness, using the UT techniques and probes optimized for the task.
5 Various methods of oxide layer detection and measurement

At first, it is probably worthwhile to briefly review typical currently existing UT and eddy-current (EC) techniques used for thin layer detection and measurement.

The conventional UT PE method, the pulse transference method, and the resonance testing methods are avoided for detection and sizing of thin oxide layers, whose thickness are comparable or smaller than the wavelength [2]. High frequency UT waves can be used, but they are also averted due to very high attenuation, besides these methods continue to be quite expensive. In addition, the high-frequency waves suffer the interference from micro-structural details of the thin layers.

To detect ultra-thin layers of thickness within the range of micrometers and even nanometers, it is necessary to perform an UT experiment at extremely high frequencies. To generate acoustic vibrations in such ultrathin films, a laser-UT system should be used [3]. But of course, a laser-UT system (the most appropriate for this application is a fiber laser-UT system) has all advantages and disadvantages typical for such systems.

The other options are to use the Lamb waves, Love waves or Rayleigh (surface) waves, whose velocities of propagation depend on the relationship between frequency (or wavelength) and oxide layer thickness (dispersion phenomenon). This ratio determines the effective stiffness of the layer and hence the velocity of the wave. Recall also that surface waves become Lamb waves when layer thickness is less than wavelength. Lamb waves can propagate long distances with low attenuation in plate-like structures, and are highly sensitive to small imperfections. Like guided waves in structures, Lamb waves have multiple modes and complicated dispersive characteristics.

However, all these methods, using Lamb waves, Love waves and/or surface waves, and respective equipment sets, also have their own serious disadvantages. For example, Lamb wave technique has a rather low sensitivity, focussing issues, difficulty to excite only one mode required for inspection, sensitive to thickness non-uniformities, and others; surface wave is very sensitive to the surface boundary conditions; shear wave has high attenuation and low signal-to-noise ratio, and so on.

There were also attempts to use high-frequency UT waves (75-100MHz) to detect and size thin ZrO$_2$ oxide layer (20-40µm) in PT by using digital spectral analysis [4].

Oxide layer thickness can be also measured by using EC technique [5-6]. The respective probe contains an EC coil driven by a high frequency current which produces a varying magnetic field around the coil. The high frequency electromagnetic field produced by the coil penetrates the nonconductive oxide layer (recall that ZrO$_2$ has properties of electric insulator) and induces eddy currents in the conductive substrate (PT material). The eddy currents produce an opposing magnetic field that affects the impedance of the coil. Since the impedance variations are strongly dependent on the distance from the coil to the conducting base metal (Zr-2.5%Nb), the probe produces a voltage signal proportional to the thickness of the non-conductive oxide layer (lift-off effect) [5-6]. The respective a lift-off effect based curve can be constructed, with a help of oxide layers thicknesses standards.
Note for comparison, that there is a very different situation with respect to iron oxide (magnetite) layer in boiler, feeder and steam generator tubes. Resistivity of magnetite layer greatly depends on the magnetite composition, age, base material, environmental conditions, temperature, and other factors. As result, the magnetite cannot be always considered an insulator or semiconductor. Sometimes, it becomes a conductor. Magnetite is also very unusual in that it can be both a conductor of electricity at higher temperatures and a strong insulator at low temperatures.

6 Typical UT concepts of oxide layer detection and measurement

In principle, the ability to perform NDE UT characterization and sizing of flaw with oxide layer and measurements of the oxide layer thickness in any required local area of PT or by mapping the whole tube is very attractive.

Generically, the UT measurements are routinely used for PT inspection [1]; they are rapid, sensitive, simple, safe, accurate, reliable and inexpensive. UT data will be obtained in real time in various convenient formats and can be easily analyzed; performed scans can be saved and accessed any time for further analysis. The UT results (images) can be obtained performing 2D B-scans (circumferential and axial), C-scans (amplitude and time-of-flight), and 3D B-scans, providing 3D-images, which are very informative and convenient for analysis. Inspection can be performed using NB and angle probes with different focal lengths, frequencies, diameters, water-paths, and incident angles; applying longitudinal and shear waves, propagating in the axial and circumferential directions in the PE and PC modes of operations [1]. The UT detection and sizing of oxide layer in PT is based on the reflection and refraction of UT waves due to different acoustic parameters of the PT material Zr-2.5%Nb and oxide layer ZrO₂, which are presented below in Table 6-1.

