NONDESTRUCTIVE TESTING OF THE LOW-LEVEL RADIOACTIVE WASTE DRUMS FOR UNI-AXIAL COMPRESSIVE STRENGTH AND FREE LIQUID CONTENT

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ABSTRACT

This paper summarizes the nondestructive test to determine the uni-axial compressive strength and free water content of solidified low level radioactive waste. The uni-axial compressive strength is determined by ultrasonic wave propagation speed, and the results are compared with those of compressive tests. Three methods of detecting the surface free water by ultrasonic testing are established, the ultrasonic wave speed, wave form and pulse height are used to determine the existence and amount of the surface free liquid. Possible difficulties are discussed.

INTRODUCTION

For the purpose of reduction of the radiation dose and pollution to the persons and natural environment, the radioactive waste produced from nuclear applications must be strictly and safely processed. Civilian nuclear power plant can produce a lot of low level radioactive waste. These can be classified into three categories, gaseous, solid and liquid radwaste. The gaseous radwaste is effluented to the air under conditional release through the off-gas treatment system. The solid radwaste is packaged in the carbon steel drum, then the low level waste drums are transported to the storage place or the final disposal facility. The low level liquid radwaste must be solidified first to ensure that they are in a stable state, then followed the same treatment as the solid radwaste. Cement is one of the candidate materials under consideration for the solidification of liquid waste, because of its strength, durability, abundant availability and relatively low cost. So, the power plants usually adopt the Portland cement to solidify the waste.

The problem faced by operators is to assure that low level waste shipments meet transportation regulations and burial site requirements. The requirements of uni-axial compressive strength must have at least 15 Kgw/cm^2 for land disposal, and at least 150 Kgw/cm^2 *for sea dumping* (1). The United States of Federal Regulations. 10 CFR Part 61 (2), mandates that free-standing liquid not exceed one percent of the volume of the radioactive waste in a container, and not exceed 1/2 percent of the volume in waste processed to a stable form (3). Methodology is required to verify the uni-axial compressive strength of cemented solid and reliably and accurately determine the existence and amount of free liquid on the surfaces or internally trapped within solidified low-level waste. Current methods of visual inspection or use of a process control program have not been consistently successful. It increases the exposure dose to workers, and needs much time to inspect the waste drums. As a result it is required to develop some non-destructive testing methods to do this.

The prospective method of inspection for uni-axial compressive strength is schmidt hammer, ultrasonic wave propagation speed and vibration analysis method (4,5). The method using the ultrasonic wave propagation speed is the most practical at present (6). This method makes use of the correlation between the speed of ultrasonic wave propagation within the cemented solid and uni-axial compressive strength (6,7). The NDT techniques have been used in detection of radioactive waste canister for free liquid in several advanced countries. Under the regulations of environmental protection in the United States, X-ray radiography has been used to detect free water and the ultrasonic testing has been applied to assure the durability of waste drum. The safety of transportation is the most important factor to be considered in Japan, therefore, the infrared thermal imagine and gamma ray radiography have been developed to identify free water above the cement solid. In west Germany, the X-ray computerized tomography (CT) is being studied in detail and the quantity of free water may be estimated. Although functions of CT are impressive for NDT, the field operation may be incompetent due to the high cost in some circumstances. Up-to-date, most of NDT methods are implemented for qualitative analysis only. This paper summarizes the method of nondestructive testing to detect the free water content and uni-axial compressive strength of solidified low level radwaste.

EXPERIMENTAL WORK

The experiment includes two main parts, they are to verify the compressive strength of cemented solid and detection of surface free water by ultrasonic testing.

Test of Compressive Strength of Cemented Solid

This research simulates the low level liquid waste produced in the current BWR power plant. During this study, only simulation of the chemical composition is considered. The shape of test specimens are cubic and cylindrical, with dimensions of 5cm • 5cm • 5cm (8) and 4.4 cm diameter and 8.8 cm height (9) respectively. Portland cement type 2 and 5 were applied to solidify the liquid waste. The range of the water/cement ratios is from 0.52 to 1.0. The water/cement ratio is the ratio of the volume(c.c) of the liquid to 100g of the cement. The casted specimens were placed in air for 24 hours, then the specimens were closed. The modelled specimens were cured in air for different testing time.

The testing procedure includes two steps. First, the ultrasonic velocity was measured versus curing time to assume stability of ultrasonic propagation. Next when the specimens curing up to 28 days, they were demoulded for testing. The specimens were first measured the velocity of ultrasonic propagation, then the compressive strength of the specimens were determined by the destructive compression test. This procedure must be done quickly to prevent the moisture escaping from the specimens surface which reduces the effect of the moisture content. The applied frequency of the ultrasonic transducer is 0.5 MHz.

