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# Determination of grain size in uranium-chromium alloys by ultrasonic attenuation

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Ultrasonic attenuation was measured in cylindrical samples of uranium-chromium alloys used for nuclear fuel elements. The grain structure of the samples was previously refined by proper heat-treatment and the grain size in the range of 50 to 200 μm was determined by conventional metallographic methods. The attenuation coefficient, $\gamma$, was correlated with the mean grain diameter, $\bar{D}$, for three ultrasonic frequencies: 4, 6 and 12 MHz. The experimental results were compared with existing theories and good agreement was found with the curves based on Merkulov's analysis for cubic metals. A more practical result of this work was the development of a non-destructive testing procedure for grain-size determination in nuclear fuel elements cast from these alloys. The highest frequency (12 MHz) is normally used as it is more sensitive in the lower grain-size limit (50 to 100 μm) and only in border-line cases would one switch to the other frequencies (6 and 4 MHz) which are more applicable to the higher grain-size limit (100 to 200 μm).

## 1. Introduction

The α-phase of uranium, which is thermodynamically stable below 666°C, is orthorhombic and, therefore, highly anisotropic in its thermal expansion [1]. Since nuclear fuel elements made of uranium undergo considerable temperature cycling in the reactor, their grains expand and contract unevenly causing above a certain critical grain-size surface roughness and macroscopic deformation [2]. A fine equi-axed structure must be achieved throughout the fuel element in order to overcome this problem.

Uranium-molybdenum and uranium-chromium alloys whose grain structure can be refined by proper heat-treatment, were developed for that purpose [3]. The grain size of every fuel element must be non-destructively determined after the final heat-treatment and only those elements whose grain size is smaller than the critical may be used in the reactor.

The purpose of the presently reported work was to develop a quantitative, non-destructive, method for grain-size determination in nuclear fuel elements made of uranium-chromium alloys. This method was intended to serve as the final quality control step in the production process of fuel-elements. Ultrasonic attenuation was chosen as the basis for the method since energy dispersion of an ultrasonic wave travelling in a polycrystalline metal is under properly chosen conditions determined mainly by its grain size [4].

The early theoretical work on grain scattering by Mason and McSkimin [5] is rather qualitative and holds only for $\lambda \gg \bar{D}$ (where $\lambda$ is the ultrasonic wavelength and $\bar{D}$ the mean grain diameter). A more exact treatment was attempted by Roth [6] who also extended the formulation to include $\lambda = \bar{D}$ and $\lambda < \bar{D}$. The presently accepted theoretical equations, derived more

recently by Merkulov for cubic and hexagonal metals [7, 8], are based on the early treatment of Lifshitz and Parkhomovskii [9]. These equations relate the attenuation coefficient (defined as the logarithmic ratio between the incident and attenuated energies $I_0$ and $I$: $\gamma = \ln I_0/I$) to the average grain size of the material, $\bar{D}$, and the frequency of the travelling wave, $f$, in each of the three main ultrasonic scattering regions:

(Rayleigh scattering region: $\lambda > 3\bar{D}$)

$$\gamma = A_1 \bar{D}^3 f^4 \quad (1)$$

(stochastic scattering region: $3\bar{D} > \lambda > \bar{D}$)

$$\gamma = A_2 \bar{D} f^2 \quad (2)$$

(diffusion-scattering region: $\lambda < \bar{D}$)

$$\gamma = A_3/\bar{D} + A_4 f + A_5 f^2 \quad (3)$$

where $A_1, A_2 \ldots$, are constants which depend on the elastic parameters of the material and, therefore, reflect the crystallographic symmetry of its lattice. Merkulov quoted exact expressions for $A_1$ and $A_2$ only in the cubic and hexagonal system. Bhatia and Moore [10] added an exact expression for $A_1$ but not for $A_2$ in the orthorhombic system.

Experimental verification of the above equations was established by Merkulov in several cubic metals, such as Cu, Mg and Fe, and by Papadakis [11], in some more cubic metals and alloys. Although reports of empirical work on ultrasonic grain-size testing in uranium fuel elements can be found in the technical literature [12-17], it appears that no quantitative results relating to Merkulov's theory were as yet reported with regard to uranium alloys or any other orthorhombic metals. Such a correlation between quantitative results obtained in uranium-chromium alloys and the accepted theory is attempted in the present paper.