Table 6-1. Acoustical properties of PT material Zr-2.5%Nb and oxide layer ZrO₂

<table>
<thead>
<tr>
<th>Property</th>
<th>Material</th>
<th>PT alloy Zr-2.5%Nb</th>
<th>Dioxide ZrO₂ (from different references)</th>
<th>Difference in relation to PT, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Longitudinal velocity, mm/µs</td>
<td>4.71</td>
<td>4.8-7.0 (average ~5.9)</td>
<td>~30</td>
<td></td>
</tr>
<tr>
<td>Shear velocity, mm/µs</td>
<td>2.35</td>
<td>2.4-3.5 (average ~2.8)</td>
<td>~20</td>
<td></td>
</tr>
<tr>
<td>Density, g/cm³</td>
<td>6.48</td>
<td>5.00-6.15 (average ~5.6)</td>
<td>~14</td>
<td></td>
</tr>
<tr>
<td>Longitudinal impedance, MRayl</td>
<td>31</td>
<td>24-42 (average ~33)</td>
<td>~8</td>
<td></td>
</tr>
<tr>
<td>Shear impedance, MRayl</td>
<td>15</td>
<td>11-22 (average ~17)</td>
<td>~13</td>
<td></td>
</tr>
<tr>
<td>Young’s modulus, GPa</td>
<td>91</td>
<td>100-250 (average ~195)</td>
<td>~100</td>
<td></td>
</tr>
<tr>
<td>Poisson ratio</td>
<td>0.333</td>
<td>0.22-0.32 (average ~0.27)</td>
<td>~18</td>
<td></td>
</tr>
<tr>
<td>Shear modulus, GPa</td>
<td>34</td>
<td>53-86 (average ~69)</td>
<td>~100</td>
<td></td>
</tr>
<tr>
<td>Bulk modulus, GPa</td>
<td>97</td>
<td>72-212 (average ~150)</td>
<td>~51</td>
<td></td>
</tr>
</tbody>
</table>
Note, that a wide range of elastic and mechanical parameters of oxide, presented in Table 6-1, can be explained not only by different reference sources, but mainly by various environment conditions of their formations and various service time intervals.

This Table shows that most probably, the best technique to detect and measure the oxide layer is to use PC mode shear wave propagating at angle within the oxide layer. Then, after refraction at the interface oxide/metal, the acoustic beam trajectories will have maximum difference in amplitude and time-of-flight for the un-oxidized and oxidized cases.

Two different UT methods can be used to measure non-destructively thickness of the oxide layer on the PT ID.

The first method is using the normal beam (NB) PE single UT probe positioned inside the tube perpendicularly to the PT surface, see schematics of this technique in Fig. 6-1a. This probe transmits and receives longitudinal UT wave propagating perpendicular to the tube wall and reflected from all interfaces. Skewed representation is applied for illustrative purposes only.

The second 2-skip (or full-skip) PC method is using two angle PC UT probes, positioned inside the PT at angle to the tube surface: see schematics of this circumferential PC (CPC) technique in Fig. 6-1b. One of these probes transmits and the other receives shear UT wave propagating at angle within the tube wall and reflected from all interfaces.

Using “Imagine-3D” software package, we performed preliminary computer simulations of various UT techniques, demonstrating abilities to detect and measure the oxide layers of different thicknesses in the PE mode and particularly in the PC mode of operation.

Fig. 6-2 below shows example of schematics and computer simulated A-scans for CPC probes, demonstrating abilities to detect and measure the oxide layers.

Obtained results show that ~20μm oxide layer thicknesses determines, probably, the upper theoretical limit of sensitivity in the CPC technique (two reflected pulses, shown on A-scan in Fig. 6-2d, almost coincide). Of course, any computer simulation demonstrates only the potential abilities of the technique. In the real life, there are always many other factors affecting the inspection outcome. For example, the oxide layer non-uniformity and sharp
edges, acoustic parameters of oxide, UT beam divergence, probe position/orientation and water path, tube wall thickness, presence of flaws or scrape marks, and others. As result, the problem of oxide layer thickness measurement significantly complicates.

Figure 6-2. Computer simulation demonstrating CPC schematics and ability to detect and measure the oxide layers of different thicknesses - obtained computer simulated A-scan for: a - 200µm thick layer, b - 100µm thick layer, c - 50µm thick layer, d - 20µm thick layer. Pulse A - shear wave pulse reflected from OD of oxide layer, pulse B - shear wave pulse passed via oxide and reflected from ID of oxide layer.
There are a few possible UT techniques and probes, which in principle can be used for oxide layer thickness measurements:

1. The PE method, using NB longitudinal UT wave, is the simplest one, but it cannot be used to measure thickness of very thin oxide layers (about ~20μm). Moreover, since this method is based on the UT pulse reflection at the interface oxide/PT, depending on the difference between longitudinal acoustic impedances of PT and oxide layer (see Table 6-1), it will work only if this difference is large enough. If such difference is small, this technique will not work.

2. The other option is to use a NB probe, which generates not only the primary longitudinal wave, but also the secondary mode-converted shear wave, which has twice shorter wavelength. This shear wave provides much higher sensitivity, resolution and measurement accuracy at the detection and characterization of various inhomogeneities.