Test of Surface Free Water

Three methods of application of ultrasonic testing to detect surface free water are developed in this report.

Method (1): The velocity of propagation of ultrasonic wave in a material is one of properties of the material. The method is to utilize the UT velocity and the time that UT needs to penetrate the surface free water layer to estimate the amount of the surface free water.

Figure 1 illustrates the set-up of the experiment. The transducer emits and receives UT wave from the bottom of the specimen, the screen of the oscilloscope will display such a wave form as that at the right-bottom corner of Fig. 1, the point 1 is the position of initial pulse, the point 2 and the point 3 are separately the first reflected wave from water/gel-interface and water/air-interface. The thickness and the amount of surface free water can be estimated from the wave velocity and the interval between the two reflected waves.

The specimens are the same as the cubic ones in part 1.

Method (2): This method estimates the amount of surface free water by means of detecting the positions of interfaces of surface free water with gel and air respectively. The equipments and set-up are shown in Fig. 2. In the Fig. 2, the point 1 and point 2 are separately the first and the second reflected waves from the mold side in the surface free water, and point 2 is the second reflected wave. The contacting area of the transducer and the water layer will change as the transducer scans along the mold side, so the different intensities of pulse of emission and receiving respond to different scanning positions. Physically the contact area reflects the relative position of the water layer with respect to a referenced position. Two measurements of contact area are used to determine height of the water layer. As the transducer scans along the mold side, the relative positions of transducer and water layer and the height of the pulse of the first reflected wave are recorded, a relation curve is established. Afterwards if a pulse-height is measured, a corresponding relative position or contacting area can be used to identify the interfaces of water layer by this method.

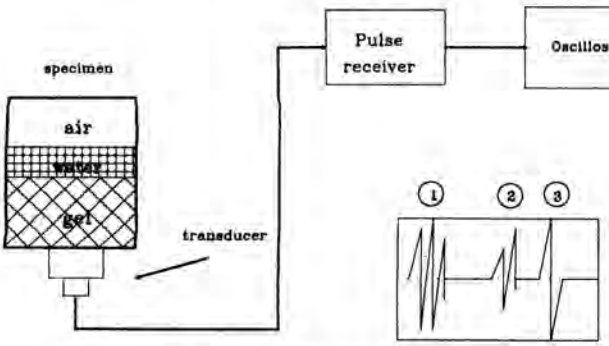

Fig. 1. Setup of UT system of method (1).

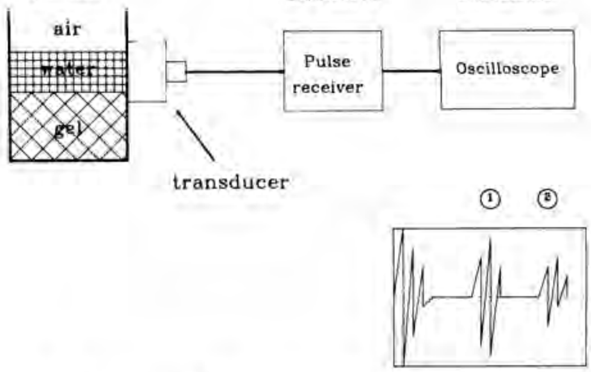

Fig. 2. Setup of UT system of method (2) and (3).

Method (3): This method utilizes the relation curve of the thickness of water layer and the maximum height of the pulse of the first reflected wave to measure the amount of surface free water. Figure 2 shows the equipments and set-up of the system.

RESULTS AND DISCUSSION

Test of Uni-axial Compressive Strength

The ultrasonic velocity increases very rapidly in the first few hours while strength of solidified radwaste develops much more slowly. For concrete, at seven days, approximately 100% of 28 days pulse velocity is attained while its strength is between 60%-80% (11). But in this case, the testing material is cement solid rather than concrete. The ultrasonic propagation velocity versus cure time for Portland cement are shown in Figs. 3 and 4. The velocity increases very rapidly in the first few days as same as the above described, followed by a oscillation period during 5-8 days. After this period, the curves become more and more flatter to approach a saturated value.

