

NBS TECHNICAL NOTE 939

U.S. DEPARTMENT OF COMMERCE / National Bureau of Standards

NBS Reactor: Summary of Activities July 1975 to June 1976



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NBS Reactor: Summary of Activities July 1975 to June 1976

FEP. on, Jan Garmant

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Issued May 1977

National Bureau of Standards Technical Note 939 Nat. Bur. Stand. (U.S.), Tech. Note 939, 140 pages (May 1977) CODEN: NBTNAE

> U.S. GOVERNMENT PRINTING OFFICE WASHINGTON: 1977

For sale by the Superintendent of Documents, U.S. Government Printing Office Washington, D.C- 20402 - Price \$2.75

Stock No. 003-003-01769-8

FOREWORD

The National Bureau of Standards Reactor was built not only to serve the needs of the NBS but also those of other government agencies and the greater Washington Scientific Community. The Reactor Radiation Division was established to operate the reactor and to foster its scientific and technological use. Toward this end, the Division has a small nucleus of scientists experienced in the use of reactors for a wide range of scientific and technical problems. In addition to pursuing their own research and developing sophisticated experimental facilities, they actively seek out and encourage collaboration with other scientists, engaged in challenging programs, whose work can benefit from use of the reactor, but who as yet do not have the reactor experience necessary to take full advantage of the facilities available. The Division also provides irradiation services to a wide variety of users as well as engineering and other technical services.

The reactor operates at 10 MW and is designed to provide more than 20 experimental facilities ranging from intense neutron beams to extensive irradiation facilities, making it one of the most versatile high flux research reactors in the country. Thus it is able to serve a large number of scientists and engineers in a broad range of activities both within and outside the NBS.

This report attempts to summarize all the work done which is dependent on the reactor including a large number of programs outside the Division. The first section summarized those programs based primarily IN Reactor Radiation Division (RRD) initiatives whereas the second and third sections summarize collaborative programs between RRD scientists and other NBS or non-NBS scientists respectively. The fourth section summarized NBS work originating entirely outside the RRD which requires no collaboration with RRD scientists. The section entitled, "Service Programs" covers those programs originating outside NBS but for which RRD provides irradiation services. The remaining sections are selfexplanatory.

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Appreciation is extended to F. J. Shorten of the Reactor Radiation Division for his extensive contributions to the editing, organization and preparation of this report, and to L. Sprecher, C. O'Connor and P. Lewis for efforts in typing manuscripts.

J. J. Rush J. J. Rush Chief, Reactor Radiation Division (Acting) National Bureau of Standards

ABSTRACT

This report summarizes all those programs which depend on the NBS reactor. It covers the period from July 1975 through June 1976. The programs range from the use of neutron beams to study of the structure and dynamics of materials through nuclear physics and neutron standards to sample irradiations for activation analysis, isotope production and radiation effects studies.

Key words: Activation analysis: crystal structure; diffraction; isotopes; molecular dynamics; neutron, nuclear reactor; radiation.

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NEUTRON SCATTERING STUDIES OF HYDROGEN IN METALS

Our long range program to study the binding and diffusion of hydrogen in metals, and related properties of metal-hydrogen systems has provided new results particularly in the study of the lattice dynamics of metal hydride crystals. Our collaborative interactions with the Argonne and Oak Ridge National Laboratories have continued and a new collaborative effort has been initiated with scientists at the Allied Chemical Materials Research Center. New results have been obtained on the crystal dynamics of VD_x and $CeD_{2.12}$ and on a detailed comparison of phonon lineshape profiles in a non-stiochiometic deuteride ($PdD_{0.63}$) with computer simulation calculations of the lattice dynamics of this disordered high density "defect" structure. We have also continued our studies of the structure and dynamics of the low concentration ($\alpha \& \beta$) phases of TiH_, as discussed in another part of this report.

1. The Lattice Dynamics of VD_{0.7} - J. J. Rush, C. J. Glinka and J. M. Rowe

The lattice dynamics of a single crystal of $\alpha(bcc)$ VD_{0.7} has been investigated by coherent inelastic neutron scattering measurements in collaboration with H. E. Flotow of Argonne National Laboratory. Due to the very small vanadium coherent cross section, essentially all the coherent scattering by this compound is associated with the deuterium (located in tetrahedral interstitial sites). Thus a study of this system provides a unique opportunity to measure the "in-band" and optical modes associated with the vibrational displacements of the deuterium (light atom) interstitials as a function of \vec{q} and ω .

The VD_{0.697} crystal prepared for these experiments had a "mosaic spread" of <1°. Some example of neutron groups measured under both constant ΔE and constant \vec{Q} conditions are shown in figure 1. Also present as a shoulder in figure 1b is the incoherent "density of states" scattering which is present in all phonon scans at higher energies and



Figure 1. Examples of phonon groups measured for α -VD_{0.7}. The arrow and dashed line in group c. ([111]TA at ξ -0.25) indicate the incoherent "density of states" peak.

makes the observation of coherent phonon peaks above ~20 meV quite difficult. The phonons measured thus far at $\xi \leq 0.5$ in the three symmetry directions are shown in figure 2. The results in figures 1, 2 represent a direct measurement of the "in band" vibrations of the interstitial deuterium atoms.

It is interesting to note from these results that: a) the observed neutron groups are well defined, with little apparent broadening (for $h\omega \le 20$ meV) due to the random distribution of interstitials, b) the phonon frequencies measured for VD_{0.7} are in general higher than those for pure vanadium, indicating a general increase in interatomic forces on hydriding in spite of a ~5% expansion of the lattice (a (V) = 3.03Å, a (VD_{0.7}) = 3.145 Å). In fact the elastic constants obtained from the long wavelength (low Q) slopes of the present measurements (figure 2) are 30-50% larger than the measured values for vanadium.

As a further attempt to elucidate the changes in vibration spectrum which occur on deuteration we have made energy transfer scans at constant

 \bar{Q} and fixed $E_{\rm F}$ for both VD_{0.7} and pure vanadium. These results were used to derive phonon "density of states" distributions for V and VD_{0.7}, assuming one-phonon incoherent scattering. The results are shown in figure 3. Both of these "distributions" reflect primarily the vibrational displacements of the vanadium atoms (although the contribution of D complicates this analysis). It is clear from the results that the addition of deuterium has considerably altered the frequency distribution. In particular the cutoff of phonon energies is apparently shifted to higher energies for the VD_{0.7} crystal.



Figure 2. Partial dispersion curves in the three symmetry directions for α-VD_{0.7}. Dotted lines indicate the long wavelength slopes of the dispersion curves predicted by the measured elastic constants of vanadium.

As discussed above the coherent inelastic scattering from $VD_{0.7}$ reflects only the D vibrational displacements so that the measurements in figures 1, 2 allow a direct test of the coupling of interstitial deuteriums to the host metal lattice. We have derived integrated intensities from careful measurements of neutron groups in several crystal directions up to phonon energies of 20 meV. The relative intensities of the groups vs. \vec{q} and ΔE show no variation within the



Figure 3. Phonon "density of states" - G'(h ω) derived from energy spectra measured at constant Q in V and α VD0.7. The pure vanadium results represent a smooth line drawn through many data points. The phonon densities are only approximately normalized. Energy resolutions (FWHM) are indicated by the horizontal bars.

estimated errors, thus providing clear evidence that the D atoms are strongly coupled to the acoustic vibrations of the vanadium lattice.

No adequate force model fit to the data in figure 2 is possible without information on phonon frequencies at larger ξ values. Such measurements are quite difficult due to the superposition of the phonon "groups" on the density of states scattering at energies ≥ 20 meV, but further work is in progress.

 Inelastic Neutron Scattering Lineshapes in PdD_{0.63} - C. J. Glinka, J. M. Rowe, and J. J. Rush

In a previous coherent neutron scattering study¹ of the lattice dynamics of the non-stiochiometric metal hydride PdD_{.63}, severely broadened lineshapes were observed for the optic phonons even at low temperature (80K). While the phonon peak positions observed in this



Figure 4. Representative constant-Q scans of the scattering in the optic mode region from PdD at 77K. The solid curves are the intensity profiles calculated from the structural model for PdD.63.

earlier work could quite adequately be derived from a second near neighbor Born-von Karman model in which the crystal was treated as stoichiometric PdD (NaCl-structure), such a model predicts sharp phonon lineshapes and thus gives no insight into the nature of the observed linebroadening. Recently however, A. Rahman has completed calculations of the coherent "one-phonon" scattering for PdD_{.63} based on a model for the lattice dynamics which incorporates the large number of vacancies in the actual structure. The calculations entail computing the normal modes of vibration of a large unit cell of 108 Pd atoms with 68 deuterium

atoms situated at random on the deuterium sublattice. The frequencies and polarization vectors obtained by repeating the computations for a statistical ensemble of such cells are then used to evaluate the onephonon cross section. In order to critically test these calculations, new, more extensive, measurements have been made of the inelastic scattering in the optic mode region. These measurements and calculations, (which have been reported in a preliminary publication²), have helped to elucidate not only the dynamics of PdD_{.63}, but also the more general problem of the effects of vacancies on the lattice dynamics of solids.

In figure 4 three typical inelastic scattering lineshapes, measured with a triple-axis spectrometer at 80K, are shown with corresponding calculated lineshapes (solid lines). Although the structural detail in the calculated lineshapes is dependent on the choice of unit cell size used in the computations, and therefore must be discounted, the overall widths are not and can be seen to satisfactorily account for the observed width in each case. This agreement demonstrates that the observed widths are primarily the result of the defect structure rather than a manifestation of anharmonic effects, for example. These results strongly suggest that the character of the excitations in a highly defected structure is markedly different from the plane-wave behavior found in stoichiometric crystals.

Efforts are now underway to produce a higher concentration single crystal sample of PdD_x with X \sim 0.9. For X>0.8, PdD_x becomes superconducting and there is currently much interest in the role of lattice vibrations in this material in the inducement of superconductivity. To achieve this higher concentration, a crystal of Pd will be loaded under a deuterium pressure of 2 kbar in a high pressure sample cell which has been especially designed for this purpose and may subsequently be mounted in a cryostat for the low temperature measurements. Further studies along these lines are planned for the alloy-hydride system $Pd_yCu_{1-y}D_x$ for which Tc's as high as 18 K have been reported, but these are awaiting the preparation of appropriate single crystals.

- 1. J. M. Rowe, J. J. Rush, H. G. Smith, M. Mostoller, and H. E. Flotow, Phys. Rev. Letters <u>33</u>, 1297 (1974).
- C. J. Glinka, J. M. Rowe, J. J. Rush, A. Rahman, S. K. Sinha, and H. E. Flotow, Proceedings of the Conference on Neutron Scattering, Gatlinburg, TN. 1976 (in press).
- Lattice Dynamics of Cerium Deuteride C. J. Glinka, J. M. Rowe, and J. J. Rush

A comprehensive study of the phonon dispersion curves of CeD_{2+x} , in collaboration with A. Maeland and G. Libowitz of Allied Chemical, has been initiated and thus far preliminary coherent scattering measurements have been made on a single crystal of $CeD_{2.12}$. It should be noted that CeD_2 has the same fcc (CaF_2) structure widely exhibited in transition-metal hydrogen systems (e.g., Ti, Zr, Hf, Y) whose physical, thermo-dynamic and electronic properties have been extensively studied.¹ The cerium hydrogen system has received considerable attention due in part to the occurrence of composition-dependent metal-to-semiconductor phase



Figure 5.Neutron groups observed in constant-Q scans at room temperature in CeD_{2,12}.

transitions at low temperature. Our measurements represent the first coherent inelastic scattering study of a nearly stoichiometric metal hydride and should, therefore, afford the opportunity to investigate the crystal dynamics (including anharmonic deuterium vibrations) and related physical properties, virtually free from any perturbation of the phonons due to nonstoichiometry. This study is also expected to provide a definitive test of the restricted central force model (in which "bond bending" forces are neglected) which has been used² in fitting to specific heat data and time-of-flight neutron spectra on this material.

Figure 5 shows two typical optic phonon scans from our preliminary data on CeD_{2.12}. The six optic mode branches all lie within 74 to 84 meV and the coherent scattering from these modes is superimposed on a considerable background of incoherent scattering which reflects the high density of states in this region. Although more detailed measurements will be needed to establish the connectivity of the closely spaced optic branches, our present results indicate that a restricted central force model is not adequate to describe their dispersion.

The peak seen in the neutron groups in figure 1 at $\hbar\omega = 94$ meV is present in all of our scans and may arise from localized vibrations of hydrogen atoms randomly distributed among the deuterium sites (our sample contains a few atomic percent hydrogen). The observation of such a mode is potentially a very exciting development since the hydrogen represents a light mass substitutional defect with (presumably) no change in the atomic forces. This system may, therefore, provide the first real test case for mass defect theory in the dilute limit.

Measurements at room temperature on the $CeD_{2,12}$ crystal are continuing. We also intend to extend our phonon measurements to low temperature and to a higher concentration of $CeD_{2,12}$.

^{1.} W. M. Mueller, J. P. Blackledge and G. Libowitz, *Metal Hydrides*, Academic Press, New York (1968).

^{2.} P. Vorderwisch et al., Phys. Stat. Sol. (b), <u>65</u>, 171 (1974).

CRYSTAL DYNAMICS, PHASE TRANSITIONS AND ORIENTATIONAL DISORDER IN IONIC CRYSTALS

We have continued and extended our studies of the dynamics and structure of orientationally disordered crystals. It is unfortunately still correct to say that these systems are poorly understood, although some progress has been made.

Our research during the past year has been centered on a study of the structure of KCN and NaCN in all three phases, on extending our measurements of the crystal dynamics of KCN and on measurements of both crystal dynamics and reorientational dynamics of deuterated ammonium halides.