3. Probably, the best option is to apply PC technique, using angle shear UT wave. This method can be used to measure very thin oxide layers (about ~20μm), because it has better resolution, sensitivity, signal-to-noise ratio, and measurement accuracy due to the following reasons:
   - When UT shear wave propagates at angle within the oxide layer, it passes a greater distance, and therefore the received pulse will come later, and it will be easier to resolve it.
   - PC method always has a higher signal-to-noise ratio, and therefore the received pulse will be easier to resolve.

4. Different parameters of the received UT pulse can be used for analysis: amplitude, time-of-flight, and spectrum.

5. Different directions of UT wave propagation, circumferential and/or axial, can be used for investigation.

6. Probe parameters (frequency, focal length, diameter, incident angle, and water path) should be optimized for the task.

7. Pulser-receiver settings (excitation pulse shape, duration and amplitude, filtering and averaging), digitizer card rate, and others parameters should be optimized for the task.

8. The spectral analysis of the received pulse can be also applied to the issue of oxide layer detection and sizing. Spectral analysis is based on a simple reasoning. If two UT pulses are close to each other, overlap, cannot be clearly distinguished in the time domain, and form just one pulse with a complex shape, then its spectrum (i.e. frequency domain) can be used in an attempt to characterize the shape of this pulse.

Emphasize that any UT method of oxide layer thickness measurements can be implemented only if both surfaces of PT are more or less smooth. If surface roughness is significant, then oxide layers whose thickness is smaller than surface roughness, cannot be measured. Typical average surface roughness of PT (both ID and OD) is about $R_a \approx 1-5\mu m$. It means that oxide layers thinner than ~10μm cannot be reliably and accurately measured.

Moreover, the received UT signals depend of course on many other various parameters related to the PT material microstructure (e.g. grain size, morphology, imperfections, inhomogeneities, inclusions, pores, cracks, flaws, scratches, etc.). Emphasize that this acoustic “structure noise” is inherent to the PT, particularly being in service for many years, and can never be completely suppressed. So the challenge is to distinguish weak indications, related to oxide layer, on the background of acoustic structure noise.
As result of our experimentations, we determined three most efficient UT methods, able to detect and size various flaws covered by oxide and also detect oxide layer on the PT ID and measure its thickness. Emphasize that all three methods are based on standard UT techniques, routinely used for flaw detection and sizing in CIGAR/ANDE/BRIMS systems.

### 7 NB technique using 20MHz ID focused probe

Good results, providing clear detection, characterization and sizing of axial ID V-notches with different depths and covered by oxide layers with various thicknesses (as received, about ~30µm, about ~100µm, and about ~190µm), were obtained using NB PE technique and the ID focused NB 20MHz transducer with focal length FL=10mm and water path WP=10mm. Various images, depicting axial ID V-notches 0.5mm deep are shown in Fig. 7-1. Similar images were obtained on V-notches 0.25mm deep with different oxide layers.

Figure 7-1. 2D circumferential B-scans and respective 3D time-of-flight images derived from 3D B-scans: a - V-notch 0.5mm deep without oxide, b - V-notch 0.5mm deep with 40µm oxide, c - V-notch 0.5mm deep with 90µm oxide, d - V-notch 0.5mm deep with 190µm oxide. Depth scale in all 3D-images is 10:1.
2D B-images in Fig. 7-1 show that all notches with different oxide layers can be reliably detected, characterized and measured. Probably, it happens because oxide layers are distributed rather uniformly along the PT ID surfaces. But inside V-notches the oxide distributions are not uniform, see examples in Fig. 7-2. Other metallographic images of cross-sections of PT samples with various V-notches and different oxide layers are similar to images presented in Fig. 7-2.

![Figure 7-2](image1)

Figure 7-2. Metallographic images of cross-sections of PT samples with non-uniform oxide layers (thickness is ~190µm) inside the V-notches 0.25mm deep (a) and 0.5mm deep (b).

Moreover, inside all V-notches in Fig. 7-1 one can clearly see multiple reflections from the oxide layer ID and from the PT material ID (i.e. oxide layer OD) because the ID focused probe, used for testing, is a highly focused probe, and it is focused on the oxide layer ID (i.e. at the interface oxide/water). As result, we have a spherical source of UT longitudinal waves and these waves impact on the sides of V-notch, and reflect and refract at different angles. As result, we can see inside all V-notches the respective multiple reflections from the oxide layer ID and from the PT material ID (i.e. oxide layer OD). 2D B-images (a, b, c, and d) in Fig. 7-1 clearly demonstrate that the thicker oxide layer, the more “fully” V-notches are “filled” with reflections. As result, depth of V-notches with thick and uneven oxide layers cannot be accurately measured.