There is a empirical correlation between compressive strength and the speed of ultrasonic propagation in concrete. This correlation can generally be shown by the following formula (6).

$$E = a \cdot F^{b}$$

Where

E = dynamic elastic modules
F = compressive strength
a, b = constants

E can also be expressed by the following formula :

$$E = \rho \cdot V \cdot (1+\nu) \cdot (1-2\nu)/g \cdot (1-\nu)$$

Where

ρ = density

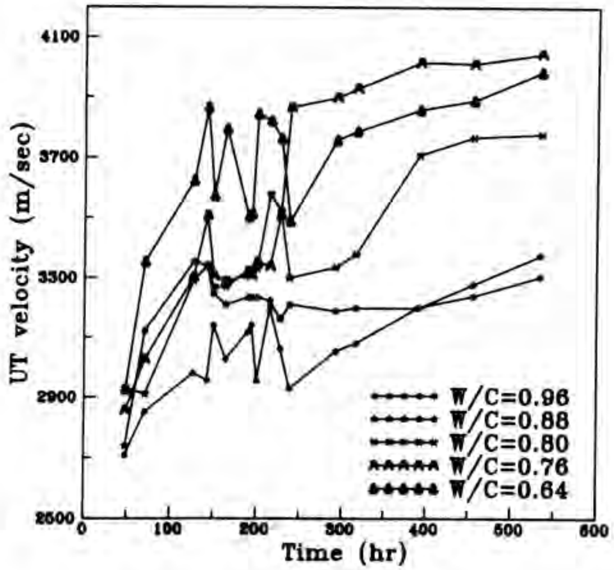

Fig. 3. The relationship between UT velocity and curing time for type 2 Portland cement.

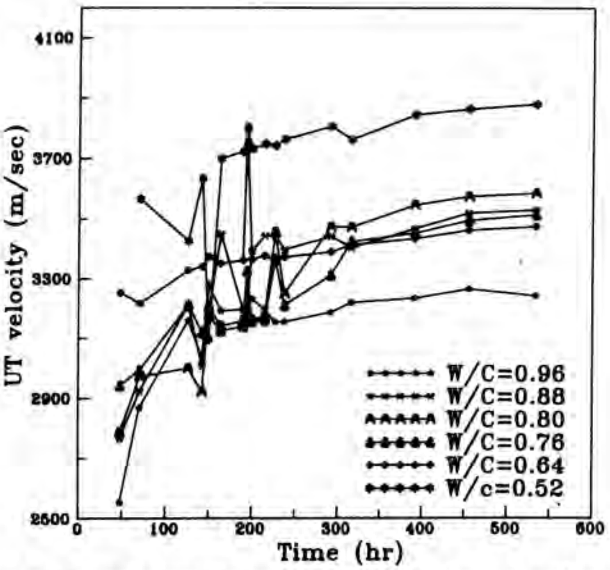

Fig. 4. The relationship between UT velocity and curing time for type 5 Portland cement.

V = speed of ultrasonic wave propagation
g = acceleration of gravity
ν = poison ratio

Let, $\alpha = \sqrt{\frac{(1-\nu)}{(1+\nu)(1-2\nu)}}$ then α varies 1.11 to 1.14 for concrete (12). This formula also suggests that the relation between the ultrasonic wave velocity and compressive strength is a power law. The results are shown in the Figs. 5 and 6. The uni-axial compressive strength of the cemented solid can be estimated by using this speed and the correction curve for uni-axial compressive strength.

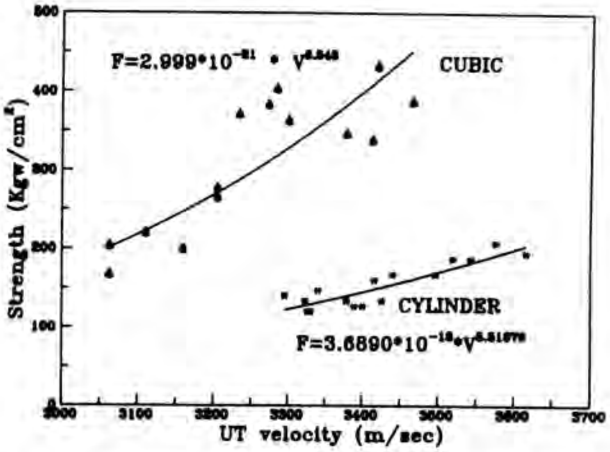

Fig. 5. The correction curve between uni-axial compressive strength and UT velocity for type 2 Portland cement.