 Alkali Cyanides - J. M. Rowe, J. J. Rush, E. Prince, N. J. Chesser, S. Susman and D. G. Hinks

We have completed a study of the crystal structures of KCN and NaCN in all three phases by neutron powder diffraction. A set of characteristic powder patterns of KCN in all three phases is shown in figure 1. The salient feature of the results shown in this figure is the lack of observable peaks in cubic KCN for scattering angles greater than about 62°. This is a reflection of the very large mean square displacements of both metal and cyanide ions in the cubic phase which arise from the soft shear mode (C_{44}) previously observed. The extra peak is a superlattice reflection characteristic of the low temperature fully ordered orthorhombic structure.

The intensity of this superlattice line is proportional to the square of the order parameter for this order-disorder phase transition. The temperature dependence of the intensity is shown in figure 2.

In the ordered low temperature structure the two (CN)- ions in the unit cell are ordered antiparallel to each other along the b axis.

In addition, we have completed a more detailed study of the dynamics of KCN. The dispersion relation for acoustic phonons has been observed over a larger fraction of the Brillouin zone as shown in figure 3. This extended information was obtained on the new BT4 threeaxis spectrometer, and it was the extra intensity and flexibility of this instrument which allowed us to obtain the new data. However, it must be emphasized that many of the neutron groups (especially at the larger energies) were very poorly defined and that the dispersion curve shown should not be interpreted in terms of harmonic or quasiharmonic models. The characteristic feature of the dynamics of KCN is the strong anharmonicity evidenced in the neutron lineshapes observed.

Finally, we have refined our measurements of the temperature dependence of the soft shear modes characterized by C44. As was shown previously, the elastic constant C_{44} tends to zero as the temperature is lowered towards the first order phase transition at 168K. Our previous measurements of the neutron scattering associated with these soft modes was complicated by the large elastic scattering associated with the Bragg peak about which the measurements were made. By changing the experimental conditions, we have been able to remeasure these modes with very little contamination and have shown that there is no detectable elastic component in the scattering for reduced wave vectors as low as 1/10 the distance to the Brillouin zone. At this wave vector the energy of the phonon is approximately 0.2 meV just above the phase transition. Recent theories have predicted the existence of elastic scattering (central modes) for these phase transitions, so that the present results were not expected. However, the predicted behavior may only occur at smaller wave vectors than could be reached in this experiment, or the first order phase transition may occur at a temperature too far above the point at which C44 extrapolates to zero.

We intend to check these and other possibilities by extending our research to mixed alkali halide-akali cyanide crystals, beginning with



Figure 1. The observed powder diffraction patterns for the three crystal phases of KCN. Note the comparative intensities of high angle reflections in the cubic and intermediate phases and the superlattice peak (blown up in insert in the low temperature phase).



Figure 2. Temperature dependence of superlattice intensity in the low temperature phase of KCN. Solid lines are result of theory described elsewhere (R. Casella).



Figure 3. Dispersion relation for "phonons" in KCN at 295K. Solid lines are guide to the eye. (o) longitudinal acoustic modes (o,∆) transverse acoustic modes.

(K Br) (KCN) 25 and proceeding to higher cyanide concentration.

2. Ammonium Halides - J. M. Rowe, J. J. Rush and N. Vagelatos

We have successfully completed our program to grow large deuterated single crystals of ammonium iodide and ammonium bromide, and have made detailed lattice dynamical measurments on the NaCl phase of $\mathrm{ND}_4\mathrm{I}$ (see last years report). In addition we have completed preliminary measurements on the lattice dynamics of ND_4 Br in the CsCl phase at room temperature. We intend to study this system as a function of temperature and hope to make measurements in the low temperature tetragonal phase. We have also made preliminary measurements of the quasilastic scattering from $\mathrm{ND}_4\mathrm{I}$ in an effort to establish the reorientation mechanism in this high temperature phase. These preliminary measurements indicate that the experiments are feasible, but also point up the difficulty associated with the large amplitude motions of the ND_4 ions and the broad distribution of equilibrium orientations in this phase.

ORDER-DISORDER PHASE TRANSITION IN KCN

R. C. Casella

Experimental work of Rowe, Rush, Prince, and Chesser¹ has indicated that below $T_c \simeq 83$ K the CN⁻ dipoles undergo antiferroelectric ordering. Earlier attempts to fit the temperature dependence of the order parameter η deduced from their data met with difficulties. I was led to consider the analogue to the Ising model in magnetism. It turns out that their data for the intensity of neutrons from the superlattice reflection ($\propto \eta^2$) lie midway between what one expects for a twodimensional and a three-dimensional Ising model and is fit (within errors) by the expression

$$\eta^2 = \left[1 - \left(\sinh \frac{T_1}{T}\right)^3\right]^{1/3} \tag{1}$$

roughly the square root of what one expects for the 3-D Ising model







Figure 2. Data from reference 1 compared with Ising model predictions: lattice symmetry corrections. The CN⁻ sites lie in a slight orthrombic distortion of what would otherwise be facecentered-cubic positions. Here the quasi-f.c.c. structure of the lattice is taken into account. Upper curve: Twodimensional square lattice as in figure 1 (for comparison). Lower curve: n^2 for f.c.c. Ising lattice, as calculated numerically.² Middle curve: the square root of the lower curve.



Figure 3. Comparison of the approximate analytical expression (a) $n = [1 - (\sinh (t_1/t))^{-3}]^{1/3}$ for the three-dimensional simple-cubic Ising lattice with (b) numerically calculated extrapolations of same.² (t = T/T_c, t₁ = T₁/T_c)

(see figures 1-3). Since T_c (= 82.9 K) is determined from specific heat measurements, there are *no free parameters* in these fits. The physical origin of this "anomalous-dimensional" behavior is not clear, but is possibly associated with long range aspects of the forces, not included in the Ising model.

- 1. J. M. Rowe, J. J. Rush, E. Prince, and N. J. Chesser, elsewhere in this report.
- 2. D. M. Burley, Phil. Mag. 5, 909 (1960).

NEUTRINO SCATTERING

R. C. Casella

The neutrino, postulated by Pauli in 1930 to save the principles of conservation of energy and momentum in nuclear β decay and experimentally confirmed by Reines, Cowan, and coworkers using a fissionreactor source of electron antineutrinos $\bar{\nu}_{a}$ in the period 1950-1960, is a unique probe of matter over a range of scales differing by at least 24 orders of magnitude. Because of its weak interaction, the neutrino carries information from the interior of stars (R $_{\rm \sim}$ 10^9 m) as well as entering deep into nucleons (r < 10^{-15} m), where the next layer of building blocks (postulated quarks and partons) can be probed. By modifying the quark-parton model of Feynman and of Bjorken and Paschos in the so-called wee region of the kinematic variable x, I have been able to account quantitatively for the high-y anomaly in antineutrinonucleon scattering experiments, the rise in the ratio of antineutrino to neutrino charged-current cross sections with energy, and qualitatively to explain the surprising high-strangeness multiplicity associated with neutrino-induced μe production. [μ = muon and y is the fractional energy loss in the laboratory frame of the lepton in the



Figure 1. dσ/dy vs. y for vN and vN scattering. Experiment, as labeled. Theory, E = 20 GeV, normalization relative to data arbitrary. Data from references 1 and 2. Crosshatched regions indicate detection-efficiency corrections incomplete.



Figure 2. $d\sigma/dy$ vs. y for vN and \sqrt{N} scattering. Experiment, as labeled. Theory, E = 100 GeV, normalization relative to data arbitrary. Data from references 1 and 2. Cross-hatched regions require further data reduction.



Figure 3. $d\sigma/dy vs. y$ for \sqrt{N} scattering (a) in small x region only, (b) excluding x < 0.15. Experiment, as labeled. Theory (a) upper curve, E = 100 GeV, lower, E = 50 GeV, x < 0.14, normalization *fixed* relative to that in (b). Theory (b) all E > 10 GeV, 0.14 < x < 0.6, normalization relative to data arbitrary. Data from reference 2.

process, ν_{μ} + nucleon $\rightarrow \mu^{-}$ + anything; i.e., $y = (E_{\nu} - E_{\mu})/E_{\nu}]$. I have done so without the necessity of introducing right-handed (V + A) currents not contained in the standard (V - A) version of Fermi's weak interaction theory as formulated after the discovery of parity violation in 1956 by Wu, Ambler, Hayward, Hoppes, and Hudson at NBS. Theoretical fits to the data^{1,2} span an energy range, 10 GeV to 150 GeV (see figures 1-4).



Figure 4. Absolute value of the total charged-current-induced neutrinonucleon cross section vs. energy. Theory, E > 10 GeV, heavy curve. The straight line is an extrapolation from lower energy Gargamelle data, not shown. Solid circles, HPWF data, open circles, CF data.

^{1.} A. Benvenuti, et al., Phys. Rev. Lett. <u>36</u>, 1478 (1976).

^{2.} A. K. Mann, paper presented at Orbis Scientiae, Univ. of Miami, Coral Gables, Florida (1976).

SPIN WAVES IN FERRIMAGNETIC ERBIUM-IRON J. J. Rhyne

and

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The group of compounds RFe_2 , where R is a heavy rare earth, crystallize in the close-packed Laves phase or MgCu₂ structure. These compounds have high Curie temperatures (T₂>550K) and show extraordinarily large magneto-elastic interactions¹. The iron moments are ferromagnetically aligned, and are antiparallel to the R moment (ferrimagnetic order). The iron moment as determined from both bulk measurements¹ and neutron scattering² on polycrystalline samples shows an anomalous reduction from the expected $2\mu_B$ to approximately $1.6\mu_B$. The rare earth moments have their free ion values.

We have studied the spin wave dispersion in ErFe_2 using inelastic neutron scattering and have compared the results to a theoretical model. Inelastic scans along three principal axis directions were made at room temperature (T_c for $\text{ErFe}_2 = 574\text{K}$)⁴ on a single crystal approximately 0.5 cm³ using a triple axis spectrometer.

 ErFe_2 has 6 atoms per unit cell with the Er atoms arranged on sites of tetrahedral symmetry and the Fe atoms on octahedral sites. One thus expects six spin wave branches, one acoustic and five optic. As the results in figure 1 show, only three of these branches have energies easily studied by neutron scattering. The data in figure 1 for spin waves propagating in the [q,q,o] and [q,q,q] directions show isotropic spectra consisting of (1) an acoustic mode with no measurable anisotropy energy gap, (2) a very flat optic mode of energy 5.1 meV degenerate with the acoustic mode at the zone boundary and (3) a highly dispersive higher optic mode starting from an energy gap of 8.75 meV. This latter mode has quadratic dispersion ($\hbar\omega = D|q|^2$) in the q range shown with $D = 240 \text{ meV-A}^2$ only slightly smaller than pure iron metal⁵ ($D = 280 \text{ meV-A}^2$)

and would represent the iron acoustic mode in the absence of rare earth spins and the R-Fe exchange interaction. This mode is shown over only a small portion of the Brillouin zone due to the very high energy transfers involved. The model calculation gives an energy of 172 meV at the zone boundary. The vanishing dispersion in the lower optic mode is a consequence of negligible Er-Er exchange in this compound; rather the Er simply sits in the molecular field from the iron spins. Data were also taken in the [q,o,o] direction which again indicated no evidence of a dependence of the dispersion on propagation direction.

A nearest neighbor interaction spin-wave model has been applied to explain the data of figure 1. The Hamiltonian included exchange and crystal field terms (the latter are negligible at room temperature) and had the form

$$H = \sum_{n,m} \sum_{i,j} 2J_{ij}^{nm} \vec{S}_i^n \cdot \vec{S}_j^m + H_{c.f}$$

where J_{ij}^{nm} describes the interaction between the i'th ion of spin S_i in cell n with the j'th ion of spin S_j in cell m (i and j thus range from 1 to 6 and n, m were restricted to only nearest neighbors). The spin wave energies were calculated from Fourier transforms of the equations of motion for the spin raising and lowering operators $S_{\pm}^{+} = S_{\pm}^{+i}S_{v}$ as follows:

$$\hbar\omega(q) S_{i}^{\dagger}(q) = -2\sum_{j=1}^{\circ} \langle S_{i}^{Z} \rangle J_{ij}(q)S_{j}^{\dagger} - \langle S_{j}^{Z} \rangle J_{ij}(o)S_{i}^{\dagger}$$

where $J_{ij}(q) = \sum_{mj} J_{ij}^{nm} \exp [i\vec{q} (\vec{r}_{mj} - \vec{r}_{ni})]$

and the six equivalent relations for S⁻. In fitting the spectra the spins on the Er and Fe atoms were fixed at 3.6 μ_B and 0.66 μ_B respectively as obtained from a combination of neutron diffraction and bulk magnetization data¹⁻². At the zone center the energies of the two optic modes are given simply by $\omega_1^{\text{op}} = 24J_{\text{Er-Fe}} < S_{\text{Fe}}^Z > \text{ and } \omega_2^{\text{op}} = 12J_{\text{Er-Fe}} (< S_{\text{Er}}^Z - 2<S_{\text{Fe}}^Z) + 22S_{\text{Fe}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Fe}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Fe}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Er}}^Z - 2<S_{\text{Fe}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Fe}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Fe}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Er}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Er}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Er}}^Z > 12S_{\text{Er-Fe}} < S_{\text{Er}}^Z > 12S_{\text{Er}-\text{Fe}} > 12S_{\text{Er}-\text{Fe}} < S_{\text{Er}}^Z > 12S_{\text{Er}-\text{Fe}} > 12S_{\text{Er}-\text{Fe}} < S_{\text{Er}}^Z > 12S_{\text{Er}-\text{Fe}} > 12S_{\text{E}-\text{Fe}} > 12S_{\text{E}-\text{$



Figure 1. Magnon dispersion of ErFe₂ along [q,q,o] and [q,q,q] symmetry directions. The solid line is the result of a nearest neighbor spin wave calculation.