## 2. Experimental procedures

### 2.1. Sample preparation

Uranium −0.5 at. % chromium alloys were vacuum cast in cylindrical form. Cylinders 30 mm in diameter and 50 mm in height were accurately machined paying special attention to external surface finish and parallelism of the flat faces (within 10 μm). In order to obtain samples of various grain size, these were isothermally transformed into the α-phase at different temperatures. The grain-size dependence on the $\alpha \rightarrow \beta$ transformation temperature in that alloy was previously demonstrated [18].

### 2.2. Ultrasonic measurements

Ultrasonic elastic waves propagated through the cylindrical samples perpendicularly to their long axis and their attenuation by the material was measured employing the pulse-echo method [19]. This perpendicular to the axis rather than parallel geometry was used in order to map accurately variations in grain size which may have occurred along the axis. The experimental set-up is depicted schematically in Fig. 1.

A piezoelectric crystal transducer contained in the probe generates ultrasonic waves in the

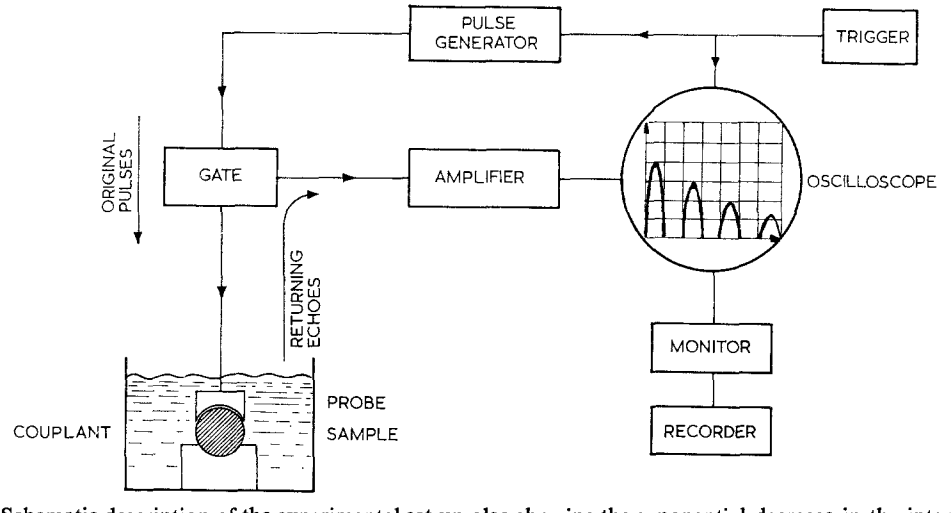


*Figure 1* Schematic description of the experimental set-up also showing the exponential decrease in the intensity of a series of echoes obtained with the oscilloscope.

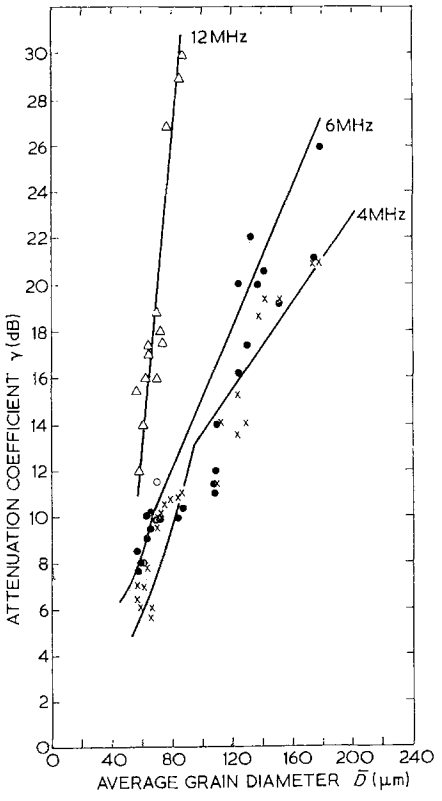


*Figure 2* Plot of the experimental results of the ultra-sonic attenuation ($\gamma$ versus $\bar{D}$) for the three frequencies: 4, 6 and 12 MHz.