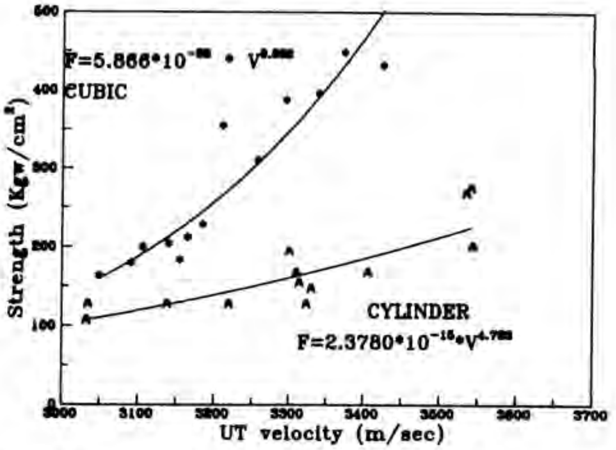

Fig. 6. The correction curve between uni-axial compressive strength and UT velocity for type 2 Portland cement.

There are some problems must be considered for the application of the ultrasonic test in the field. In the experiment the ultrasonic transducer contacted the cemented solid surface directly, but for real waste drums, the ultrasonic transducer will contact the carbon steel sheet. There generally exists an air gap between the drum sheet and the cemented solid. Because the ultrasonic wave will be reflected in the interface of medium and air. So, the ultrasonic wave can't reach the solid. In this experiment, because the size of the specimens are the same as that of ultrasonic transducer, the transducer can be treated as plane source. But for 55 gallons drum, the transducer must be treated as point source. The behaviors of ultrasonic wave propagation of point source need further study. The existence of cracks,etc,inside the solid can influence the speed of propagation. This effect can be reduced by means of measurement at several points.

Test of Surface Free Water

Method (1): Figure 7 is the relation curve of cure time and the volume of surface free water which was obtained by UT at different water/cement ratios of type 2 Portland cement. The quantity of surface free water decreases with the increase of cure time. The decreasing amount of free water served as the water that the hydration reaction of cement required. The larger water/ cement ratio is, the more amount of surface free water is. Particularly, the amount of water/cement ratio 1.0 is much more than the others, about twice larger than the ratio 0.92.

Figure 8 illustrates the relation curve of cure time and the volume of surface free water which was obtained by a straight rule at *different* water/cement ratios of type 2 Portland cement. The tendencies of curves are similar to cures of Fig. 7. Figure 9 shows the comparison of volume of surface free water measured from the UT and a rule-estimate at water/cement ratio 1.0 and 0.84. The results of UT quite match those of a rule-estimate except the results at water/cement ratio 1.0 differs more. However the mean relative errors of ratio 1.0 and 0.84 are separately 13.6% and 30.4%.

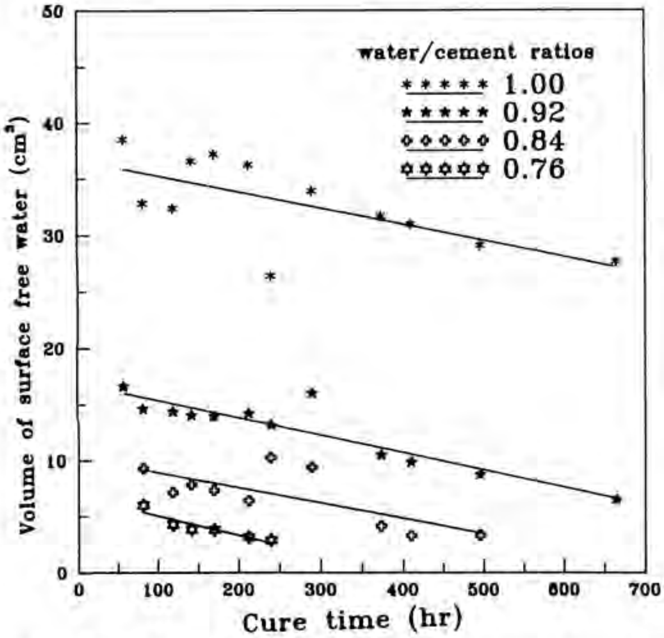

Fig. 7. The volume of surface free water measured from UT method versus the cure time of type 2 Portland cement.