The remaining exchange constant $J_{Fe-Fe} = 30$ meV was adjusted to obtain the correct dispersion in the higher optic mode. These values were similar to those for the analogous compound Ho $_{.88}$ Tb $_{.12}$ Fe $_2^{-3}$. J_{R-R} is equal to zero in both cases to an accuracy of approximately $\pm .01$ meV as determined from the maximum dispersion in the lower optic mode. Within the restricted q-range of the higher energy data, this model fits the experimental results extremely well. Its success in predicting accurately the E and q dependence of the remaining magnon branches is not known.

The measurements are currently being extended to lower temperatures to examine the effects of the strongly enhanced anisotropy and magnetoelastic interaction on the spin wave spectrum.

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ORDER-DISORDER IN V3 Ga

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It is known that the superconducting transition temperature of compounds of stoichometry near V_3 Ga can be varied by appropriate heat treating. In an effort to discover if a correlation exists between the T_c and the degree of atomic order of the V and Ga atoms a neutron diffraction study was undertaken of several samples of different heat treating and also of slight composition variation from stoichometric V_3 Ga. Neutrons are significantly superior to x rays for this type of investigation due to the very similar x-ray scattering factors of V and Ga which makes separate site occupancy factors difficult to determine. In the neutron case V and Ga scattering amplitudes have opposite signs and differ by more than a factor of 10.

Initial results have indicated that compounds even in the "as cast" state are highly ordered with long range order parameters as high as 0.95 and that heat treatment annealing at 1400° C produces only a slight increase in the long range order parameter while it lowers the super-conductivity transition temperature by about 0.5 K.

As a result, first indications suggest that the variations in T_c are not strongly correlated with long-range crystallographic order in V_3 Ga and certainly that T_c does not increase with crystallographic order as previously predicted.

APPLICATION OF ROBUST/RESISTANT TECHNIQUES TO CRYSTAL STRUCTURE REFINEMENT

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and

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A technique for fitting a theoretical model to a set of experimental data points and estimating the best values of adjustable parameters in the model is said to be "robust," or, more precisely, "robust of efficiency," if the parameter estimates have near minimum variance for a broad class of distributions for the errors in experimental data. A technique is "resistant" if the estimates are not highly dependent on any small subset of the experimental data. To date, all techniques that are robust of efficiency are also resistant. Many data analysts feel that this is inevitable, hence, the splice word, robust/resistant.

The technique of least squares, the one most commonly used for refining crystal structures, is neither robust nor resistant. Least squares was designed specifically by Gauss for normally distributed errors. With broad-tailed error distributions and/or the presence of a few bad data points in a large data set, least squares can give anomalous results. In recent years, statisticians have been developing methods of fitting and parameter estimation which work nearly as well as least squares for ideal data sets, but which are robust/resistant. The purpose of this research is to apply these methods to crystal

structure refinement, both as a non-trivial test of the robust/resistant approach and as a development in calculational crystallography.

The technique of least squares is based on finding the minimum value of the quantity $\sum_{i=1}^{\infty} (y_{i0} - y_{ic})^2$, where y_{i0} is the value of the observed data, y_{ic} is a calculated value of that point based on some theoretical function containing adjustable parameters, and w_i is a weight. For the method to work well, the weight, w_i , must be proportional to $1/\sigma_i^2$, where σ_i^2 is the variance of the observation y_{i0} . This variance is, however, usually not known a priori, but is approximated by a formula containing a term arising from an assumed Poisson distribution of random counts and one or more other terms which estimate additional experimental error associated with the observed value, y_{i0} .

Tukev¹ describes a robust/resistant alternative to least squares estimation which is particularly attractive for crystal structure refinement. In the least squares program, the response of a refinement is modified by adjusting the weight according to the formula w' = $w_i W(x)$, where x = $(y_{i0} - y_{i0})/K\hat{\sigma}_i$. $\hat{\sigma}_i$ is the standard deviation approximation described above and K is a robust/resistant estimate of The function W(x) is called a "W-function." Characteristics scale. that a W-function should have in order to create a robust/resistant refinement are discussed elsewhere by the authors.² To perform the refinement, an initial set of parameters is used to form normalized residuals $r = (y_{i0} - y_{ic})/\hat{\sigma}_{i}$. These are used to construct K. Independent work of Andrews and Beaton and Tukey 4 suggest a K of 6 times the median absolute residual. With K specified, W(x) is calculated and the weighted least squares iteration performed. After each iteration, K is recalculated as above.

The crystallographic least squares program RFINE4⁵ was modified to include, as options, several different W-functions. One of these W-functions was used to investigate the operational characteristics of the robust/resistant approach using a lead deuterium orthophosphate

data set. The details of the investigation are described elsewhere.^{2,6} A second option is the biweight W-function $W(x) = (1-x^2)^2$ ($|x| \le 1$); = 0 (otherwise).

RFINE4 with the Tukey biweight W-function is being used to repeat the refinements of the structure of D(+) tartaric acid originally carried out by Hamilton and Abrahams' as part of the single-crystal intensity project of the International Union of Crystallography. Three of the 17 experiments considered by Hamilton and Abrahams, selected on the basis of reasonably large numbers of reflections, have been analyzed so far. The refinement for each data set has three stages: 1) attempt to "recreate" the results of Hamilton and Abrahams by repeating as closely as possible the condition of their refinements; 2) add secondary extinction to the classical weighted least squares refinement; and 3) incorporate the biweight W-function and do a robust/resistant refinement. The results of Hamilton and Abrahams are not reproduced exactly, possibly because of small differences in the handling of atomic scattering factors in the original refinement. Inclusion of the Zachariasen model for secondary extinction significantly improves the refinements of all three experiments. Inclusion of the biweight function again significantly improves the refinement with weighted r indices decreasing by up to 40%.

Based on the refinement of these three experiments, the robust/ resistant approach looks very promising. Individual atom positions and thermal vibration parameters agree more closely than in the original analysis. For each experiment, a small fraction of the data fail to fit the robust/resistant estimated model and, hence, are identified as extreme outliers. These outliers can be identified on theoretical grounds as very imprecise reflections. For example, for one experiment the extreme outliers were mostly weak reflections. Such residual patterns suggest that the biweight approach succeeds in minimizing the influence of poor data on the refinement. The above work was reported at the 1976 winter meeting of the American Crystallo-

graphic Association, Clemson, SC. A written version is being submitted for open literature publication.

Work is now in progress on refinement of the remaining experimental data sets reported by Hamilton and Abrahams. Several of their refinements give very discrepant individual atom parameter estimates. Elimination or a least reduction of these discrepancies would establish robust/resistant techniques as a viable approach to crystal structure refinement.

A FLAT-CONE DIFFRACTOMETER UTILIZING A POSITION-SENSITIVE DETECTOR

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The use of flat-cone geometry to take advantage of linear position-sensitive detectors for single crystal neutron diffractometry was described in last year's progress report.¹ An instrument employing these principles is under construction. The detector itself has been received and tested, and most of the electronic components are in hand. Work on hardware and software is proceeding.

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CONVERSION OF THE SPECTROMETER CONTROL SYSTEM TO THE DECLAB 11/40

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and

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It was apparent several years ago that the existing data acquisition system for controlling beam tube experiments would be inadequate for controlling the new 4-axis spectrometer being installed at BT-4. More recently, various developments made it desirable to phase out the existing system, which was installed in 1967, and which was based on obsolete equipment that was becoming more and more difficult to maintain adequately. After extensive consideration of the alternatives, the new system chosen was a Digital Equipment Corporation DECLAB 11/40, supplemented by an industrial control subsystem (ICS-11) provided by the same manufacturer. A major effort to adapt a DEC supplied operating system (RSX11M) so that it could handle spectrometer and diffractometer operations was nearly complete at the end of the year. Most of the task of adapting user programs to the new system was also complete, and the work of rewiring the instruments was proceeding. It is anticipated that the conversion will be complete well before the end of calendar year 1976.

The new operating system possesses much greater flexibility than the existing system. It can accommodate an arbitrary number of motors for each instrument, and also an arbitrary number of detector scalers. Furthermore, it is now possible to do program development, including compiling and linking of programs and editing of source text, without interrupting the data collection process.

THE NEUTRON RADIOGRAPHY PROGRAM

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and

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The goals of the Neutron Radiography Group of the Reactor Radiation Division are to develop neutron radiographic standards, techniques and hardware and to participate in the National Bureau of Standards Nondestructive Evaluation Program. In these capacities, the group provides assistance to both the public and private sectors in the development of neutron applications.

Accomplishments of the Neutron Radiography Group during the past year consist of a number of collaborative efforts with outside agencies in the public sectors. Internal efforts have been devoted to the development of three dimensional laminagraphy, three dimensional image enhanced laminagraphy, extensive beam diagnostics on the thermal column, modifications to the thermal column beam facility and fabrication of the new vertical beam facility. Preliminary conceptual design work has been completed on the external hardware for both the thermal and fast vertical facilities.

1. Three-Dimensional Inspection by Thermal-Neutron Laminagraphy

Radiographic inspection of thick, complex objects presents problems because overlaying radiographic shadows from different object depths complicate interpretation and because it is often difficult to assign an object depth to an observed shadow. Three-dimensional (3-D) radiographic methods including computerized image reconstruction optical and holographic methods, and multiple-film laminagraphy mimimize these inspection problems. In this work, multiple-film laminagraphy has been demonstrated with thermal neutrons, thereby expanding the capability of neutron radiography to inspect complex objects.

In this technique, radiographic views from several angular orientations are obtained and superimposed to bring a desired object image plane into registration (focus) while information from other planes is blurred. New developments reported here include the use of thermal neutrons as the imaging radiation and the rotational movement of the object-detector assembly as opposed to the usual translational source movement to obtain the angular views.

One object used to demonstrate the method was a simulated EBR-II fast reactor fuel subassembly. A hexcan, 5.7 cm across the flats, normally holds 91 rod-shaped fuel elements in 11 rows. For our tests the can was filled with solid aluminum rods and occasional Bakelite rods. These neutron attenuating rods were used to demonstrate spatial resolution at various locations in the assembly. To aid in that determination several through holes varying in size from 0.5- to 2.5- mm diam were drilled in each Bakelite rod.

Thermal-neutron radiograph of the fuel subassembly were obtained over a 40-deg angular coverage, with a 5-deg rotation between exposures. These nine individual neutron radiographs were viewed superimposed to focus on any desired plane. Since it is necessary to view through all the films, a light exposure on each film (typical density 0.4) and a clear base are desirable. This was accomplished by one of two methods. In one approach the neutron radiographs were obtained on a clear base x-ray film. In the other, neutron radiographs were obtained with conventional x-ray film; the films were then copied on clear-base graphic arts film (Kodak types 6127 and 4135 were used). In both cases, neutron radiographs were prepared with gadolinium vapor-deposited screens in vacuum cassettes.

Figure 1 pictures example of two laminagraphic views from these tests. The results show that one can focus on individual fuel rows and that detail as small as 0.5 mm can be resolved in the hexcan assembly. In tests with a high-contrast cadmium test object, a spatial resolution of 0.25 mm was obtained for a similar thickness object.



Figure 1. Photographs of 9-film thermal neutron laminagrams of an EBR-II fuel subassembly hex-can containing aluminum and Bakelite rods instead of fuel. View a focuses on a plane adjacent to one hex-can face and shows a single Bakelite rod in focus; four holes 2.5 mm, 1.5 mm, 1.0 mm and 0.5 mm (left to right) are detectable. View b focuses on the middle row of simulated fuel rods; one Bakelite rod is in focus. Three holes (1, 1.5 and 2.5 mm in diameter) are visible on the print. Note the different images of the radiographic markers at the left of each view. These two laminagrams were obtained with one set of neutron radiographs.

Three-dimensional thermal-neutron radiography with a spatial resolution better than 1 mm can now be accomplished with a technique requiring minimum equipment investment. This broadens the application of thermal-neutron radiography. The 3-D method will prove useful for assembly inspection. Additional capabilities of isotopic differentiation and radioactive material inspection make this neutron laminagraphic method particularly attractive for nuclear problems.

2. Film Investigations

Work has continued on the investigation of the suitability for neutron radiography of other emulsions than the standard x-ray emulsions. Most of this work has been done with an electron-sensitive emulsion developed for transmission electron microscopy. This film is now produced on a regular basis by Eastman Kodak as Type SO 163. It has from two and a half to three times the speed of Type SR, depending on the development. Qualitatively the resolution approaches that of SR, but a radiograph on SO 163 does not have the "snap" of one of the same density done on SR. The ratio of neutron sensitivity to gamma ray sensitivity is definitely better than that for the double emulsion x-ray films.

We have also looked at the possibility of using graphic arts films for neutron radiography. Generally these are high contrast emulsions with extremely fine grain with resulting high resolution. They are also single emulsion on a clear base. With proper development they will produce a gray scale satisfactorily. Preliminary work indicates that the ones tried so far are much too slow when used with a gadolinium foil converter to be useful. There does appear to be a possibility that they can be used for certain applications with a scintillating screen such as ⁶LiFZnS:Ag or Gd_2O_2S :Tb. They may also be useful for neutron laminagraphy where low density and a clear base are required.

3. Image Enhancement

A Spatial Data Image Enhancement system has been installed and placed in operation in the image analysis laboratory of the Neutron Radiography Group. The purpose of this unit is to perform various operations on the radiographic image which causes subtle details to become more easily visible to the eye. This image enhancement system operates strictly in the analog mode, utilizing a standard video camera and monitor. The signal from the video camera is routed to the enchancement circuitry which manipulates the image in such a way that it can be dis-

played in one of three ways, i.e. (a) edge enchancement in which the first derivative of the black and white image is displayed, (b) a normal enlargement of the original image is displayed and (c) color enhancement in which the shades of gray of the radiograph are divided into the hues of twelve colors. A standard radiographic image is illustrated in figure 2. Color enhanced and edge enhanced images of this radiograph are illustrated in figure 3 and figure 4 respectively.