frequency range of 1 to 100 MHz. The transducer is excited by a burst of RF pulses emerging from the pulse generator. The width of each pulse is of the order of 1 μsec. After entering the sample the elastic waves echo back and forth between its external round surfaces. The same piezoelectric crystal transducer, which is cylindrically curved to fit concentrically with the sample surface, is now acting as a receiver rather than transmitter, producing electrical signals which are proportional to the intensity of the echoes. These are passed to the oscilloscope through an electronic gate which prevents the original pulses from reaching the oscilloscope. In order to avoid interference between the returning echoes and the original pulses, the pulse repetition frequency is 1000 to 2000 per second so that there is sufficient time between the pulses (500 to 1000 μsec) to allow the series of echoes to decay. This decay is evidenced on the oscilloscope face as an exponential decrease in intensity (Fig. 1). If the shape of the echoes is conserved, it is possible to measure their amplitude instead of their intensity and the attenuation is then expressed simply by:

$$\gamma = -20 \log \frac{A_{i+1}}{A_i} \tag{4}$$

where $i$ is the order of the echo reflection. The actual values of $\gamma$, expressed in dB are obtained by comparing the amplitude of the first echo of the examined sample to the first echo of a metallographically examined standard specimen.

The probe and the sample were submerged in an acoustical couplant, in this case water, in order to avoid total reflections at solid-air interfaces that would have been otherwise formed between the probe and the sample, and between the sample and its holder.

## 3. Results and discussion

The average grain size, $\bar{D}$, of the examined samples was in the range of 50 to 200 μm. This was determined by conventional metallographic methods which resulted in a mean grain-cross-line length, $\bar{L}$, for every micrograph. The relation

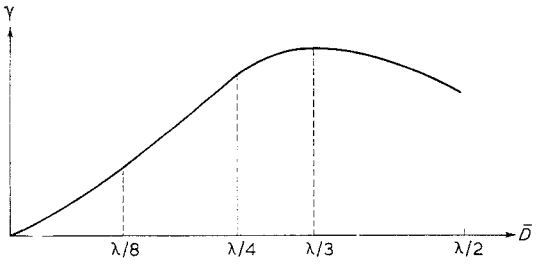


*Figure 3* Plot of theoretical equations relating acoustical attenuation, $\gamma$, versus average grain size, $\overline{D}$, for a constant frequency.

between $\bar{L}$ and $\bar{D}$ was chosen as $\bar{D} = 1.5\ \bar{L}$, intermediate between the expressions given by Fullman [20]: $\bar{D} = (\pi/2\bar{L})$ and Papadakis [11]: $\bar{D} = 1.45\ \bar{L}$.

The coefficient of ultrasonic attenuation, $\gamma$, was measured for every sample. Results for the three frequencies used, 4, 6 and 12 MHz, are presented in Fig. 2 in the form of $\gamma$ versus $\bar{D}$ plots.

These experimental plots can now be compared to the theoretical curves (Fig. 3) obtained by the graphical presentation of Equations 1, 2 and 3 as discussed in the introduction. The first equation ($\bar{D} < \lambda/8$) describes the Rayleigh scattering region. The second ($\lambda/8 < \bar{D} < \lambda/4$), the linear region and the third ($\bar{D} > \lambda/3$), the diffusive scattering region.

After comparing Figs. 2 and 3 it can be seen that our experimental data for 4 MHz fall into the first two regions, for 6 MHz they are confined almost entirely to the second (linear) region and for 12 MHz they fit the end of the linear region. This can be better seen in Fig. 4 which shows the experimental data (dots) together with the theoretical plots obtained using Bhatia's [10] expressions for orthorhombic crystals (broken line) as well as Merkulov's [7, 8] relations for cubic metals (solid line).