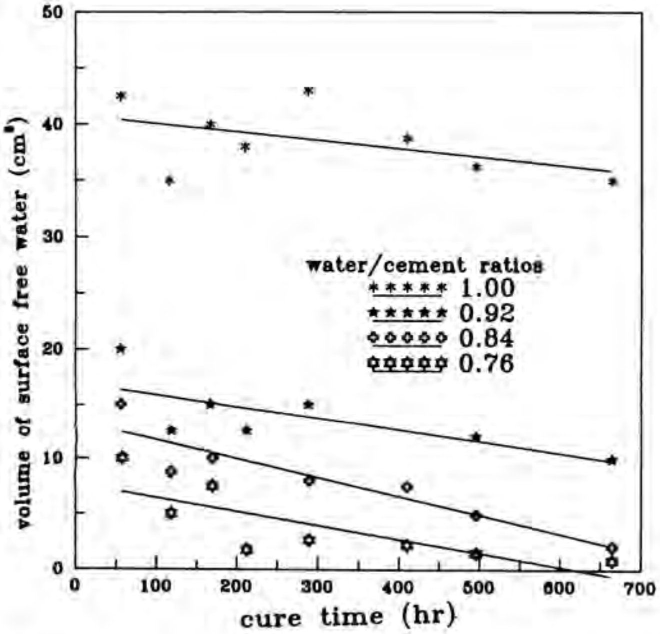

Fig. 8. The volume of surface free water measured from a rule-estimate versus the cure time of type 2 Portland cement.

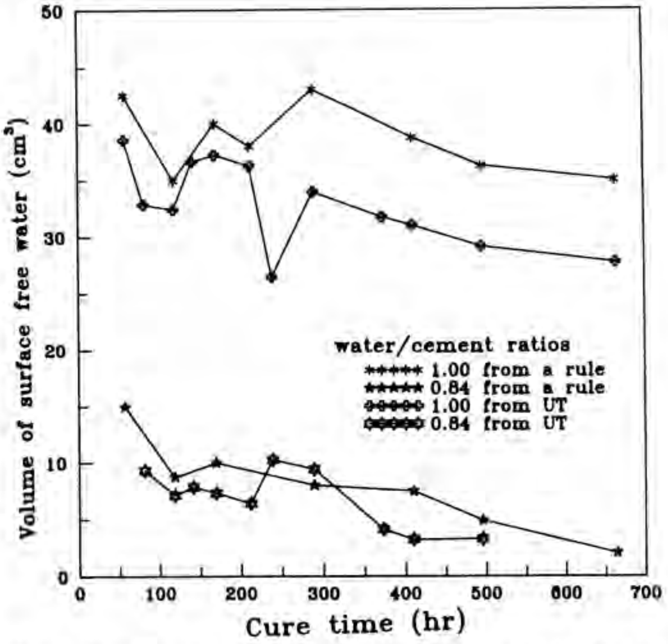

Fig. 9. The comparison of the volume of surface free water measured from UT and a rule-estimate versus cure time of type 2 Portland cement.

The curves of linear fit in Fig. 7 and Fig. 8 have the approximate slopes, their hydration reactions have the approximate absorption rate of water. In addition, the specimens with water/cement ratios lower than 0.68 will have no surface free water after 57hr of cure time; the specimens of w/c ratio 0.72 will also have no surface free water after 57 ~ 118hr of cure time.

Method (2): Figure 10 shows the measurements of the pulse height of the first reflected wave versus the contacting area between a transducer and water layer. The pulse height increases with contacting area, and the variations of pulse-height are smaller for larger or smaller contacting area; but the variation is more intense at the middle region. The reflected waves for the contacting-area above 2.7 ~ 3.0 square centimeter or the thickness of water layer above 1 centimeter are identified. The maximum of the pulse-heights is about 800 ~ 900 mV.

Figure 11 illustrates that the 1.0 MHz transducer is also studied in the method. The tendencies of these curves are essentially similar to Fig. 10. The main difference is that the pulse-height is almost triple larger than that in Fig. 10. Water is a homogeneous media, so the factor of attenuation is negligible (13). It is not clear why does the 0.5 MHz transducer emit stronger stress waves but receives much smaller pulse-height response than that of 1.0 MHz transducer. More research is needed in this area. The existence of water layer just above 0.6 cm thickness can be detected with 1.0 MHz transducer. However 1.0 cm thickness water layer can be detected with a 0.5 MHz transducer. Transducer with higher frequency reflects higher resolution of detecting capability for the free surface water layer.

Figure 12 makes a comparison of different thickness of water layer. The maximum pulse-heights of 5.0 cm and 3.0 cm