Figure 2. Normal neutron radiograph, no image enhancement.



Figure 3. Neutron radiograph, edge enhancement mode.



Figure 4. Neutron radiograph, color enhancement mode.

4. Image Enhanced Laminagraphy

A successful demonstration has been made to adapt the color image enhancement system to three dimensional laminagraphy viewing. The video camera of the image enhancement system views the laminagraph image as the depth below the surface of a test object is viewed. As the layer image comes into view, the enhanced image of the structure under observation is displayed on the video output of the enhancement system.

The method can be employed to view complicated structures or it can be employed to view in three dimension microstructure below the surface of components. Figure 5 illustrates microshrinkage below the surface of a stainless steel casting covering a depth of approximately 1/4 in. in equal depth steps.



Figure 5. Color enhanced laminagraph microshrinkage in stainless steel casting showing the shape of the cavity at various depths below the surface.

5. Pacemaker Studies

The Food and Drug Administration requested the group to investigate the feasibility of using neutron radiographic techniques to inspect pacemakers and the individual components used in their manufacture. Specifically, the FDA hoped to have developed a method for detecting leaks between the insulation surrounding the electrodes and the seal entering the pacemaker module. A second series of studies was to be conducted on neutron radiography of mercury and lithium iodide batteries used to power the pacemaker.

a. Seal Leakage Studies

The pacemaker was inspected by neutron and x-radiography in an attempt to visualize water that had been injected between the module seal and the electrode insulation. Neither technique could be used to visualize details of the water. In the case of neutrons, the hydrogen content of the insulation was probably very close to that of the water. In the case of x rays, the density of the insulation was possibly very close to that of water.

b. Mercury Cell Studies

A set of two new and four depleted mercury cells were radiographed with neutrons and x rays. Two views of the cells were taken for each specimen - one view along the cell axis and the other at right angles to the cell axis. A schematic diagram of a typical cell is illustrated in figure 6.

In the case of neutron radiography good detail could be visualized in the views taken down the cell axis; some detail could be visualized but not as satisfactorily at right angles to the axis. In both cases, migration of the mercury toward the cell axis could be observed in the depleted cells, but not in the unused cells. The new cells could unambiguously be identified by the absence of the mercury migration.



Figure 6. Schematic diagram of mercury cell for pacemaker application.

The mercury cells were x-rayed at an optimum energy. Although no detail could be observed in the right angle shots, mercury migration could be observed in the axial x-rays of the depleted cells.

The possibility of a microbeam neutron or x-ray transmission measurement along the cell axis to determine remaining battery life remaining in the cell appears to be a real one. No method exists to produce this specification at this time. A microbeam gamma ray backscatter gauge could possible be considered for insitu measurements.

c. Lithium Battery Studies

Lithium batteries are being seriously considered as power sources for pacemakers because of their long life - 13 years as compared to 5 years for a mercury cell. X-radiography is unsatisfactory for lithium battery evaluation because of the double stainless steel case and the relativley low atomic number of the materials used in the electrolyte.

With neutrons it was possible to observe a phenomenon which appeared to be a function of cell life. This apparent electrolyte migration or





gas pocket production is not understood at this time. The cell is fabricated in a slab geometry consisting of a rectangular lithium slab sandwiched between two slabs of iodine. As the battery becomes depleted, lithium iodide is formed causing no change in the total attenuation to the neutron beam.

A series of four lithium batteries was subjected to use tests ranging from 10% to 105% of the expected life. Neutron radiographs of the four units are shown in figure 7. It can be seen that as the use factor increases, the formation of gas pockets appears to take place. Further study on this project will continue once a set of new batteries from the same manufacturing batch are received from the FDA.

6. Pair Production Generated In Objects Under Inspection

A lack of contrast sensitivity has been observed from film fogging when thin Al (a few mils) window cassettes are used. We believe it is due to pair production products generated in the object under inspection. Presumably the radiation products generated do not possess great penetrating power in Al since the effect caused by an object in front of the cassette appears to disappear when aluminum of 0.125 in thickness is employed as an entrance window. We attribute the effect to the high energy prompt gamma rays resulting from radiative capture of thermal neutrons in D_2 (6.2 MeV) and graphite (4.4 MeV). Experiments are in progress to determine the cause of the phenomenon and the effect it may have on cassette design.

The magnitude of the film fogging is illustrated in figure 8 which is a copy of a radiograph made in a neutron-free beam, i.e., when the thermal column curtain was in the closed position.



Figure 8. Film fogging due pair production in specimen under inspection.

7. Standard Measurements

Preliminary design work has been started on two standard measurements which will make it possible to compare individual neutron radiographic facilities. These standard measurements are:

1. Thermal Neutron to Gamma Ray Ratio

A means to measure the thermal neutron to gamma ray ratio of a neutron beam for radiographic purposes. At the present time, these measurements are made using a gold foil to measure the thermal neutron flux and TLDS to measure the gamma content. The gamma measurement is the most inaccurate since the presence of the neutrons has an effect on the readings.

The effect of the gamma ray content on the image recorder is the important factor to be considered, and not necessarily the mr content. Both the mr and the energy spectrum of the gamma beam will have an effect on the gamma film fogging.

We propose to accomplish this measurement using a standard package which will consist of a film in contact with a Gd converter screen sufficiently thick to stop the neutron beam. This will be backed by an aluminum filter sufficiently thick to stop the electrons generated by the Gd. A second film will be employed to determine the effect of the gamma ray content of the beam plus the effect of knock on protons created in the film base by fast neutrons. The ratio of these two numbers at the same front film density would be the meaningful number for comparison of facilities.

2. Beam Divergence Meter

One requirement that has been presented to the NBS Neutron Radiography group has been that of measuring the new beam divergence of a radiographic facility. Preliminary design work has been started on such a device which essentially measures the L/D ratio of the beam. The concept is illustrated in figure 9.



Figure 9. Schematic diagram of the beam divergence meter.

8. Facilities

Extensive modifications to the thermal column neutron radiography facility. Swellage of the magnetite concrete outer shield caused the collimator to shift, directing the neutron beam into neighboring experiments. A new outer shield box fabricated from 0.5 in. thick welded steel plate and filled with lead has been installed. Larger beam pipes have been designed into the new system to produce a larger beam and to run cryogenic cooling to crystal beam filters.



Figure 10. The components comprising the meter consist of a pancake fission beam monitor, two single zone plates of Gd, and the distributed neutron source. As C is moved in the direction of the source, the counts above background in the stop when the neutron beam can no longer be seen by the fission chamber. The distance x can then be used as a measure of the beam divergence.

Pinhole radiographs of the thermal column aperture have exhibited extensive haloing at the image plane. Further diagnostic work will be necessary to determine the cause of this effect.

A problem has arisen in that gamma scatter from the object being radiographed raise the background to an unacceptable level. A new lead lined cave is being designed to eliminate this problem. In addition, the design of a gravity-operated shutter mechanism for the thermal column facility A schematic diagram of the proposed shield and shutter is illustrated in figure 10.

THERMAL NEUTRON FLUX MEASUREMENTS

V. W. Myers and M. Ganoczy

Analysis of the data for calibrating boron loaded cobalt glass beads in a known thermal neutron beam is being completed. An unknown thermal flux can then be determined by comparing the Co-60 counting rate from a bead activated in the unknown flux to a calibrated bead.

SOLUTIONS OF THE KLEIN-GORDON EQUATION

V. W. Myers

The time-independent Klein-Gordon equation for a particle of rest mass m is $(E-V)^2 \psi = (m^2 - \nabla^2) \psi$. This equation is valid in the classical field limit for a spin zero particle, for example a I meson in an external field. Solutions for ψ such as e^{-r} , e^{-r^2} have been investigated to determine if they conform to a real potential V over all space $0 \le r \le \infty$. There are rather stringent limitations on solutions because of the presence of $(E-V)^2$ rather than E-V of the nonrelativistic Schrödinger equation. For example, e^{-r} corresponds to a real potential over all space, but e^{-r^2} does not. There are similar restrictions on solutions of the Dirac equation.

SEPARATION OF OVERLAPPING PEAKS IN NEUTRON DIFFRACTION POWDER PATTERNS

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and

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1. Method

Important applications of powder diffraction data, such as quantitative analysis and determination of lattice parameters, lattice distortions, crystallite sizes etc., are possible only if the patterns contain a large number of single, non-overlapped peaks. This requirement constitutes a serious limitation in most cases and, not surprisingly, attempts have been made to circumvent the problem. In the case of x-ray diffraction the possibility of separating overlapping peaks has been analyzed in detail, especially by Parrish and co-workers. A major difficulty in the use of x-ray patterns is due to the fact that the x-ray peak shapes are not well known and therefore cannot be accurately described by means of a simple mathematical function. However because the neutron diffraction peak shapes are nearly gaussian, the treatment of overlapping reflections relatively simple in this case. The use of neutron powder patterns for the characterization and the analysis of the crystalline components of Portland cements has been considered in this laboratory for this reason. With this approximation the total intensity y, observed at the angular position $2\theta_i$ is given by the expression

$$y_{i} = \sum_{n=1}^{m} y_{i,n} = \sum_{n=1}^{m} \left\{ a_{n} \left[1 - p \frac{\left(2\theta_{i} - 2\theta_{n}\right)^{2}}{\tan \theta_{n}} s \right] \exp \left[-b_{n} \left(2\theta_{i} - 2\theta_{n}\right)^{2} \right] \right\}$$

where $y_{i,n}$ is the contribution of reflection n to the intensity measured at $2\theta_i$, θ_n is the Bragg angle of reflection n, a_n the peak height and $b_n = (4\ln 2)/H_n^2$ where H_n is the full width at half maximum (fwhm). P is







Figure 2. Comparison between the theoretical and the experimental values of the full widths at half maximum of neutron diffraction peaks. The "experimental" points of figure 1a. have been obtained by least-squared fitting of the peaks of KBr by assuming gaussian shapes. The points of figure 1b. are those given by Caglioti et al. (loc. cit.). The broken lines represent the theoretical preditions according to the equation $H^2 = Utan^2\theta + Vtan\theta + W$. The continuous lines are the leastsquares lines through the experimental points. The parameter a is given by a = $tan\theta/tan\theta_M$ where $2\theta_M$ is the take-off angle

the so called "asymmetry parameter", introduced to correct for the distortions caused by the vertical divergence and s = -1, 0, +1, depending on the sign of $2\theta_i - 2\theta_n$. m is the number of reflections contributing to the intensity y_i . A least-squares program has been written which minimizes the function

$$F = \sum_{i}^{K} W_{i} \left[y_{i} (observed) - y_{i} (calculated) \right]^{2}$$

with respect to the parameters H, a, 2R and P of each reflection. In the above expression W_i are weights, assigned according to the expression $W_i = 1/(y_i + background)$. The main difference between Rietveld method (*J. Appl. Cryst.*, <u>2</u>, 65, 1969) and the one described here is that the latter does not require any structural information.



Figure 3. Lattice constant of KBr calculated from the indicated reflections and plotted against the function $(\cos^2\theta)/\sin\theta + (\cos^2\theta)/\theta$. The extrapolated value of the lattice parameter - a = 6.596 - is in good agreement with the literature values which range between 6.596 and 6.599.

2. Results

Examples of the application of the least-squares program are illustrated in figures 1a, b, c, and d for cases of increasing degree of overlapping. The data obtained after refinement are given in table 1 and compared, whenever possible, with the theoretical predictions.

The powder pattern of KBr contains good quality, non overlapped peaks. Therefore, a detailed analysis of these peaks has been carried out in order to clarify a number of questions related to the profile fitting procedure proposed by Rietveld and now widely used. Preliminary results of this study seem to suggest the following conclusions:

a. The neutron diffraction peaks are very nearly gaussian. The analysis of all the peaks, however, shows that with our experimental arrangement the least-squares fit results in a value of the R_W factor around 4-5%. This means that the so called "expected" R factor cannot be evaluated on counting statistics alone and that it is unrealistic to expect structural refinements with R_W factors lower than 4-5%.

b. There is a discrepancy between the theoretical values of the full widths at half maximum (fwhm) calculated with the procedure of Caglioti

Table 1. Results from the least-squares separation of overlapping peaks

	²⁰ LS	²⁰ THEORY	^H LS	H _{THEORY}
KBr	20.56° 23.78	20.53 23.76	30 ' 29	27 ' 26
$R_{W} = 4.4\%$				
25Li ₂ 0:75Ti0 ₂	31.34 32.12 32.99		28 38 31	26 26 26
$R_{W} = 3.6\%$				
M-LiTa308	20.62 21.51 22.08	20.63 21.51 22.05	32 32 32	27 27 27
$R_{W} = 8.2\%$				
c ₄ AF+β-c ₂ S	35.45 36.39 36.94		33 35 37	26 26 26
$R_{W} = 5.2\%$				

et al. (*Nucl. Instr.*, <u>3</u>, 223, 1958) and the experimental data. The nature of the discrepancy is illustrated in figures 2a, b. These figures show that the functional relationship between fwhm and dispersion parameter seems to hold. It is clear, however, that more careful analysis of the equation relating the fwhm and θ is required before we can apply with any confidence the equation to a profile fitting procedure such as that of Rietveld.

c. In figure 3 the lattice parameter of KBr obtained from each diffraction line is plotted against the Nelson and Riley extrapolation function (Klug & Alexander, "X-Ray Diffraction Procedures," 2nd Edition, Wiley, 1974, p. 594). While the choice of this function may not be quite appropriate for our particular case, the figure shows clearly that the peak positions are affected by systematic errors as in the x-ray case. The nature of the systematic errors does not have to be necessarily the same in x-ray and neutron diffraction, however. For example, the vertical

component of the eccentricity error cannot be eliminated experimentally in the neutron patterns (as it is done in the x-ray patterns) because this would require measurements of peak positions in the unfavorable anti-parallel geometry. According to our preliminary results, this error unfortunately varies with θ according to cos2 θ cotan θ , making it difficult to apply the ordinary extrapolation procedures. Another significant difference between x rays and neutrons is that in the latter case the problem of absorption is only rarely important, while in the former case is the major source of errors. The refinement procedure of Rietveld does not take into account systematic errors in the lattice parameters. The above results strongly indicate that this is another area where further study is required.