Since no theoretical expression was proposed for the linear region in the case of orthorhombic materials, we can only discuss the degree of agreement of our experimental data in this region with the theoretical curves obtained using expressions developed for cubic materials. Indeed, we find this agreement to be quite good in all of the three frequencies employed. Moreover, in the only frequency (4 MHz) where sufficient experimental data for comparison with both theoretical curves was available, we find a

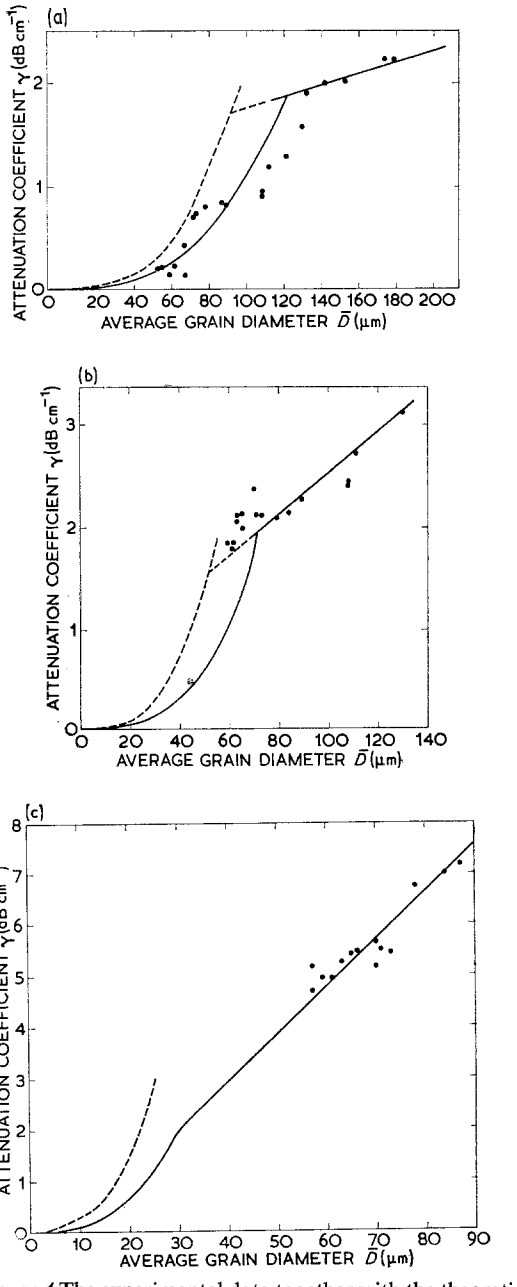


*Figure 4* The experimental data together with the theoretical plots of $\gamma$ versus $\overline{D}$ for: (a) 4 MHz, (b) 6 MHz, (c) 12 MHz.

better fit with the theoretical expression for cubic rather than with that for orthorhombic materials (Fig. 4a).

Another result of our work, which was perhaps of a more immediate and practical importance, was that, by using the three frequencies, we made it possible to determine non-destructively,

reproducibly and fairly accurately the grain size of the uranium-chromium fuel elements in the range of 50 to 200 μm. This is feasible, as shown in Figs. 2 and 4, due to the fact that the linear regions of the three frequencies cover these grain sizes with the higher frequencies corresponding to the smaller values. Thus, in an industrial testing set-up where the smallest possible grain size is preferred, one would tend to use the highest frequency of 12 MHz and only for testing questionable specimens would one switch to the lower frequencies which are more sensitive for larger grain sizes (100 to 200 μm).

It should be emphasized that in all of our experiments the effect of grain size on the ultrasonic attenuation was singled out so as to obtain meaningful results. In other words, effects such as impurities, inhomogeneities and preferred orientation which also considerably affect the ultrasonic attenuation were carefully eliminated.

## 4. Conclusions

Experimental results on ultrasonic attenuation in orthorhombic uranium-chromium alloys in the frequency range of 4 to 12 MHz were found to be in agreement with the theoretical predictions based on Merkulov's treatment for cubic metals.

A practical procedure for non-destructive determination of grain size in nuclear fuel elements made of these alloys was developed employing the three frequencies 12, 6 and 4 MHz and covering the grain-size range of 50 to 200 μm.

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