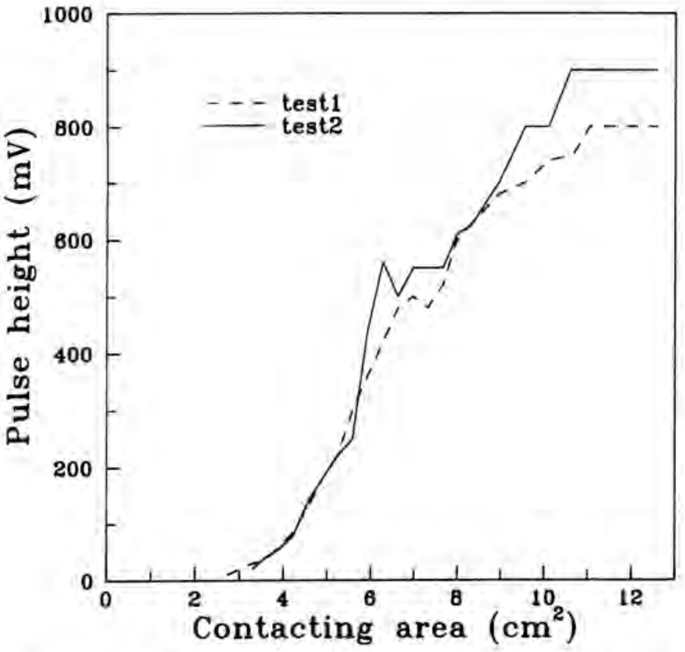

Fig. 10. The relation between pulse height and contacting area at water/gel-interace. 0.5 MHz probe, total area is 12.57 cm^2; ATTEN. = 0.0 db; water thickness: 5.2 cm

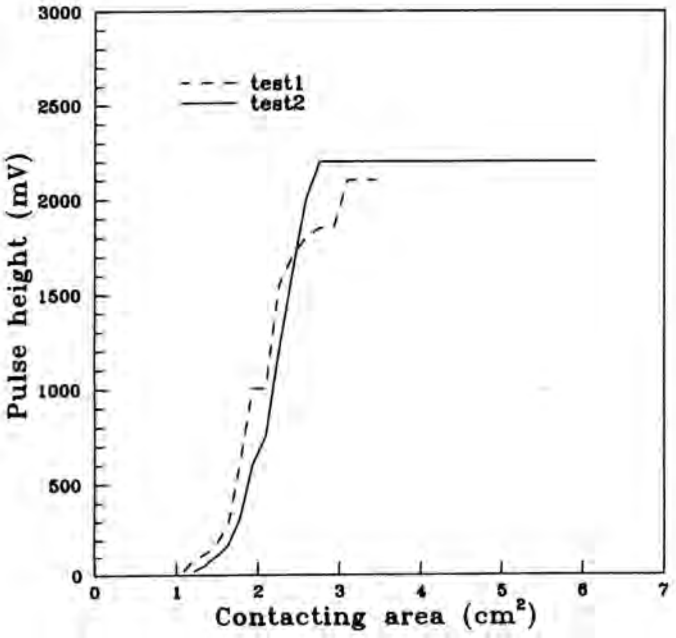

Fig. 11. The relation between pulse height and contacting area at water/air-interface. 1.0 mHz probe, total area is 6.16 cm^2, water thickness: 5.2 cm.

water layer are the same, but pulse height of 1.0 cm is smaller than the other two. The above phenomena suggest that there is a critical thickness between the water layers of 1.0 cm and 3.0 cm thickness. The formula of the divergent angle of ultrasonic wave is (14):

$$\sin\theta = 1.22\lambda/D$$

$$\text{or } \sin\theta = 1.22\, C/fD$$

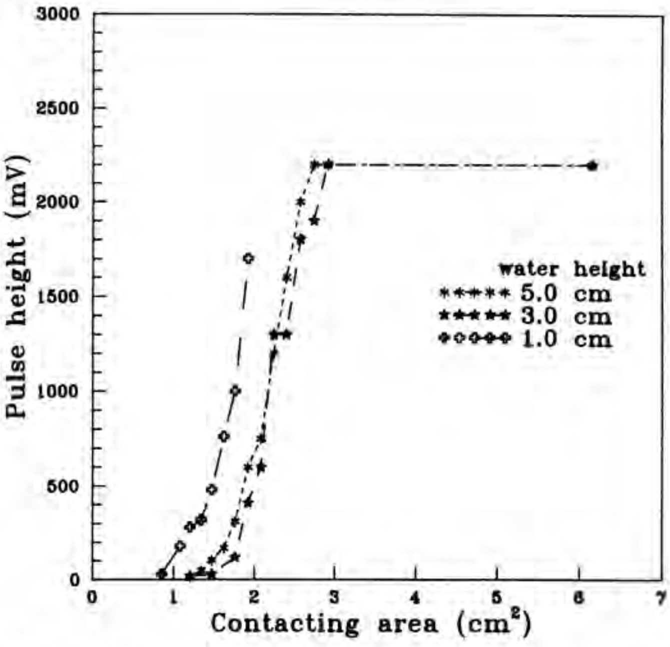

Fig. 12. The variation of pulse height for different water heights versus the contacting area at water/air-interface. 1.0 MHz probe, total area is 6.16 cm^2.