POWDER NEUTRON DIFFRACTION ANALYSIS OF THE CRYSTAL STRUCTURES OF LiNb308, M-LiTa308 AND H-LiTa308

A. Santoro

and

R. S. Roth and D. B. Minor (Inorganic Materials Division)

The principles of the crystal chemistry of Li⁺ in oxide structures is largely unknown, as this ion generally cannot be located with any accuracy by x-ray diffraction methods. However, neutron diffraction offers unique advantages over x rays in scattering of both Li⁺ and oxygen and opens up the promise of learning some of the details of the Li-oxygen coordination scheme. The compound LiTa₃0₈ was found to be trimorphic (Roth et al. in Fast Ion Transport in Solids, p. 218, Ed. W. vanGool, North Holland Publish Co., Amsterdam, 1973). As the crystal structures of all the polymorphs are known in principle, it appeared that these three phases might be appropriate to use as a starting point in this study. However, the lowest temperature polymorph has never been synthesized as

a pure single phase, so the apparently isostructural compound $\text{LiNb}_{30_8}^{0}$ was substituted. Stoichiometric $\text{LiTa}_{30_8}^{0}$ was prepared from LiTa0_{3}^{0} and $\text{Ta}_{20_5}^{0}$, and converted to the M- and H-polymorphs by heating at 1025°C and 1200°C respectively. The $\text{LiNb}_{30_8}^{0}$ was prepared from LiNb0_{3}^{0} and $\text{Nb}_{20_5}^{0}$ by reaction at 1100°C. All heat treatments were performed in open containers of pure platinum.

Neutron diffraction measurements were made at room temperature on a powder diffractometer of moderate resolution at the National Bureau of Standards reactor. The details of the experimental conditions are summarized in table 1.

Table 1. Experimental conditions used to collect powder diffraction data

Monochromatic beam: reflection 220 of a Cu monochromator Mean neutron wavelength = 1.358(1)Å

Take-off angle = 63.8°

Horizontal divergences: (a) In-pile collimator α_1 =21 min. arc

(b) Monochromatic beam collimator $\alpha_2 = 24$ min. arc

(c) Diffracted beam collimator $\alpha_3=23$ min. arc

Monochromator mosaic spread ~10 min. arc Theoretical values of full width at half maximum (fwhm) parameters: (*)

 $U = 7200, V = -4650, W = 2420 \times 10^4 \text{ deg}^2$

Sample container: Vanadium can of ~ 1 cm. diameter

(*) The parameters U,V,W appear in the expression

 $H_n^2 = Utan^2\theta_n + Vtan\theta_n + W$ where H_n is the fwhm of reflection n with Bragg angle θ_n . Their theoretical values are calculated with the expressions given by Caglioti et al. (Nucl. Instr., <u>3</u>, 223, 1958). The structural models of the title compounds were refined by using the profile fitting procedure proposed by Rietveld (J. Appl. Cryst., <u>2</u>, 65, 1969).

Starting Models

a. $LiNb_{3}0_{8}$ - The structure of lithium triniobate has been determined by single-crystal x-ray techniques by Lundberg (Acta Chem. Scan., <u>25</u>, 3337, 1971) and by Gatehouse & Leverett (Cryst. Struct. Comm., <u>1</u>, 83, 1972). It has been re-considered in this work not only to provide a useful comparison between x-ray and neutron diffraction results, but also to test if the total profile analysis proposed by Rietveld can be extended to the refinement of crystal structures of the complexity encountered in the study of this class of compounds. The results of the refinement, reported in table 2, show that the method can be safely applied to the study of oxide structures. Figure 1 shows the calculated and observed profiles for this compound.

b. M-LiTa₃0₈ - The x-ray analysis of the intermediate temperature phase of lithium tantalate (Gatehouse, Negas & Roth, J. Solid State Chem. <u>18</u>, 1, 1976) failed to reveal the exact location of the Li atoms. An attempt made by powder neutron diffraction techniques showed immediately that the lithium ions are located in the octahedral sites indicated by the idealized structure represented in figure 2. The results of the profile refinement are summarized in table 3.

In figure 3 are shown the calculated and observed profiles for M-LiTa₃0₈. A comparison between the atomic coordinated obtained by single-crystal x ray analysis and by powder neutron diffraction is shown in table 4.

Table 2. Results of the refinement of the structure of LiNb 308 $LiNb_{3}0_{8} -P2_{1}/c$, a = 7.457(2), b = 5.033(1), c = 15.270(2) Å, β = 107.34(1)° $2\theta_{initial} = 6.00^{\circ}, 2\theta_{final} = 64.50^{\circ}, step = 0.1^{\circ}$ No. of reflections = 269, no. of observations above background = 519 Scattering amplitudes: b_{Nb}=0.711, b₀=0.575, b_{1.i}=-0.194x10⁻¹²cm U = 15700(1400), V = -11600(1100), W = 4300(200) $R_{p} = 6.3\%, R_{pw} = 7.6\%, R_{expected} = 4.2\%$ (**) B(*) Atom \mathbf{x} У z Nb1 0.493(2)0:228(3)0.331(1)0.56 0.60 ΝЪ2 0.751(2)0.232(4)0.073(1)0.56 Nb3 0.987(1) 0.247(4)0.333(1)0.74 01 0.998(2)0.055(4)0.098(1)02 0.413(3)0.079(4)0.216(1)0.74 0.096(3)0.345(1)0.74 03 0.767(3)0.419(4)0.105(1)0.74 04 0.539(2)0.74 0.418(5)0.216(1)05 0.911(3)0.74 06 0.641(2)0.384(3)0.461(1)0.74 07 0.149(2)0.071(3)0.453(1)0.74 80 0.275(3)0.417(4)0.345(1)0.082(4)2.3 0.245(9)0.245(20)Li

(*) The isotropic temp. factors were kept fixed at the values found by x rays.

(**) For the definitions of R_p, R_{pw} and R_{expected} see Hewat, ILL report #74H62S, April 1974.

Table 3. Results of the refinement of the structure of M-LiTa308 $M-LiTa_{3}O_{8} - C2/c$, a = 9.409(1), b = 11.519(1), c = 5.050(1) Å $\beta = 91.11(1)^{\circ} 2\theta_{\text{initial}} = 9.00^{\circ}, 2\theta_{\text{final}} = 78.00^{\circ}, \text{ step} = 0.1^{\circ}$ No. of reflections = 247, no. of observations avove background = 641 $b_{Ta} = 0.691 \times 10^{-12} \text{cm}$ U = 15000(1100), V = -10900(900), W = 4300(200) $R_{p} = 6.1\%, R_{pw} = 7.3\%, R_{expected} = 5.3\%$ R(*) Atom z х y Tal^(*) 0.2415 0.0864 0.2466 0.27 Ta2^(*) 0 0.3299 1/40.27 0.4386(11)01 0.3533(5) 0.4502(5) 0.62 02 0.3839(5)0.1794(5)0.4226(10)0.61 0.1322(5)0.4366(5) 0.0834(12)0.48 03 04 0.1187(5)0.1961(4)0.1050(10)0.64 0.8357(20) 1/40.70 0 Li

(*)All thermal parameters and the positional parameters of the Ta atoms were kept fixed during refinement.

c. H-LiTa₃0₈ - The structure of the high temperature phase of LiTa₃0₈ has not been determined previously, but lattice parameters measurements indicated that the structures of H-LiTa₃0₈ and LiNb₆0₁₅F (Lundberg, Acta Chem. Scan., <u>19</u>, 2274, 1965) must be closely related. A refinement based on this assumption gave a final R factor of 15%, i.e. significantly higher than the expected value. These preliminary results indicate that the gross structural features of the two compound are the same. The high value of the R factor, however, suggests that the symmetry of H-LiTa₃0₈ may be lower than that of the space group Pmam reported for LiNb₆0₁₅F. Single-crystal measurements do in fact show that many reflections h0% with h = 2n + 1 have appreciable intensity, thus indicating that the structure does not contain the glide plane. Further study on the structure of this compound by neutron single-crystal and powder techniques is still in progress.







Figure 2. Layers of octahedra which build up the structure of M-LiTa308

Table 4. Comparison of x-rays and neutron atomic parameters for oxygen and lithium atoms in the structure of $M-LiTa_{3}O_{8}$

X RAYS			NEUTRONS					
Atom	x	У	z	В	x	У	Z	В
0(1)	0.3541(20)	0.4516(17)	0.4413(37)	0.62(28)	0.3533(5)	0.4502(5)	0.4386(11)	0.62
0(2)	0.3846(22)	0.1788(17)	0.4270(39)	0.61(27)	0.3839(5)	0.1794(5)	0.4226(10)	0.61
0(3)	0.1332(20)	0.4375(18)	0.0792(36)	0.48(26)	0.1322(5)	0.4366(5)	0.0834(12)	0.48
0(4)	0.1185(22)	0.1967(18)	0.1069(38)	0.64(28)	0.1187(5)	0.1961(4)	0.1050(10)	0.64
Li	-	-	-	-	0	0.8357(20)	1/4	0.7





APPLICATION OF SIMULTANEOUS DIFFRACTION STUDIES TO THE ANALYSIS OF X-RAY SPECTRA

A. Santoro

and

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1. Introduction

X-ray spectra from the sun are now being intensely studied as they are rich in structural details of importance for the understanding especially of flare phenomena. As the time of maximum solar activity approaches, large scale plans for sophisticated multi-spectral measurements are well underway. One of these, the Solar Maximum Mission (SMM), a 300 kg satellite containing more than a dozen spectrometers, is being pursued under NASA sponsorship by a group at Mullard Industries and Appleton Laboratories in England and Lockheed Research Laboratory in Palo Alto, California.

This group has asked the NBS for assistance in the quantitative characterization of the dispersion crystals and for comment on the crystallographic choices which must be made. X-ray spectroscopic resources (232.06) provided help in optimizing diffraction characteristics for the several spectral features to be studied on SMM. One additional feature of crystal choice, namely the avoidance of simultaneous diffraction, was studied at the Reactor Division (314.03).

Simultaneous, or multiple, diffraction of x-rays takes place whenever two or more sets of crystal planes are in position to diffract the incident beam. The occurrence of this effect produces complicated variations in the measured intensity of reflections (Zachariasen, *Acta Cryst*. <u>18</u>, 705, 1965) and it has to be avoided in structural investigations (Santoro and Zocchi, *Acta Cryst*. <u>17</u>, 597, 1964; *Acta Cryst*. <u>21</u>, 293, 1966; *Acta Cryst*. <u>22</u>, 331, 1967) as well as in x-ray spectroscopy work.

In what follows we will outline the method employed in this study and we will point out some properties of the diffraction geometry which may be of general interest.

2. Method

The crystal is initially oriented so that the reciprocal lattice vector (scattering vector) corresponding to the selected "primary reflection" $h_{0}k_{0}l_{0}$, is perpendicular to the incident beam. The orientation needed to bring $h_{0}k_{0}l_{0}$ into reflecting position is then achieved by means of the appropriate rotation θ_{0} about the direction perpendicular to the "equatorial plane", i.e. the plane defined by incident and diffracted beams. Obviously the diffraction condition continues to be satisfied also if we rotate the crystal of any arbitrary angle ψ (azimuthal rotation about the scattering vector. Since the crystal orientation, the lattice parameters and the values of θ_{0} and ψ are all known quantities, it is possible to evaluate the matrix T which transforms the coordinates h of a reciprocal lattice point, expressed in the reciprocal lattice system, into the coordinates expressed in an appropriate Cartesian system attached to the laborator/

\sim = Th

From this equation it is easy to derive the geometrical conditions (that is the value of ψ or that of the wavelength λ) under which a second reciprocal lattice point hkl is in simultaneous diffraction with the primary reflection h k l. Starting from the same equation it is also possible to prove a number of theorems which have been found useful in interpreting the geometry of multiple diffraction and in designing experiments which minimize its effects on the measured intensities. These are the following:

a. Simultaneous diffraction may be λ -dependent or λ -independent. (In the first case the angle ψ at which a node hkl is in simultaneous diffraction with the primary reflection h k l is a function of λ , in the second case it is not).



b. Simultaneous diffraction λ -independent takes place if, and only if, the points $h \underset{O O O}{k} 1$ and hkl lie on a vertical net, i.e. on a net perpendicular to the equatorial plane.

c. If $h \underset{O \ O}{k} 1$ and hkl are in simultaneous diffraction λ independent, then also the reciprocal lattice point

$$h' = h_0 - h,$$
 $k' = k_0 - k$ $l' = 1_0 - 1$

diffracts simultaneously with hkl and $h \underset{O}{k} 1$. In addition, the three nodes all lie on the same vertical net.

d. If $h \underset{O}{k} 1_{O}$, hkl and h'k'l' are in simultaneous diffraction λ -independent, the net of these three nodes is rectangular.

e. If a node hkl diffracts simultaneously with the primary reflection $h_{0}k_{0}l_{0}$ at a value ψ of the azimuth, then the node

$$h' = h_0 - h,$$
 $k' = k_0 - k,$ $1' = 1_0 - 1$

diffracts simultaneously with h k 1 at the azimuth

 $\psi' = 180^\circ + \psi$

3. Results

A computer program which evaluates the azimuthal coordinates ψ of multiple reflections has been developed. The values of ψ were calculated for the lower and upper limits of each wavelength interval to be studied and for each dispersion crystal employed in SMM. When these values are plotted as indicated in figure 1, one often finds orientation intervals entirely free of multiple diffraction. When possible, it is obviously desirable to choose such orientations for spectroscopy. Sometimes, however, there are no such regions, as indicated in figure 2, or the desired orientation is not accessible because of growth morphology. For these cases a second program has been developed which enumerates the Bragg angles (or wavelengths) at which trouble might be expected.