However, the oxide layer is relatively uniform not only on the PT ID, but also on the bottom of wide flat-bottom notches. See Fig. 7-3, where various images depicting axial ID flat-bottom notches 0.5mm deep with various oxide layers are shown.

Similar images were obtained on flat-bottom notches 0.25mm deep with different oxide layers.
Figure 7-3. 2D circumferential B-scans and respective 3D time-of-flight images derived from 3D B-scans: a - flat-bottom notch 0.5mm deep without oxide, b - flat-bottom notch 0.5mm deep with 30µm oxide, c - flat-bottom notch 0.5mm deep with 120µm oxide, d - flat-bottom notch 0.5mm deep with 150µm oxide. Depth scale in all 3D-images is 10:1.
2D B-images in Fig. 7-3 show that all notches with different oxide layers can be reliably detected, characterized and measured. Probably, it happens because oxide layers are distributed uniformly along the PT ID surfaces and along wide flat-bottom notches. But at the edges inside the flat-bottom notches we have long multiple reflections; the thicker oxide layer, the longer these reflections are.

For clarity, metallographic images of cross-sections of PT samples with thick oxide layers (thickness is ~160µm at the PT ID surface and ~120µm at the notch bottom surface) on coupon samples with flat-bottom notches 0.25mm deep and 0.5mm deep are shown below in Fig. 7-4. One can clearly see the uneven oxide layers at the edges of both notches and even at the notch bottoms. Other metallographic images of cross-sections of PT samples with various flat-bottom notches and different oxide layers are very similar to ones presented in Fig. 7-4.

![Figure 7-4. Metallographic images of cross-sections of PT samples with thick oxide layers (thickness is ~150-160µm) on samples with flat-bottom notches 0.25mm deep (a) and 0.5mm deep (b). On such samples with relatively uniform oxide layer d_{true}=d_{measured}.](image)

Results of notch depth and width measurements, obtained using standard 20MHz NB ID focused probe, show that all different notches covered by various oxide layers can be reliably detected, characterized and sized. Measured values of received signal amplitudes and measured values of notch depths and width differ insignificantly for notches covered by oxide layers with various thicknesses. Probably it happens because oxide layers ZrO₂ are relatively uniform and have almost the same acoustic impedance as Zr-2.5%Nb. As result, flaw detectability and sizing accuracy, obtained in our experiments, practically do not depend on the oxide layer thickness. The reason is that, due to oxide layer uniformity, we were able to perform measurements only from oxide-to-oxide, as if there is no oxide at all; this case is actually presented in Fig. 7-4b, where d_{true}=d_{measured}. Respectively, the
accuracy of measurements was rather high. However, in general, as it has already been mentioned earlier, the oxide layer in PT in reactor can affect flaw detection, characterization and sizing in very different ways.

Recall that our specimens, used for testing, had pretty uniform oxide layers with relatively smooth surface. However, in order to simulate the actual oxide layers in reactor, the test specimens should have non-uniform oxide layers with comparatively rough surface.

Emphasize that, unfortunately, this method cannot be used for oxide thickness measurement on the PT ID and on the bottoms of wide bearing pad frets, because it is impossible to distinguish two UT pulses reflected from the ID of oxide layer and from the PT ID. Most probably, it occurs because oxide layers on tested samples are pretty uniform and due to very close values of longitudinal acoustic impedances of oxide and PT material.

8 NB technique using 17MHz material focused probe

Good results, able to estimate oxide layer thickness were obtained using NB PE technique and material focused NB 17MHz transducer with a stretched focal zone FZ=17-42mm and WP=21mm [7]. 2D B-images, depicting ID V-notches and flat-bottom notches, covered by oxide layers with various thicknesses (as received, about ~30µm, about ~100µm, and about ~190µm), are shown below in Fig. 8-1. Similar images were obtained on V-notches 0.5mm deep and flat-bottom notches 0.25mm and 0.5mm deep with different oxide layers.

Unfortunately, we did not get clear direct reflections from oxide layers. The reason, probably, is the closeness of acoustic impedances of PT material Zr-2.5%Nb and oxide layer ZrO$_2$. In this case, we have a very weak reflection from oxide layer, and respective responses are hard to distinguish at the level of typical acoustic noise in PT. However, despite this disadvantage, we got another rather reliable method of oxide layer detection and more-or-less accurate estimation of its thickness.

This technique is based on measurement of the duration of shear-longitudinal mode-converted response located at ~30.6µs on all B-scans presented in Fig. 8-1. Mode-converted UT pulse, located at ~30.6µs, is pretty wide and has rather complex “distorted” shape, because this shear-longitudinal UT pulse is actually consists of two different mode-converted pulses, reflected from the PT ID and oxide layer.

The obtained width of a complex-shape pulse, consisting of two close and overlapped pulses, strongly depends on the thickness and parameters of the oxide layer. At the same time, it, of course, depends on various changes in the PT surface roughness and material microstructure: e.g. grain size, material morphology, imperfections, inhomogeneities, inclusions, pores, cracks, flaws, scratches, etc.