where θ is divergent angle, λ is a wavelength of ultrasonic wave, D is the diameter of a transducer, C is the propagation velocity of UT wave in a material, f is the frequency of a transducer. The higher the frequency is and the larger the diameter is, the divergent angle is the smaller. Variation of water layer falling in the projected zone of the divergent angle reflects different pulse-height. However if the water layer thickness exceeds the projected zone of divergent angle, the UT pulse height variation disappears. Therefore there exists a critical thickness of water layer. The maximum pulse-height will not change with the thickness of water layer after the thickness excesses the critical value. Figures 12 and 13 illustrate the conception of the maximum pulse height variation and the critical thickness water layer.

Method (3): Figure 15 shows the two relation curves of the maximum pulse-height of the first reflected wave and the thickness of the water layer with a 0.5 MHz transducer. From curve 1 and curve 2, we can see that the maximum pulse-heights increase with the thickness of the water layer, and the existence of saturated region at larger thickness agrees with the formula of the divergent angle. Results of Figs. 15 and 16,

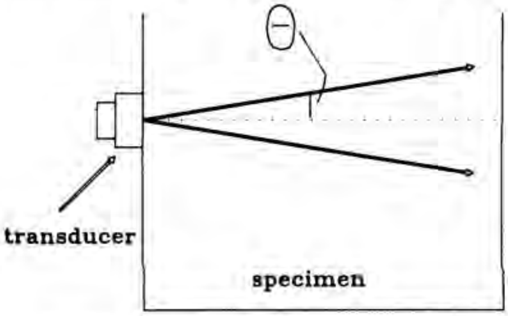

Fig. 13 The divergent angle of ultrasonic wave.

show that the maximum pulse-heights of 1.0 MHz are much larger than those of 0.5 MHz.

Because the emission and receiving stress waves of a transducer in method (3) are more effective than those in method (2). The former is more suitable to evaluate the case with less water. The method (3) can identify the existence of water of 0.3 cm thickness, but the method (2) just can detect the surface free water layer which is above 1.0 cm.

But in the condition of much water (about above 2.0 cm thickness), if the thickness excesses the critical value, the method (3) can't estimate actually the amount of water. Therefore a recommendation of the process of examining surface free water is that the first step to scan the water layer, and check whether there are the reflected wave in water layer, if the maximum pulse-height is near or above the saturated region, the method(2) will be applied, otherwise, the method(3) is applied.

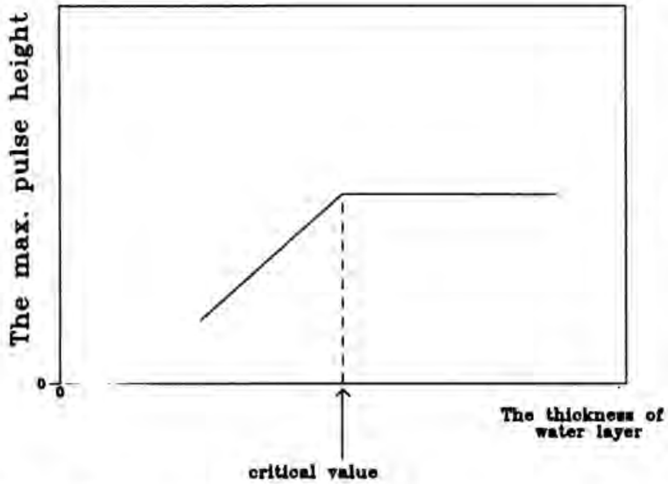

Fig. 14. The maximum pulse height versus the thickness of water layer.

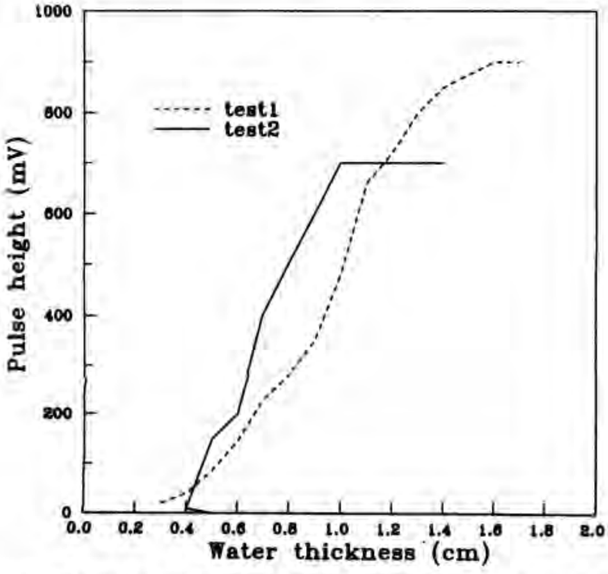

Fig. 15. The relation between the maximum pulse height and the thickness of water layer. 0.5 MHz probe, total area is 12.57 cm^2.

