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ANNUAL REPORT OF RESEARCH PROGRESS ON CONTRACT AT(30-1)-1772

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This report constitutes a brief review of the work undertaken, entirely or in part, under the Contract AT(30-1)-1772 with the United States Atomic Energy Commission.

The work published during 1959 is listed at the end of this report and copies of reprints or reports are attached where available.

These investigations have been concerned with the examination of defect production and behavior as it may be observed by high frequency ultrasonic attenuation and velocity (modulus) changes in such materials as quartz, silicon, germanium, alkali halides (especially NaCl and KCl), high purity aluminum, and glass containing boron. The irradiations used are cobalt 60 gamma-ray irradiation and the reactor irradiation obtained in the Brookhaven National Laboratory graphite reactor.

Co^{60} Gamma-ray Irradiation Studies on NaCl and KCl

The irradiation of the alkali halides by cobalt 60 gamma-ray irradiation is discussed in a separate report entitled "The Effect Of Cobalt 60 Gamma-ray Irradiation on Ultrasonic Attenuation and Velocity in NaCl and KCl". This report describes work done in the past as well as most of the work done during the current contract. In the past year the main effort in this area of work has been that of obtaining better measurements of the velocity changes concurrently with the attenuation measurements during irradiation. The improved velocity difference measurements were essential to showing whether or not the ideas about the mechanisms involved were or were not correct. A description of the ultrasonic velocity measurement unit has been submitted for publication. It has been established beyond any doubt that the effects are dislocation effects, and it seems that a number of the details of the assumed mechanism are also correct judging by the fact that there is quantitative agreement between the model and the experimental results. With much improved instrumentation and techniques it now remains to determine the attenuation and velocity behavior under irradiation conditions at liquid nitrogen temperatures and below. The understanding of the details of the effects observed is obviously a function of the quality, purity, etc. of the materials, and with alkali halides there seem to be even more questions of this type to consider than usual. It is quite probable that oxygen, OH ions, etc., may have to be considered as well as annealing, temperature, deformation and so on. At present efforts are being made to obtain high purity material from other sources than those used thus far.

The details of an investigation involving the study of sodium chloride and potassium chloride <u>during</u> deformation and <u>during</u> the recovery <u>immediately</u> following deformation are the subject of a report now in preparation. There are some surprising results from this work. One of these results is that it has been found that during recovery immediately after deformation the attenuation does not follow even approximately a $t^{2/3}$ law with time. These measurements have been made on many samples and over a range of temperatures between +100°C and -70°C. The differences between these results here and those of Gordon and Nowick as discussed by Granato, Hikata and Lücke are large and cannot be explained by errors of any reasonable sort. The measurements were made over the temperature range indicated in order to obtain an activation energy for the point defects responsible for pinning during recovery - assuming of course a diffusion mechanism such as that used by Granato, et al. The activation energy obtained by Granato, et al was about 1 ev. The recent work yields a value of 0.033 ev. There are a number of possible explanations of this difference but it is difficult to understand why it is so large. One of the main features of the recent work is that the measurements are made during the process of deformation and immediately thereafter - this was not the case in previous work. Another point is the fact that the earlier work was in the kilocycle range of frequencies and this work was done in the megacycle range. It is going to require some work to understand these differences and some of the work will have to be repeated under different conditions. Some work has been done on the temperature dependence of the velocity or modulus in the NaCl. This work is discussed in the report just mentioned.

Effects of Reactor Irradiation on Elastic Moduli of Germanium

Studies of the effect of reactor irradiation on germanium single crystals by means of measurement of the temperature dependence of v in a [100] direction before and after irradiation have shown the following effects. Irradiation of approximately 2×10^{19} nvt produced a change in $\frac{1}{v} \frac{dv}{dT}$ (equivalent to $\frac{1}{c_{11}} \frac{dc_{11}}{dT}$) of 12%. This result was obtained using four samples originally as nearly identical as possible. The $\frac{1}{v} \frac{dv}{dT}$ measurements were made using various combinations of the unirradiated samples using combinations (after two samples were irradiated) of one irradiated sample against one unirradiated sample. These measurements were made at 45 Mc/sec with compressional waves.