Other x-ray spectroscopy programs are benefitting from this work also. For example, the spectra of magnetically confined and laser driven

plasmas are rich in this region and need quantitative spectrophotometry. Work done in support of the ERDA supported studies at the Princeton Plasma Laboratory will include use of this systematic approach to the avoidance of multiple diffraction effects. Finally, an NBS experiment destined for installation at the Stanford Storage Ring was recently troubled by this effect because of a failure to apply the systematic tests in advance.

THE CONFORMATION OF PS-PMMA DIBLOCK COPOLYMER IN TOLUENE BY SMALL ANGLE NEUTRON SCATTERING

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and

B. Mozer

A deuterated polystyrene-poly(methyl methacrylate) (dPS-PMMA) diblock copolymer and its deuterated polystyrene homopolymer precursor were studied by small angle neutron scattering and light scattering experiments using toluene as the solvent.

The radii of gyration of the deuterated PS homopolymer (M=88,000) measured by both methods and that of the PMMA block (M=203,000) in the diblock copolymer measured only by neutron scattering were determined as 210Å and 85Å respectively. In addition, the second virial coefficients and the chain excluded volume exponents were also obtained.

The copolymer chain configuration in toluene at 23°C is inferred from these measurements. It is suggested that the PMMA block forms the interior core from which the PS block exudes out as an expanded chain.

SUMMARY OF EXPERIMENTAL RESULTS

<pre>Second Virial Coefficient A2 (mol. ml/g²)</pre>) (5.2±1.0)x10 ⁻⁴ (LS)			
Volume t v	(NS)		(NS)	(SN)
Excluded Exponen	1.15~1.20		1.03±.05	1.11±.03
'2 (Å)	(SN) (LS)	(IS)	(SN)	
<r<sub>G²,1/</r<sub>	215±18 210±40	225±40	85±7	
Wt.	(I.S.)	[(ST)	(EA)	
Molecular	(88±3)x10 ³	[(95±3)x10 ³	203x10 ³	60.6x10 ³
	dРS (homo)	dPS (block)	PMMA (block)	PMMA (homo)

RRD-NBS COLLABORATIVE PROGRAMS

STUDIES ON THE STRUCTURE OF DEFECTIVE HYDROXYAPATITE

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and

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Structural studies of defective hydroxyapatite using the method of total profile analysis have continued in the past year. The results so far obtained do not permit to determine the nature of the defects present in the structure, mainly because a "standard" sample with which comparisons can be made is not yet available. Attempts are now being made at preparing non defective samples of the compound.

C. INTERAGENCY AND UNIVERSITY COLLABORATIVE PROGRAMS

DEUTERIUM-SITE OCCUPANCY IN THE & AND & PHASES OF TIDX

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and

H. Flotow (Argonne National Laboratory, Argonne, IL)

and

J. Rush and J. Rhyne

A neutron diffraction study on polycrystalline samples of the $\alpha(hcp)$ and $\beta(bcc)$ phases of TiDx has been completed. The objective was the determination of the sites that are occupied by deuterium atoms in each of the two phases. The compositions and temperatures chosen to achieve the α and β phases were TiD_{0.075} at 375C and TiD_{0.67} at 400C respectively. The results show that both phases exhibit considerable mixed - octahedral (y) - tetrahedral (1-y) site occupancy. The results are for the α -phase: y=0.68±.02 while for the β -phase: 0.15<y<0.30. A comparison of observed and calculated intensities are given in Tables 1 and 2. The uncertainty in y for the β -phase is due to a strong correlation with the octahedral deuterium temperature factor. The site occupancy results may be correlated with the Ti-D distances for each site. In both phases it appears that the deuterium atoms predominantly occupy sites with the larger holes.

Single crystals of titanium have been obtained and diffraction measurements will be made to further refine the octahedral and tetrahedral temperature factors. Work is underway using the time-of-flight spectrometer to measure the quasielastic scattering from TiH_x in the α and β phases in order to determine the diffusion of hydrogen in the lattice. Finally we expect to measure the phonon spectra on single
hk1	I _{obs}	I ^a calc
100	51370	53024
002	54575	53068
101	199045	196319
102	21085	23269
110	34498	34694
103	48779	47774

Table 1. Comparison of observed and calculated neutron scattering intensities for $\text{TiD}_{0.075}$ at 375C-- α (hcp) phase.

^aCalculated with $B^{Ti} = 1.875$, $B_D^{oct} = 4.6$, $B_D^{tet} = 3.0$ and deuterium octahedral site occupancy, y = 0.68.

Table 2.	Comparison of	observed a	and calculated	neutron scattering	
	intensities f	or β-phase	(bcc) TiD _{0.67}	at 400C.	

hkl		I _{obs}	I a calc
110		230700	237573
200		4529	5001
211	-	28822	28566
220		24936	23784

^aCalculated with $B^{Ti} = 2.5$, $B_D^{oct} = 5.0$, $B_D^{tet} = 2.5$ and deuterium octahedral site occupancy, y = 0.20.

crystals of pure α -Ti and α TiH_{.075} to obtain information on how the presence of hydrogen affects the metal-metal bonds. This work has already begun with the measurements of several phonon branches for pure α -Ti at room temperature.

SMALL ANGLE MAGNETIC SCATTERING FROM A DILUTE AMORPHOUS Fe(Tb) ALLOY

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and

J. J. Rhyne

Low temperature neutron scattering measurements on a rapidly sputtered bulk sample of composition ${}^{\rm Tb}_{0.018}{}^{\rm Fe}_{0.982}$ reveal anomalous small-angle magnetic scattering similar to that observed in the more rare earth-rich amorphous alloys ${}^{\rm TbFe}_2$ and ${}^{\rm HoFe}_2$ reported previously¹. The data in figure 1 show a broad hump (as distinct from a sharp critical peak) about T_c (determined from magnetic measurements²) and the anomalously large magnetic component below T_c. Above T_c the scattering can be fitted to a Lorentzian from which we deduce a correlation length which increases from 13A at 260K to 38A at T_c. Below T_c the scattering can be fitted by the asymptotic Porod Law for a random distribution of spherical particles.

- * Physics Dept., University of Rhode Island
- S. Pickart, J. Rhyne and H. Alperin, Phys Rev. Lett. <u>33</u>, 424 (1974); AIP Conf. Proc. 24, 117 (1975).
- 2. H. Alperin, J. Cullen, A. Clark AIP Conf. Proc. 29, 186 (1976).



Figure 1. Scattered neutron intensity, background subtracted as a function of temperature. The data at lower temperatures for q = .04, which are not plotted for reasons of convenience, scale very closely to the square of the observed spontaneous moment².

DIFFRACTION MEASUREMENTS FROM RAPIDLY SPUTTERED BULK Fe(Tb) AND Ni(Dy) ALLOY SAMPLES

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and

J. J. Rhyne

We have previously measured bulk samples (50 mm³ to 150 mm³ in volume) of $\text{Tb}_{x}\text{Fe}_{1-x}$ (x = 0.118, 0.25, 0.33, 0.45 and 0.75) alloys pre-



Figure 1. Diffraction pattern of amorphous Tb_{0.018} Fe_{0.982} at room temperature. If the pattern were due to small polycrystalline particles of pure bcc iron the peaks would have to appear at the locations indicated.

pared by dc rapid sputtering. Diffraction patterns were found with very broadened peaks. The atomic 'structure' that can give rise to such patterns has been described as amorphous, arising from a random close packed atomic arrangement. We have now made similar measurements on the compositions Tb_xFe_{1-x} for x = 0.018, 0.167 and also for Ni(1wt.%Dy) which corresponds to Dy_{0.0036}Ni_{0.9964}. The terbuim content in the x = 0.018 sample was analyzed by neutron activation to an accuracy of +2% by the Activation Analysis Section of the Analytical Chemistry Division. The diffraction pattern for this composition shown in figure 1, is most likely also due to an amorphous random close packed atomic arrangement. However, if one should try to attribute the broadened diffraction peaks to small-size crystalline iron particles, these particles could be no larger than an average diameter of 15A if we analyze only the broadening about the (110) Fe position. The displacement of the measured peaks from the (211) and (220) Fe positions however makes even such a crystalline interpretation doubtful. For the x = 0.167composition, if the particles are crystalline they could be no larger The Ni(Dy) sample exhibited sharp diffraction peaks indicative than 10A. of a crystalline atomic structure.

NONDESTRUCTIVE DETERMINATION OF GRAIN-ORIENTATION OF METAL LINERS FOR SHAPED-CHARGE MUNITIONS

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The optimum functioning of shaped-charge ammunition requires that the projectile have essentially zero spin (i.e., axial rotation) at impact with the target. This requirement is very difficult to achieve in practice. It has been generally accepted that at relatively low spin rates ($\frac{1}{2}$ 50 r.p.s.) optimum functioning can be achieved in spite of spin if the liner cone is manufactured in a specific way. That is, by preferentially aligning the individual grains of the liner such that as the liner collapses along preferred slippage directions, a counterrotation equal and opposite to that of the in-flight projectile is imparted to the explosive blast jet. It has also been suggested that residual stresses are induced in liners by the same manufacturing technique which produces grain orientation (i.e., shear spinning), and that it is residual stress, not grain orientation, which causes "spin compensation" in collapsing liners. The principal difficulty which has prevented the resolution of this conflict has been the lack of nondestructive test methods to measure grain orientation and residual stress, which would then allow test firing of the characterized liners.

In the present program, we have explored the feasibility of neutron diffraction as a nondestructive probe for grain orientation determination in intact, copper shaped-charge liners. Initial measurements were performed on a piece cut from a liner which had been flattened and examined by x-ray diffraction. Neutron and x-ray results were found to be comparable. Subsequent neutron diffraction studies were made on twelve intact liners from a current production batch. In figure 1 are shown pole-figure representations of the neutron diffraction results for (111) and (220) planes for two of the copper liners.



CONE I (III)



CONE I (220)





CONE 2 (220)

Figure 1. Pole figures for (111) and (220) grain orientation in intact shaped charge liners from neutron diffraction measurements. The heavy solid contour corresponds to "random" grain orientation; lighter counters correspond to increases in orientation by approximately 30% of random per contour.

These data represent the extremes of the differences observed for the twelve liners, which in the case of (111) reflections are rather significant. If grain orientation is the critical parameter, one would expect the performance of these two liners to be measurably different.

Further studies to establish grain orientation as a function of depth (total thickness \sim 3 mm) and to examine the feasibility of nondestructive residual stress measurements using neutrons are in progress.

REINVESTIGATION OF CRYSTAL STRUCTURE OF $\alpha - P_b(N_3)_2$

C. S. Choi (Picatinny Arsenal, Dover, NJ)

and

E. Prince

Lead azide is a well-known primary explosive. The explosive nature of the azide compound is known to be correlated very closely with the degree of covalency in the azide bond in the crystal. Therefore, it is very important to know the structure of the azide groups and their surroundings accurately in order to understand the explosive properties. The crystal structure of $\alpha - P_{h}(N_{3})_{2}$ was determined previously by using a partial three-dimensional neutron diffraction data set. The present study was aimed at improving the accuracy of the structure by collecting a complete set of three-dimensional data. A total of 1466 independent reflections were measured within the limiting sphere defined by a maximum 20 angle of 106° and a neutron wavelength of 1.226A. With this new set of data, the refinement of the structure converged rapidly to the final discrepancy indices, R = 0.038 and wR = 0.043 for 1328 observed reflections. The structure obtained in this study is essentially the same as that of previous study, except for the detailed values of some parameters and their accuracy. The configurations of the four different azide groups in α -Pb(N₃)₂ are given in table 1. The azide I and azide III are essentially symmetric, azide II is slightly asymmetric and azide IV is quite asymmetric. The azide structure becomes more symmetrical when the bond lengths are corrected for the effect of the thermal vibration by using the riding model.

			0	
Туре	Bond	Bond Ler	ngth (A)	<u>N-N-N Angle (°)</u>
		Raw	<u>Riding</u>	
I	N ₁ -N ₂	1.166(1)	1.179	177.9(2)
II	N ₃ –N ₄ N ₅ –N ₄	1.181(2) 1.160(2)	1.186 1.173	178.2(1)
III	N ₆ –N ₇ N ₈ –N ₇	1.172(2) 1.167(2)	1.181 1.176	179.2(1)
IV	N9-N10 N11-N10	1.191(2) 1.149(2)	1.196 1.164	178.3(2)

Table 1. The structure of four different azides in α -Pb(N₃)₂.