However the acoustic “structure noise” is, in average, approximately the same everywhere in the clean areas of PT (i.e. in the areas without oxide layer). This type of average acoustic structure noise is inherent to the PT, and can never be completely suppressed. So the challenge is to distinguish weak signal variations, related to the oxide layer, on the background of response fluctuations due to acoustic structure noise.
Figure 8-1. 2D circumferential B-scans derived from 3D B-scans: a – V-notch 0.25mm deep without oxide, b - V-notch 0.25mm deep with 40µm oxide, c - V-notch 0.25mm deep with 90µm oxide, d - V-notch 0.25mm deep with 190µm oxide, e – flat-bottom notch 0.25mm with 30µm oxide, f - flat-bottom notch 0.25mm with 100µm oxide, g - flat-bottom notch 0.25mm with 160µm oxide.

We used different coupons and correlated oxide thickness, measured on metallographic images (“true” values), with width of shear-longitudinal mode-converted response located at ~30.6µs on all B-scans presented in Fig. 8-1. Measurements were also performed at different axial locations. The obtained averaged correlation curve is shown in Fig. 8-2 below.

It is obvious that this correlation is pretty good (correlation coefficient is ~0.95) and almost linear. It means that width of shear-longitudinal mode-converted response, obtained using NB material focused probe can be used for oxide layer thickness estimation.
Figure 8-2. Averaged correlation between the oxide layer “true” thicknesses, measured using metallographic images, and widths of shear-longitudinal mode-converted response. Averaging was performed using different coupons and various axial locations.

However, this method is definitely not a direct measurement technique. It has various issues and difficulties, related to the oxide layer detection and sizing. The obtained correlation is just an average and approximate one, there are always many other factors affecting the outcome of measurements: the oxide layer non-uniformity and sharp edges, UT beam divergence, probe position and orientation, variations in the tube wall thickness, presence of scrape marks, surface roughness, structure imperfections, and others. Respectively, the problem of detecting and sizing non-uniform oxide layer on the background of PT with significant microstructure noise is not simple. Moreover, the obtained results were based on the testing of “artificial” oxide layers, while the actual oxide layers in the reactor might differ from the tested ones.

Because of all these issues and difficulties, we decided to analyze the signal spectrum. In general, the UT spectroscopy is widely used for qualitative and quantitative microstructure material characterization, when spatial resolution in time domain is not sufficient. Analysis of the UT spectral features can yield quantitative correlations with material properties that are governed in turn by microstructure. If two UT pulses (reflected, in our case, from the tube material and oxide layer) are very close to each other in the time domain, overlap, cannot be clearly distinguished, and form just one pulse with a complex shape, then, probably, in order to overcome this uncertainty, its spectrum can be used to characterize the shape and width of this pulse.

The trade-off between the compaction of a function and its Fourier transform can be formalized in the form of the uncertainty principle by viewing a function and its Fourier transform as conjugate variables. Recall that term uncertainty comes here from the fact that a function and its Fourier transform cannot both be sharply concentrated (localized) at once. This result is a generalization of the fact, well known, for example, in quantum mechanics (pair the momentum and position and/or pair the energy and time are Fourier transform pairs, and cannot be both accurately measured) and in communication theory (a signal cannot be sharply limited in both, time and band domains, simultaneously).
The obtained spectrum of a complex-shape pulse, consisting of two close and overlapped pulses, strongly depends on the thickness and parameters of the oxide layer. At the same time, this spectrum, of course, depends on the various changes in the surface roughness and material microstructure of the PT. It looks like that spectral analysis allows distinguishing between responses, related to the oxide layer, from responses, related to the surface roughness and microstructure imperfections, i.e. such spectral analysis provides some type of “filtration from average structure noise”.

The obtained results demonstrate that responses from different tested PT samples with the same oxide layers have in average rather similar time-domain images and spectra. Consistency and robustness of these results, i.e. images in time and frequency domains, obtained on different tested PT coupons, demonstrate that used UT methods and equipment, employed for PT testing, provide rather reliable results. In general, one can assert with high probability that images in the time-domain and their spectra can be used to characterize the average oxide layer thickness.

Using a few PT coupons with various thicknesses of oxide layers, we established a correlation between averaged oxide layer thicknesses measured metallographically (“true” values) and averaged widths of spectra at -6dB level of shear-longitudinal mode-converted responses, obtained using standard NB material focused probe. We analyzed spectra of responses, shown in Fig. 8-1, and obtained averaged correlation curve. Measurements and subsequent averaging were performed using various coupons and also different axial locations on each coupon.

The obtained averaged correlation curve oxide “true” thicknesses vs. averaged width of spectrum at -6dB level of mode-converted response, is presented below in Fig. 8-3.