These $\frac{1}{v} \frac{dv}{dT}$ measurements were originally made to find out whether or not this type of measurement is a sensitive indicator of changes produced by irradiation. It has become of more interest to determine the manner in which the irradiation produces these changes. The velocity change for comparable irradiation has never exceeded about 1% and while the absolute velocity changes on these particular samples has not yet been measured, it is safe to assume about a 1% increase in v for these circumstances. Consequently it is seen that the change must be an increase in $\frac{dv}{dT}$ of approximately 10 - 12%. It will be necessary to measure the absolute values of v under the same set of conditions as those under which $\frac{1}{v} \frac{dv}{dT}$ is measured if $\frac{dv}{dT}$ is to be determined and studied. The sensitivity of this method to small changes is large, but the accuracy and reproducibility is not what had been hoped for. The accuracy and reproducibility can certainly be greatly improved but whether the effort should be expended in this or other directions has not been decided.

"In Reactor" Measurements During Irradiation

A large amount of effort during the past year has gone into the "in reactor" measurements of attenuation and velocity during irradiation. A number of detailed technique problems had to be settled by experiment. "In reactor" measurements have been made in a fairly satisfactory way on a high purity aluminum single crystal and on quartz. In the case of alkali halides the results to date indicate that damage effects would anneal out so rapidly at normal reactor temperatures that appreciable effects will not appear.

The attenuation change in aluminum at 58°C in the reactor was a decrease from 0.46 db/μ sec initially to 0.36 db/μ sec at the end of 47 hours of irradiation - approximately a 20% decrease with a flux of about 1.7×10^{18} nvt. Interpretation of the attenuation decrease in aluminum can be made on the basis of a dislocation pinning behavior, but for real confirmation it will be necessary to have velocity measurements together with attenuation measurements and at low temperatures as well. In addition it is desirable to have such measurements over a frequency range.

The "in reactor" attenuation changes measured in quartz have been the most successful thus far. The results of reactor irradiation of a single crystal of quartz have been obtained using a thick crystal operating as its own transducer (and sample) at 60 Mc/sec. This eliminates the need for a transducer to be attached externally and makes the measurements much easier and without any question about bond effect. Unfortunately only piezoelectric or magnetostrictive materials can be studied in this way. A plot of the results of an attenuation-irradiation run for quartz are shown. From this plot it is seen that for a period of slightly more than a minute the attenuation was constant, and between one and one and one-half minutes the attenuation began to decrease by approximately 10% as shown. After three to five minutes the direction of the change reversed, and the attenuation began to rise in a very irregular way. After about twenty minutes, trouble with the automatic attenuation equipment stopped the data collection for some hours. When the equipment was again operating the attenuation was increasing and continued to increase beyond the points shown.

While the interruption by the instrument trouble makes it necessary to rerun this experiment, the main features (early decrease in attenuation, the reversal, and later increase in attenuation) should remain the same. The early decrease in attenuation was accompanied by a small velocity increase of approximately 0.1%. The attenuation decrease of about 10% and the accompanying velocity increase of about 0.1% have the proper directions and magnitudes for a dislocation effect. One might ask whether this early change could also arise from heating when the sample goes into the reactor. One cannot rule out the possibility of a temperature effect, but the attenuation change does not have the characteristics of temperature changes previously observed. The matter will be checked by a separate temperature experiment, but this has not yet been done.

The increase in attenuation at the later stages of this experiment is almost certainly caused by the increase in the number of regions of damage in the lattice. Before speculating further in this direction, however, it is necessary to repeat these experiments at several temperatures, if possible, to help establish the mechanisms involved. Both velocity and attenuation measurements are needed as usual and can now be made in a satisfactory way. The automatic attenuation equipment was designed and built during the past year or more to follow fatigue effects in solids over a long period of time; obviously it is equally useful here. There are, naturally, plans to run a large number of materials in the reactor in a manner similar to that of the quartz. In many materials that have been tried, such as some of the alkali halides, the effects observed are rather small, apparently because the damage anneals out as fast as it is produced at temperatures in the reactor (40°C to 70°C). Lower temperature irradiation facilities are obviously essential here.

Additional Studies

Results of multiple scattering theory have been applied to the analysis of attenuation and velocity data in order to estimate the size of damage regions produced in silicon single crystals by fast neutrons. A report of this work appears in PHYSICAL REVIEW for November 15, 1959. Further work has been done on the study of a layered sphere as a model for a damage region; the frequency and size dependence of the scattering cross section is being studied numerically.