PHASE TRANSITION STUDY OF NaN₃ BY POWDER NEUTRON DIFFRACTION

C. S. Choi (Picatinny Arsenal, Dover, NJ)

Sodium azide undergoes a phase transition at approximately 14 °C from β -phase (rhombohedral R $\overline{3}$ m) to α -phase (monoclinic C2/m) with decreasing temperature. Iqbal and Christoe¹ have studied the phase transition by a method of polarized Raman scattering and concluded that the phase transition is a second order type. Subsequently, they proposed a mechanism for the phase transition based on the soft mode model developed by Chihara et al.;² the azide axis, in the phase transition, gradually changes its orientation with temperature over a wide temperature range, and this uniform tilt of the azide axis is coupled anharmonically to its librational mode in accordance with the formulas given by Chihara et al. The present study was aimed at determining the tilt of the azide axis at various temperature by the neutron powder diffraction method. Using 0.948Å neutrons, powder diffraction data of NaN₃ were collected at four different temperatures; 270 K, 174 K, 78 K, and 4 K, within the scanning range of 8° to 60° in

scattering angle with a 0.1° interval. The portion of the spectrum contaminated by the aluminum container was excluded from the refinement. The structure and lattice parameters were refined by the Rietveld technique of profile refinement using a program modified by Hewat. The profile residuals, R and R_w as defined by Reitveld, are 0.10 and 0.12 for 270 K, 0.067 and 0.114 for 174 K, 0.048 and 0.066 for 78 K, and 0.055 and 0.069 for the 4 K structure, respectively. The structure of α -NaN₃ are very similar to that of β -phase in general atomic arrangement, except for slight distortions due to a shear movement of the lattice (or β -angle changes) and an internal rotation of the azide axis. The tilt of the azide axis is the compounded effect of these two distortions.

Table 1. The distortion parameters of NaN_3 at various temperatures. The parameters are β for the angle of monoclinic cell, Θ for the internal rotation of the azide axis measured from the c-axis, and the tilt angle of the azide axis measured from the normal direction of the cation layer. The 300 K data refer to the equivalent monoclinic cell of the rhombohedral structure.

lemperature	<u>β(°)</u>	Θ (°)	<u>Tilt(°)</u>	Tilt(°) (soft mode Model)
300 K	112.54	22.54	0.0	0.0
270 K	110.7(1)	25.88(4)	5.2	4.4
174 K	108.63(4)	28.80(1)	10.17	9.9
78 K	107.63(1)	29.64(1)	12.01	. 12.2
4 K	107.46(1)	29.94(1)	12.48	12.48

The tilts of the azide axis obtained in this study agree well with those obtained by Iqbal et al. based on the soft mode model as shown in table 1. The rate of tilt is largest at near transition temperature and therefrom it decreases gradually until it saturates at near 78 K.

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QUASI-ELASTIC NEUTRON SCATTERING STUDY OF SINGLE CRYSTAL AMMONIUM PERCHLORATE

N. J. Chesser and H. J. Prask (Picatinny Arsenal, Dover, NJ)

The rotational motions of the ammonium ions in ammonium perchlorate (NH_LC10_L) have been the subject of considerable study for more than a decade, primarily because this salt has been thought to provide a prototype for quasi-free rotation in an ionic solid. An earlier quasielastic neutron scattering (QNS) study of polycrystalline NH_4C10_4 indicated that in the temperature range 78 to 130K the ammonium ions were reorienting via "instantaneous" 120° jumps about C₂ axes of the ion.¹ Thus three protons would move 120°, about an axis containing the nitrogen and fourth proton, to a new configuration identical to the original. A recent crystal structure determination² at low temperature has established that three of the four NH_{λ}^{+} protons are in non-equivalent equilibrium positions and that their thermal amplitudes differ. It would seem likely, therefore, that the jump probability for a proton is site dependent and that a QNS study of single-crystal NH4ClO4 might differentiate the different jump probabilities, and the present report summarizes the results of such a single crystal study. This work represents the most complex material for which QNS has been applied to establish the details of orientational disorder in a crystalline solid.

As a guide to the analysis of our experimental data, we have extended the "instantaneous" jump reorientation model of Skold³ to include the possibility of three different residence times, τ_i , associated with the three non-equivalent hydrogen equilibrium sites² of an NH⁺₄ - appropriate to NH₄ClO₄. This model yields three different Lorentzian contributions and a δ -function contribution to the scattering law. Of these, the δ -function and one of the Lorentzians have structure factors independent of residence times whereas the structure factors of the second and third Lorentzians depend on the relative ratios of the residence times.Based upon average TS obtained from QNS studies of polycrystalline



- Figure 1. Intensity contour map for the hOl and Okl planes of NH4ClO4 at 120K. Areas marked A represent intensities which are, on the average, approximately ten times those of areas marked F. Diamonds indicate the value of Q selected to obtain relatively pure Lorentzian contributions. While these contours are those calculated for the residence times derived from 120K data, they are similar to those derived for all of the temperatures examined.
- Table 1. Activation Energies E_0 and Arrhenius Prefactors τ_0 for each atom.

Hydrogen Site	E _o (kcal/mole)	τ _o (psec)
1	.58±.04	.15±.03
2	.61±.04	.15±.03
3(3')	.52±.05	.28±.03



Figure 2. Residence times derived from analysis of the Lorentzian widths (see text). The lines are obtained under the assumption of an Arrenhius temperature dependence for the residence times $(\tau=\tau_0\exp(E_0/T)$ for each proton. Parameters are listed in table 1. Powder data are from reference 1.

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 NH_4C10_4 and the ratios of librational amplitudes inferred from the low temperature neutron diffraction study² the structure factor contour map of figure 1 is obtained.

Constant Q scans of the quasielastic scattering were made for single crystals³ of NH_4ClO_4 using a triple axis spectrometer configured such that FWHM (ful-width-at-half-maximum) $\simeq 0.1$ meV. Measurements were made at temperatures of 60 to 120K, at the reciprocal lattice points indicated in figure 1.

The data were fit using a non-linear least squares fitting routine with six adjustable parameters: the Lorentzian width, Lorentzian and δ -function intensities, "thermal" (i.e. analyzer efficiency dependent) and "fast" (i.e. flat) neutron background levels, and offset of the peaks from the nominal elastic position. The calculated curves were folded with the appropriate Gaussian resolution function and corrected for analyzer efficiency ($K_f^3 \cot \theta_A$).

From the structure factor map and the limitation that the model requires the width of Lorentzian two to be greater than that of Lorentzian three, residence times for the different sites were obtained at each temperature as shown in figure 2. Assuming Arrenhuis behavior, activation energies and prefactors as shown in table 1 are obtained. The results shown in figure 2 and table 1 clearly indicate the anisotropic character of ammonium uon reorientations in NH_4ClO_4 . Work is in progress to further examine this behavior.

REINVESTIGATION OF THE CRYSTAL STRUCTURE OF $NH_{A}C10_{A}$ AT 298 K

C. S. Choi and H. J. Prask (Picatinny Arsenal, Dover, NJ)

and

E. Prince

A refinement of the structure of ammonium perchlorate at room temperature has been reported by Peyronel and Pignedoli, ¹ who concluded that the structure cannot be refined in the centrosymmetric space group Pnma, but can be refined in the space group Pna21. Since this conclusion is in disagreement with our study at low temperature using neutron diffraction data, we have performed further least-squares refinement of the room temperature structure in the centrosymmetric space group Pnma using their published data. Two cycles of refinement with isotropic temperature factors, followed by three more cycles of refinement with anisotropic temperature factors and an isotropic secondary extinction parameter, reduced the R indices to $R_{1} = 0.044$ and R = 0.049 for the 800 observed reflections. A comparison of this centrosymmetric structure result (R = 0.044 for the 35 parameters) with their acentric structure refinement (R = 0.055) for the 55 parameters) shows clearly that Pnma is the correct space group for NH_4C10_4 at room temperature as it is at lower temperatures. The bond parameters and rigid body motions of the perchlorate group obtained in this study are given in table 1. The librational motion of the perchlorate group, as obtained from the rigid body refinement, is almost isotropic and causes the apparent C1-O bond length to be shortened by about 0.02A. The translational motion, however, is extremely anisotropic and closely resembles the thermal ellipsoid of the nitrogen atom of the ammonium group.

Table 1. The bond parameters and rigid body motions of the perchlorate group.

Bond length (A)	Uncorrected (A)	Corrected (A)
C1-0(1)	1.432(3)	1.452
C1-0(2)	1.424(3)	1.445
C1-0(3)	1.452(2)	1.462
Bond angle (°)	Uncorrected (°)	Corrected (°)
0(1)-C1-0(2)	111.3(2)	111.2
0(1)-C1-0(3)	108.7(1)	108.7
0(2)-C1-0(3)	109.7(1)	109.7
0(3)-C1-0(3)	108.6(2)	108.6

Principal	Amplitudes	Directi	ons based on cr	ystal system
axis	r.m.s. (A)	<u>α (°)</u>	<u>β (°)</u>	<u>γ (°)</u>
T(1)	.314	89.	0	1.
T(2)	.171	1.	0	91.
T(3)	.168	0	1.0	0
	r.m.s. (°)			
L(1)	7.1	0	1.0	0
L(2)	6.9	138.	0	132.
L(3)	6.5	48.	0	138.
<u>N-atom</u>				
U(1)	.325(3)	88.(1)	90	2.(1)
U(2)	.191(3)	90	180	90
U(3)	.179(3)	2.(1)	90	92.(1)

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SMALL ANGLE SCATTERING: INSTRUMENT AND APPLICATIONS

C. S. Schneider (U. S. Naval Academy, Annapolis, MD)

The double perfect crystal diffractometer has been considered for measuring the scattering of thermal neutrons from a Christiansen filter to determine the scattering amplitude^{1,2} of the powder atoms. The scattered intensity, as a function of the difference ΔNb for powder and liquid, changes in form with the phase shift across the particle, $\phi = 2\delta kR = 2\lambda R\Delta Nb$ and the probability of scattering from particles in the sample, $P = N_{p}\sigma_{x}$ where x is the powder thickness. In the single diffraction region $\phi < 1$ and $P \doteq x\phi^{2}/4R < 1$ for a packed powder of particle radius R. Thus

$$I_1 = PfI_o = (\Delta Nb)^2 \lambda^2 xRfI_o$$

where I_o is the central peak intensity and f<l accounts for absorption $e^{-\mu x}$ and slight broadening from the initial resolution to λ/R . When P exceeds unity the central peak is completely diffracted and multiple diffraction begins causing reduction in the intensity seen at the base of the central peak: in region two where $\phi<1$ and P>1

$$I_2 = fI_0/\sqrt{P} = fI_0/\lambda\sqrt{xR} \text{ ANb.}$$

Finally, as ϕ exceeds unity the scattering becomes multiple refractive being broadened through roughly $\lambda^2 \sqrt{P} \Delta Nb/2\pi$ where P = x/R. Thus

$$I_{3} = f' I_{o} / \lambda \sqrt{xR} \Delta Nb$$

continuing the $(\Delta Nb)^{-1}$ decrease with a small change in coefficient f'.

The precision of using the double crystal diffractometer to determine Nb by finding the center of equal intensities in region one from the plot in figure 1, is one part in $2\sqrt{N_0}$ where N₀ is the total counts recorded. In one day's counting using perfect Ge crystals, $\lambda = 0.4$ nm, R = 40 µm and x = 1 mm for highly absorbing powders, the precision approaches a few parts per thousand.

Because a rapidly decreasing tail of the resolution in a diffractometer is necessary to observe small angle scattering, which decreases as θ^4 , Darwin reflectivities are not ideal with their θ^2 decrease soon over powering the sample scattering. Bonse-Hart triple reflection monochromator and analyzers have been made of Ge single crystals with the hope of exploiting their θ^6 tail drop-off, but the modest peak intensities of 10^4 n/min made the advantage marginal. Several modifications have been considered: first, highly asymetric channel cutting (beam incident upon monochromator nearly parallel to the channel) can largely reduce the rocking width but the intensity would stay nearly the same.

Second, inducing thermal gradients perpendicular to the reflecting planes can broaden the integrated reflectivity³ to $\frac{\Delta d}{d} = \alpha \Delta T$ where α is the thermal expansion coefficient and ΔT the temperature difference across the crystal. Due to the small value of α , the large thermal conductivity and the large temperature dependence of α , power densities of 20 W/cm² did not cause a significant reflectivity increase from Si (111) planes. The techniques of adhering a graphite wafer to the back surface and preparing a resistive pathway proved a very effective method of applying several hundred watts to the crystal. Air jet cooling appears sufficient but is not highly uniform.

Finally, strain induced reflectivity⁴ is being intensively studied with a strained perfect crystal spectrometer recently fabricated, with reversible dialable strain normal to the reflecting plane

$$e_{max} = \frac{\Delta d}{2d} \sigma = \frac{\sigma t}{P}$$

existing furthest from the neutral axis of the crystal sheet where σ is Poisson's ratio. Slices of both Si and Ge have been optically polished and their curvatures measured as a function of the strain device micrometer reading, by observing the focussing of a nearly parallel laser beam. Anticlastic (saddle shaped) curvature is 25 percent of the applied curvature between constraint lines. Strain is applied vertically



Figure 1. Intensity scattered by a Christiansen filter as viewed at the base of the central peak in the double perfect crystal diffractometer.

so that the defocussing of the double crystal diffractometer is minimized: two smearing effects are present. First is the permanent smearing due to finite horizontal collimation $\beta_{\rm H}$ and second crystal curvature, $1/\rho_{2\rm H}$:

$$\Delta \theta_1 = \beta_H \times /\rho_{2H}$$

where x is the separation of monochromator and analyzer crystals. Second is the relative curvature smearing which can be eliminated by setting $\rho_{2H} = \rho_{1H} - 2x$, the focus condition:

$$\Delta \theta_{2} = \frac{W_{1}}{\rho_{1H} \rho_{2H}} (\rho_{2H} - \rho_{1H} + 2x)$$

where W_1 is the width of the monochromator crystal. If the smearing is not to affect the induced mosaic, $\Delta \theta = 2ef \tan \theta$ where f<1 is due to dynamical effects⁵ then in the focussed condition for $\rho >> x$,

$$\Delta \theta = fttan \theta / \rho > \Delta \theta_1 = \beta_H x / \rho_H$$

or $t > \beta_H x \cot \theta \rho_V / \rho_H f$. Since $\rho_V / \rho_H \leq .25$ and $\beta_H = 0.01$ and x = 50 cm is ample sample and shielding space, then t > 1.2 mm/f. Present studies on



Figure 2. Intensity reflected from various thicknesses of 18x25 mm area Ge (some imperfect) and Si crystals. Note saturation of thin crystal reflectivity.

piezoelectrically strained crystals^{6,7} should eliminate focussing problems.