![Figure 8-3. Averaged correlation between the oxide layer “true” thicknesses, measured using metallographic images, and widths of spectrum of shear-longitudinal mode-converted response. Averaging was performed using different coupons and various axial locations.](image)
It is obvious that this correlation is pretty good (correlation coefficient is ~0.85) and almost linear. It means that width of spectrum of shear-longitudinal mode-converted response, obtained applying NB material focused probe can be used for oxide layer thickness estimation. Using the obtained correlation curves, presented in Figs. 8-2 and 8-3, it is easy to get the respective trendlines and extrapolate them to any required range of oxide thicknesses. Obtained correlation plots and respective trend-lines can be used as calibration curves (at least, in the first approximation) for actual measurements of unknown oxide layers.

It is well known that the shorter pulse, the wider its spectrum is. On the other hand, the wide pulse should have rather complex shape, because it consists of two different pulses reflected from the PT ID and oxide layer. As result, its spectrum also should be wide. These two factors generically work in the opposite directions. However, we have a very weak reflection of longitudinal waves from the interface oxide/PT, because we do not see any clear reflection from this interface using NB ID focused probe (see respective A-scans in Figs. 7-1 and 7-3) and NB material focused probe (see respective A-scans in Fig. 8-1). It means the longitudinal acoustic impedances of the oxide and PT material are almost equal; therefore the wide pulse (containing in principle two different pulses) is not distorted, has rather simple shape, and consists of a few “clean” cycles (see again A-scans in Figs. 7-1, 7-3 and 8-1). Most probably, it occurs because for NB technique, where we are using material focused probe, the contribution of longitudinal waves is significant (because we are analyzing the mode-converted shear-longitudinal wave), and therefore the 1st factor plays the main role, i.e. the shorter pulse, the wider its spectrum is.

If we could use only shear waves, then wide pulse would have rather complex “distorted” shape, because it should consist of two different shear wave pulses, reflected from the PT ID and oxide layer. As result, its spectrum should be wider than spectrum of a “clean” pulse. It means that when only shear waves are used, the 2nd factor plays the main role, i.e. pulse with complex “distorted” shape, consisting of two different pulses reflected from the oxide layer and PT ID, has a wider spectrum. The results, presented in the next section for CPC technique, using only shear waves, confirm this idea.

Emphasize that, unfortunately, described method cannot be used to detect, characterize and size different ID notches covered by various oxide layers, because NB material focused probe is not sensitive enough for this task. This probe has been developed specifically for detection and sizing of material and OD flaws. Applying this probe for oxide layer thickness estimation, we are just using one of its advantages – ability to generate strong shear-longitudinal mode-converted response. This feature was so far a neglected weapon in the arsenal of UT tools; now it is time to use it. Since mode-converted response contains a shear wave with a small speed of propagation (and respectively, small wavelength), it is more sensitive to oxide layer detection and provides more accurate sizing.

9 CPC technique

Good results, providing clear detection, characterization and sizing of axial ID V-notches with different depths and covered by oxide layers with various thicknesses (as received, about ~30µm, about ~100µm, and about ~190µm), were obtained using CPC technique.
with UT probes having frequency $f=15$MHz, $FL=50$mm, incident angles $25^\circ$, and $WP=21$mm. Various images, depicting ID axial V-notches 0.5mm deep are shown in Fig. 9-1. Similar images were obtained on V-notches 0.25mm deep with different oxide layers.

Figure 9-1. 2D circumferential B-scans and respective 3D time-of-flight images derived from 3D B-scans: a - V-notch 0.5mm deep without oxide, b - V-notch 0.5mm deep with 40µm oxide, c - V-notch 0.5mm deep with 90µm oxide, d - V-notch 0.5mm deep with 190µm oxide. Depth scale in all 3D-images is 10:1.
Various images depicting axial ID flat-bottom notches 0.25mm deep are shown in Fig. 9-2. Similar images were obtained on flat-bottom notches 0.5mm deep with different oxides.

Figure 9-2. 2D circumferential B-scans and respective 3D time-of-flight images derived from 3D B-scans: a - flat-bottom notch 0.25mm deep without oxide, b - flat-bottom notch 0.25mm deep with 30µm oxide, c - flat-bottom notch 0.25mm deep with 120µm oxide, d - flat-bottom notch 0.25mm deep with 150µm oxide. Depth scale in all 3D-images is 10:1.
2D B-images in Figs. 9-1 and 9-2 show that all notches with different oxide layers can be reliably detected, characterized and measured. Recall that oxide layers are distributed rather uniformly along the PT ID surfaces and along wide bottoms of flat-bottom notches.

Results of notch depth and width measurements, obtained using CPC technique, show that measured values of received signal amplitudes and measured values of notch depths and width differ insignificantly for notches covered by oxide layers with various thicknesses.