A report in which measurements of compressional wave and shear wave velocities along the z-axis of Al_2O_3 is being prepared. The results check well with data in the literature. These samples will be studied under irradiation.

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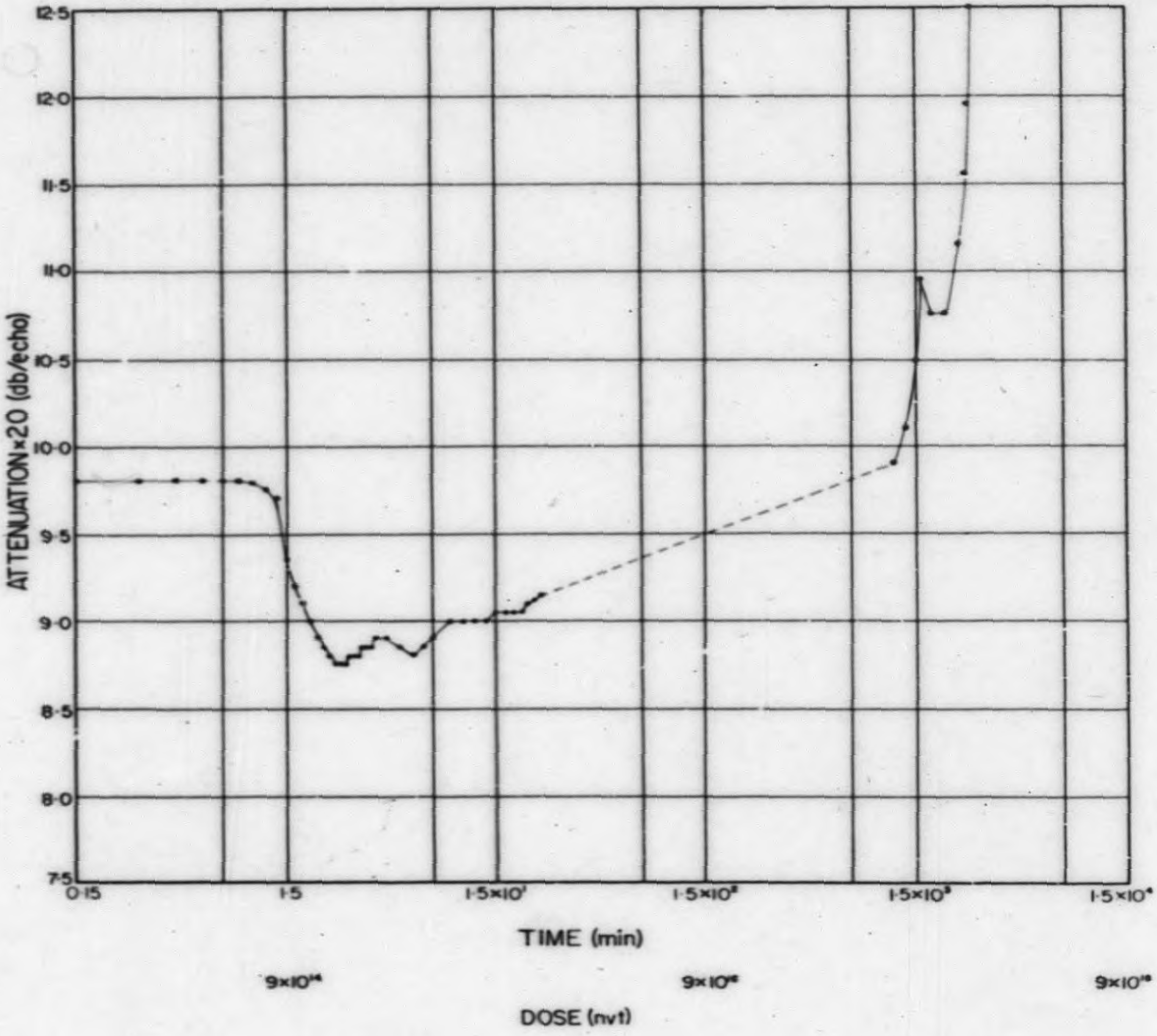

Attenuation in Single Crystal of X-Cut Quartz
Subjected to Reactor Irradiation