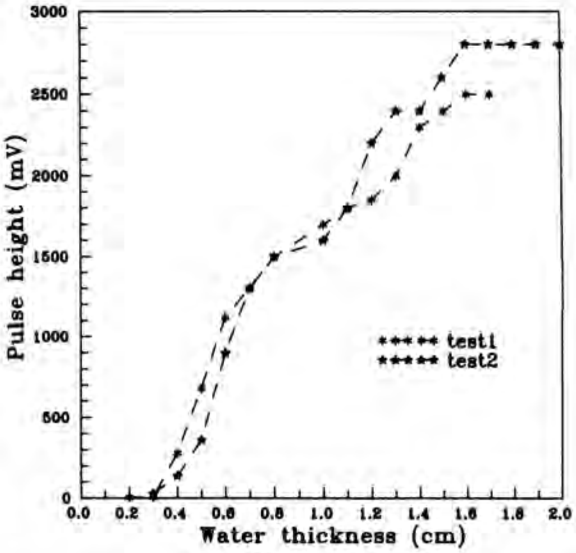

Fig. 16. The relation between the maximum pulse height and the thickness of water layer. 1.0 mHz probe, total area is 6.16 cm^2.

CONCLUSIONS

The objective of the study was to evaluate the applicability of NDT of LLWR drums for uni-axial compressive strength and free water content. The following conclusions are drawn:

- Measurements of the ultrasonic speed of propagation provide an estimate of the uni-axial compressive strength from a correction curve.
- Tests with the larger water/cement ratios exist more surface free water. The less and less amount of surface free water is detected with the increase of cure time of cement.
- In method (1) of UT, success was judged according to how closely the amount of surface free water from the UT method agreed with that from a rule-estimate. The larger the water/cement ratio is, the mean absolute error is larger, but the mean relative error is smaller. In the real application to waste drums, the penetration of ultrasonic wave and the homogeneity of the solidified waste should be considered.
- In method (2) and (3) of UT, method (2) suits better to the condition of existence of more surface free water, and method (3) suits to that of less surface free water.

REFERENCES

1. "Low-level Radioactive Waste Policy Act Report", U.S. Department of Energy, DOE/NE-0015, 1981.
2. "Licensing Requirements for Land Disposal of Radioactive Waste", Code of Federal Regulations, 10 CFR Part 61 Rev. Jan, 1987.
3. R. CLARK and D. BRADLEY, "Nondestructive Evaluation of Low- Level Radioactive Waste Canisters for Free Water Content", EPRI RP 2412-20.
4. V. M. MALHOTRA, "In Situ/Nondestructive Testing of Concrete--A Global Review" V. M. Malhotra, Ed. 1984, American Concrete Institute, Detroit.

5. AKASHI TOYOKY and AMASKI SYOUJI, "Study of The Stress Wave in the plunger of a Rebound Hammer at the Time of Impact" publication SP-82, American Concrete institute, Detroit.

6. I. ODA and H. SATO, "Non-destructive Inspection Technology for Waste Drum and Waste", Consulting Material, JGC on publication.

7. YASUO TANIGAWA, KENJI BABAS, and HIROSHI MORI, "Estimation of Strength by Combined Nondestructive Testing Method" Publication SP-82, American Concrete Institute, Detroit.

8. ASTM C109-80 "Standard Test Method for Compression Strength of Hydraulic Cement Mortars".

9. ASTM C39-81 "Test for compressive strength of cylindrical concrete specimens".

10. M. W. CHENG and L. S. CHI, Radiography, 1st Ed, R.O.C. 1986.

11. V. R. STURRUP, F. J. VECHIO, and H. CARATIN "Pulse Velocity as measure of Concrete Compressive Strength" Publication SP-82, American Concrete Institute, Detroit.

12. Ioan Facaoaru "Romanian Achievements in Nondestructive strength Testing of Concrete" Publication SP-82, American Concrete Institute, Detroit.

13. N. J. CARINO, "Laboratory Study of Flaw Detection in Concrete by the Pulse-Echo Method". in situ/Nondestructive Testing of concrete, SP 82-28.

14. C. C. HUANG, The Ultrasonic Testing, 2nd Ed, R.O.C. 1988.