Probably it happens because oxide layers ZrO$_2$ are pretty uniform and have almost the same acoustic impedance as Zr-2.5%Nb. As result, flaw detectability and sizing accuracy, obtained in our experiments, practically do not depend on the oxide layer thickness. The reason is that, due to oxide layer uniformity, we were able to perform measurements only from oxide-to-oxide, as if there is no oxide at all. Respectively, the accuracy of measurements was pretty high. This case is illustrated in Fig. 7-4b, where $d_{\text{true}} = d_{\text{measured}}$; of course one should take into account that in the PC mode all measurements are performed in the direction from the OD to ID. However, in general, as it has already been mentioned earlier, the oxide layer in PT in reactor can affect flaw detection, characterization and sizing in very different ways.

Recall that our specimens, used for testing, had pretty uniform oxide layers with relatively smooth surface. However, in order to simulate the actual oxide layers in reactor, the test specimens should have non-uniform oxide layers with comparatively rough surface.

Unfortunately, using 2D B-images, shown in Figs. 9-1 and 9-2 and depicting axial ID V-notch and flat-bottom notches, covered by oxide layers with various thicknesses, we did not get clear direct reflections from the oxide layers. The reason, probably, is the closeness of shear acoustic impedances of PT material Zr-2.5%Nb and oxide layer ZrO$_2$. In this case, we have only a weak reflection from oxide layer, and respective responses are hard to distinguish at the level of typical acoustic noise in PT.

However, despite this disadvantage, we got another comparatively reliable method of oxide layer detection and more-or-less accurate estimation of its thickness. This technique is based on measurement of the duration of shear wave CPC response on all B-scans presented in Figs. 9-1 and 9-2. CPC pulse is pretty wide and has rather complex “distorted” shape, because this shear wave UT pulse is actually consists of two different shear wave pulses, reflected from the PT ID and oxide layer. The obtained width of a complex-shape shear wave pulse, consisting of two close and overlapped shear wave pulses, strongly depends on the thickness and parameters of the oxide layer.

We used different coupons and correlated the oxide layer thicknesses, measured on the metallographic images (“true” values), with widths of CPC responses on all B-scans presented in Figs. 9-1 and 9-2. Measurements were also performed at different axial locations. The obtained averaged correlation curve is presented in Fig. 9-3 below.

It is obvious that this correlation is pretty good (correlation coefficient is ~0.95) and almost linear. It means that width of shear wave CPC response can be used for oxide layer thickness estimation.
Figure 9-3. Averaged correlation between the oxide layer “true” thicknesses, measured using metallographic images, and widths of shear wave CPC response. Averaging was performed using different coupons and various axial locations.

However, this method is definitely not a direct measurement technique. It has various issues and difficulties, related to the oxide layer detection and sizing and described in details in previous section #8. Because of these issues and difficulties, we decided to analyze spectra of shear wave CPC responses.

Using a few PT coupons with various thicknesses of oxide layers, we established a correlation between averaged oxide layer thicknesses measured metallographically (“true” values) and averaged widths of spectra at -6dB level of shear wave CPC responses. We analyzed spectra of responses, shown on A-scans in Figs. 9-1 and 9-2, and obtained averaged correlation curve. Measurements and subsequent averaging were performed using various coupons and also different axial locations on each coupon.

The obtained averaged correlation curve - oxide “true” thicknesses vs. averaged widths of spectra at -6dB level of shear wave CPC responses - is presented in Fig. 9-4.

It is obvious that this correlation is pretty good (correlation coefficient is ~0.9) and almost linear. It means that width of spectrum of shear wave CPC response can be used for oxide layer thickness estimation.

Using the obtained correlation curves, presented in Figs. 9-3 and 9-4, it is easy to get the respective trendlines and extrapolate them to any required range of oxide thicknesses. Obtained correlation plots and respective trend-lines can be used as calibration curves (at least, in the first approximation) for actual measurements of unknown oxide layers.
The CPC method can be used simultaneously for oxide layer thickness estimation and also for detection, characterization and sizing of different ID flaws covered by various oxide layers, because shear wave angle CPC probes are sensitive enough for both tasks.

Despite all approximations, disadvantages, inaccuracies and limitations, mentioned above, the obtained results are encouraging, and reasons for limitations are, most probably, correctly understood. We can conclude that performed investigation is a promising start; it definitely demonstrates that UT technology is an efficient technique for detection, characterization and sizing of flaws covered by oxide, and for oxide layer thickness measurements. Generically, the UT measurement of oxide layer thickness is based on the technology, well-established and proven in different areas and currently adapted for this particular application. In addition emphasize that none of the currently available NDE methods for oxide layer detection and sizing is better. Thus in general, the UT techniques and probes, described above and optimized for the task (within our abilities) in the laboratory conditions (NB high-frequency ID focused probe, CPC angle method using shear waves, NB material focused probe using mode-converted shear-longitudinal response, and spectral analysis of responses) allow detecting and measuring various oxide layers and also sizing different ID flaws covered by oxide layers. However, remind again that actual oxide layers in PT in reactor are not uniform and homogeneous and actual oxides might have different composition and acoustic parameters in comparison with “artificial” oxide layers, grown up in the lab conditions. All these factors will definitely complicate tasks of oxide layer detection and sizing and also characterization and sizing of different ID flaws covered by oxide.

10. Conclusions

1. Oxide layer on the PT ID masks flaws and complicates their detection, characterization and sizing; entails degradation of the material properties and brittle microstructure; and
leads to many other different issues. That is why oxide layer detection and sizing is an important problem.

2. The ability to perform UT examination of flaw characterization and sizing in PT with oxide layer and measurements of the oxide layer thickness in any required local area of PT or by mapping the whole tube is very attractive, because the UT measurements are routinely used for PT inspection; they are rapid, sensitive, accurate, and reliable. UT data will be obtained in real time in various convenient formats and can be easily analyzed; performed scans can be saved and accessed any time for further analysis.

3. Calibrated computer-based UT immersion scanning system was employed for performed lab experiments. This UT system includes the Winspect™ data acquisition software, a SONIX STR-8100 digitizer card, and a UTEX UT-340 pulser-receiver. The tests were performed using various UT techniques and probes with different center frequencies and focal lengths.

4. The laboratory tests were conducted on the PT coupons made contained ID axial flaws of different shapes and dimensions. These coupons contained oxide layers with various thicknesses: as received, about 30-40µm, about 90-110µm, and about 160-190µm. All our specimens, used for testing, had pretty uniform oxide layers with relatively smooth surface. However, in order to simulate the actual oxide layers in reactor, the test specimens should have non-uniform oxide layers with comparatively rough surface.

5. Performed computer simulations demonstrated that oxide layer thickness about 20µm determines, probably, the upper theoretical limit of sensitivity.

6. Experimental investigation was performed using various PE and PC circumferential 2D-scans and 3D-scans, applying NB and angle UT transducers with different parameters. Longitudinal and shear waves propagating in different directions were used for testing; signal processing methods (filtering, averaging and spectral analysis) were also employed for investigation. Different images, including 3D images, were obtained.

7. Obtained results clearly show that various ID flaws in all coupons can be reliably detected, characterized, and sized.

8. Measured values of received signal amplitudes and notch dimensions differ insignificantly for notches covered by oxide layers with various thicknesses. Probably it happens because oxide layers are uniform. Respectively, we were able to perform measurements only from oxide-to-oxide, as if there is no oxide at all. As result, flaw detectability and sizing accuracy, obtained in our experiments, practically do not depend on the oxide layer thickness.

9. Unfortunately, we did not get clear direct reflections from oxide layers. Probably, the reason is the closeness of acoustic impedances of PT material Zr-2.5%Nb and oxide layer ZrO₂. In such a case, there are only weak reflections from oxide layers, and respective responses are hard to distinguish at the level of typical acoustic noise in PT.

10. However, despite this disadvantage, we managed to determine three UT techniques able to detect oxide layer and estimate its thickness.

11. Obtained results show that, using NB ID focused 20MHz probe, the oxide layer can be detected because all V-notches and edges of flat-bottom notches are “filled” with multiple UT reflections from the oxide layer ID and PT material ID (i.e. oxide layer OD). The thicker oxide layer, the more “fully” V-notches and sides of flat-bottom notches are “filled” with UT reflections. However, unfortunately, this method cannot be used for oxide thickness measurement.
12. Measurement results show that width of shear-longitudinal mode-converted response and width of its spectrum, obtained using NB material focused probe, can be used for oxide layer thickness estimation. Respective correlation curves have large correlation coefficients $R \approx 0.85$-$0.95$, and their trend-lines can probably be used (at least, in the first approximation) as calibration curves. However, this method cannot be used to detect, characterize and size different ID notches covered by various oxide layers.

13. Obtained results show that all different notches covered by various oxide layers can be reliably detected, characterized and sized using CPC technique. Moreover, this CPC technique is also able to provide measurement of oxide layer thickness. The width of shear wave CPC response and width of its spectrum can be used for oxide layer thickness estimation. Obtained correlation curves have large correlation coefficients $R \approx 0.85$-$0.95$, and their trend-lines can probably be used (at least, in the first approximation) as calibration curves.

14. Despite all issues, mentioned above, the obtained results are encouraging, and reasons for limitations are, most probably, correctly understood. We can conclude that performed investigation is a promising start; it definitely demonstrates that UT technology is an efficient technique for detection, characterization and sizing of flaws covered by oxide, and also UT can be used for oxide layer thickness measurements.

11 References