Deeper penetration of the beam into a mosaic or strained crystal also requires a minimum thickness to reflect fully. This thickness is roughly one millimeter as above and can be extracted from the intensity measurements made with the Si and Ge crystals on the strain device. The intensity saturates either due to beam penetration of the thin crystals or to yield of the thicker crystals, which was at a strain of roughly

 10^{-3} , corresponding to a few minutes arc mosaic. Preliminary data is shown in figure 2.

Insertion of the 2 mm thick amorphous TbFe₂ sample into the beam of the perfect crystal diffractometer caused severe attenuation, largely from scattering. This suggests the need for the broader resolution of the bent crystal diffractometer. Critical scattering in iron is also under consideration as the very fine q resolution allows direct observation of long correlation lengths.

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STRUCTURE OF THE ANTIFERROELECTRIC PHASE OF COPPER FORMATE TETRAHYDRATE

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and

E. Prince

A three-dimensional neutron diffraction data set was collected from a crystal of copper formate tetrahydrate in the antiferroelectric phase at 77 K. The antiferroelectric phase is characterized¹ by small deviations, in alternate unit cells, from more symmetric positions in the room temperature, paraelectric phase. Consequently, reflections with odd values of ℓ are much weaker, on the average, than reflections with even values of ℓ . Nevertheless, some of the odd ℓ reflections

have appreciable intensity. Attempts to refine the structure using conventional least-squares techniques have met with difficulties, particularly with respect to reflections of the class 5, k, ℓ with ℓ odd. There may be features in the structure not included in the model which have a particularly strong effect on these reflections. Work is continuing on refinement of the structure using robust/resistant methods.

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ACTIVATION ANALYSIS: SUMMARY OF 1976 ACTIVITIES

Harry L. Rook and Thomas E. Gills (Analytical Chemistry Division)

This summary of the activities of the Activation Analysis Section covers the period of July 1975 to June 1976. During this period the Section has continued its efforts to attain a high degree of versatility in activation analysis and has directed efforts toward developing a greater expertise in multielemental analysis particularly coupled with automation and computer data reduction. In addition, research is continuing in the area of the investigation of parameters which affects accuracy and precision in activation analysis.

While the majority of the activation analysis work is in support of the Analytical Chemistry Division's programs, a large portion of the Section's activities this year has been spent in service analyses for organizations outside the Bureau of Standards and for groups within the Bureau of Standards. Such cooperation affords an opportunity to transfer analytical capabilities or expertise to the analytical community.

The Activation Analysis Section has at present eleven members and has programs in a variety of areas related to the trace elemental characterization of materials. These areas include research projects, SRM certification analyses and service analyses. Summaries of the work in these areas are included.

- 1. Basic Research in Activation Analysis
 - a. A Method for the Determination of Iodine in Biological and Environmental Matrices

H. L. Rook

Iodine is an element with excellent intrinsic sensitivity when determined by thermal neutron activation. However, in most real samples,

the preponderance of chlorine and bromine, relative to iodine, makes the direct determination of iodine virtually impossible. Over the past 20 years, there probably have been as many publications on the separation of iodine as there have been for any other radionuclide. Upon review, however, the methods are essentially the same. After irradiation, the samples are subjected to a rapid destructive process to free the iodine from the matrix and then the iodine is separated from the other halides either by liquid-liquid extraction or by liquid ion exchange. Both of these procedures are, however, rather complex and do not effect a complete separation of the halides in one pass. Also, most of the separations are not quantitative in the recovery of iodine, thus, they require a radiochemical yield determination, with its associated problems and additive errors.

The best assessment of the present ability of NAA to determine iodine in real matrices is the published results of iodine in Bowen's Kale and in certain NBS SRM's such as Orchard Leaves, Bovine Liver and Coal. These matrices have been widely studied and numerous publications have appeared with data on their trace element compositions. In Bowen's Kale, no consensus iodine result was obtained, because values reported by cooperating laboratories varied by more than two orders of magnitude. In the NBS SRM's, no iodine values have appeared in open literature, except on Coal, and they appear to be in error.

In the work presented here, a simple, rapid procedure is developed for the quantitative determination of iodine in botanical, biological and environmental matrices.

For the separation of iodine from general matrix activity, the sample is burned in a flowing oxygen atmosphere using a quartz combustion tube, similar to the one described for mercury separations. Following combustion, the sample is heated to 1100° C with a resistance oven to completely distill the volatile elements. During this process, the iodine is separated from the chlorine and bromine, in the gas phase, by a trap containing hydrated manganese dioxide (HMD). A second trap,

of silvered glass wool, is placed downstream from the HMD trap to collect the iodine. The HMD trap, heated to 80° C, has been found to completely remove chlorine while quantitatively passing iodine through to the final collector trap. With this separation system, samples can be processed in 10-15 minutes with separations of greater than 10^{6} for chlorine and 10^{4} for bromine.

The described procedure has been used for the determination of iodine in numerous new and old SRM's at the NBS. Table 1 gives some representative data on SRM 1571, Orchard Leaves. From this work, it is estimated that $<10^{-9}$ g of iodine can be easily determined in real matrices.

Table 1. Iodine Concentration in SRM 1575 Orchard Leaves

Sample #	µg/g Iodine	Sample #	µg/g Iodine	Mean
1	0.176	5	0.160	
2	0.165	6	0.166	
3	0.171	7	0.153	
4	0.180			0.167 ± 0.009

 b. The Measurement of Radial Efficiency Gradients in Ge(Li) Gamma Detectors

R. M. Lindstrom

In the decade since the introduction of lithium-drifted germanium detectors, their availability from commerical suppliers with steadily increasing counting efficiency has led to their nearly universal use in measuring gamma-rays from radioactive substances. The rapid replacement of the sodium iodide scintillation detector for gamma assay is due almost entirely to the forty-fold sharper energy resolution of the newer detectors. In addition to assuring qualitative identification of gamma emitters, this resolution allows a forty-fold reduction of the background

area under a peak, with the result that despite a two to ten-fold decrease in counting efficiency the Ge(Li) detector is still superior to NaI for quantitative determination.

In order to minimize the counting time required for a given statistical precision, the analyst is often impelled to place the source close to the detector, often in contact with the end cap. But, because of the small size of the crystal, small differences in shape or positioning of the counting sample compared to a standard may lead to significant differences in counting efficiency. It is the intent of the present work to examine this position sensitivity where the source is in contact with the detector can. With a ¹³⁷Cs point source directly on the vacuum can of 80 cm³ Ge(Li) detector, the radial gradient in efficiency (i.e., across the can) is 1.6%/mm at r = 10 mm; this is three times the efficiency gradient of a 7.6 x 7.6 cm NaI(T1) detector. The present work extends these considerations to different detector configurations.

- c. Selective Radiochemical Separation for the Activation Analysis of Biological Standard Reference Materials
 - T. E. Gills

New biological materials such as Tomato leaves, Pine Needles and Spinach are currently being characterized at the National Bureau of Standards as part of the Standard Reference Materials Certification Program. Of the elements whose concentrations are usually certified, Copper, Arsenic, and Antimony could be determined accurately and precisely by activation analysis if sensitivity were the only consideration. However, the matrix radioactivities in most cases prevent instrumental determination of those elements or nuclides. The most useful (n,γ) reactions involve short-lived nuclides of Cu, As and Sb thus requiring chemical separation of those elements from the activated matrix. Separation procedures by Kosta et. al. for Cu, As, and Sb are capable of producing accurate results at the sub-ppm level of concentration. However, such procedures are highly

dependent upon the pH of the extraction system and are not easily adaptable to multielemental analyses.

Among possible radiochemical procedures and reagents for group separation of elements, inorganic ion exchangers have received increased interest arising from the high selectivity which can often be obtained by their use. Furthermore, by separating elements in groups instead of individually, and utilizing the high resolution of semiconductor detectors, simple, low cost and rapid analyses can be achieved.

Table 1

A Comparison of NAA Results Using The Described Separation Procedure to other Independent Methods

Code	Element	Spinach	Pine Needles	Tomato Leaves
1	Cu	10.8 ± .2	2.98 ± .16	11.1 ± .2
4	Cu	12.7 ± .4	3.2 ± .4	11.5 ± .2
3	Cu	11.5	2.9	10.6
1	As	.12 ± .01	.20 ± .02	.23 ± .03
2	As	.17 ± .05	.21 ± .04	.29 ± .05
1	Sb	.043 ± .004	.22 ± .01	.12 ± .03
			_	

Codes: 1 - Neutron Activation Analysis with Chemical Separation. 2 - Colorimetry.

- 3 Isotope Dilution Spark Source Mass Spectrometry.
- 4 Atomic Absorption Spectrometry.

This work involved the use of two chromatographic columns separately filled with Cuprous Chloride and Tin Dioxide. The columns were placed in series to provide a selective separation procedure for Cu, As and Sb from neutron activated biological materials. This separation scheme, was initially tested by reanalyzing previously certified NBS Orchard Leaves (1577). The procedure was then used to determine Cu, As, and Sb in new proposed Standard Reference Materials: Spinach, Pine Needles and Tomato Leaves. A comparison of NAA results using this procedure to other independent methods is given in table 1.

 d. Evaluation by Activation Analysis of Elemental Retention in Biological Samples After Low Temperature Ashing
G. J. Lutz, J. S. Stemple and H. L. Rook

Low temperature ashing (LTA) is the low temperature ($\sim 100^{\circ}$ C) decomposition of organic or biological samples using atomic oxygen produced by an electrodeless radio-frequency discharge. There are a number of advantages in using LTA as the first step in a general analytical method such as the avoidance of contamination during wet or high temperature ashing. With respect to activation analysis, freeze-drying followed by LTA has the advantage that with the elimination of organic carbon, the possibility of charring a sample with a very high neutron flux is removed. In addition, wet digestions can be substantially speeded up and hence short-lived nuclides can be more readily separated for counting.

The possibility exists in the LTA method of the loss of some volitile elements during ashing. We have studied, using activation analysis, loss or contamination of trace elements during LTA.

A commerical model LTA apparatus capable of 100 watts forward power was used in these experiments. A schematic of the apparatus is shown in figure 1. The sample was placed in a borsilicate glass ashing chamber which was briefly rotated by hand to induce the organic sample to spread as evenly as possible on the inside surface. The cold trap was filled

with crushed dry ice (liquid nitrogen would condense explosive ozone). The system was pumped down to 1.3 - 2.6 Pa (10-20 millitorr). The oxygen flow rate was adjusted so as to maintain a pressure of 13-40 Pa (100-300 millitorr). Forward power was usually set to the maximum of 100 watts. Tuning to optimum was accomplished by observing the maximum intensity in discharge glow in the tube.

Preliminary experiments involving addition of tracers to biological materials and determining retention of each isotope after ashing verified previously published data and served to establish and perfect the basic ashing technique. After these experiments, it was decided to study three NBS Standard Reference Materials in detail: 1571-Orchard Leaves, 1577-Bovine Liver, and 1632-Trace Elements in Coal.

Samples of about one gram were ashed for a period of several hours. The relative amounts of the elements in ashed and unashed samples were determined by nondestructive neutron activation analysis with the NBS 10 megawatt nuclear reactor and photon activation analysis with the NBS electron linear accelerator. Suitable irradiation and counting schedules were arranged to detect and quantify both long- and short-lived activities.

In the activation analysis experiments, the losses of iodine, bromine, chlorine and mercury in addition to carbon were observed.

Figure 2 shows, in the format of the periodic chart, the elements detected and not lost during ashing in all experiments. The symbols in the individual square of each element, OL, C and BL stand for orchard leaves, coal and beef liver, respectively, in which the element was determined. In addition, some elements reported retained in the literature determined by neutron activation analysis, atomic absorption spectrometry, or spectrophotometry are included.

It is also significant to note that, equal in importance to loss of trace elements, there was no pick-up of contamination during the ashing process as would be evidenced by an increase in the amount of the element after ashing. This is in spite of the fact that only very ordinary precautions were taken to guard against contamination during cleaning of the

borosilicate glass ashing tube with ordinary laboratory detergent followed by rinsing with water, hot dilute hydrochloric acid and finally deionized water and drying in an oven at 110° C.



Figure 1. Block diagram of LTA apparatus.

2. Cooperative Research Projects

a. The Determination of the Absorption of Metallic Ions on Tooth Tissue

The American Dental Association Health Foundation Research Unit at NBS (ADA-NBS) is involved in a research effort to improve adhesive bonding between hard tooth tissues and materials for use in preventive and restorative dentistry. The initial direction of this project is to determine which candidate metallic ions, are the best suited to serve as a mordant to improve the adhesion of dental resins and composites to hard tooth tissues. Success in this objective could save millions of teeth annually from decay and the costly procedure of drilling and filling with expensive silver alloy restorations.

Neutron activation analysis is currently being used to determine the concentration of four elements (A1, Cu, Fe and Zn) in hard tooth

Table 1. Trace Element Variations in a Whole Bovine Liver Compared to Replicate Analysis of NBS Bovine Liver.

			Averag	ge Concentra	ations[ppm]			
	As	Se	Cu	Co	Fe .	Zn	Мо	La
Whole Bovine Liver	00/1 001							
n = 12 (2-gram samples) (wet wt.)	.004 <u>+</u> .001 (25%)	<u><</u> .1	(11%)	.056 <u>+</u> .003 (6%)	240+18 (8%)	42 <u>+</u> 5 (12%)	1.3 <u>+</u> .2 (15%)	1.7 <u>+</u> .3 (18%)
NBS Bovine Liver Standard n = 6 (gm dry wt.)	.055 <u>+</u> .003 (5%)	1.2+.1 (8%)	193 <u>+</u> 14 (7%)	.18 <u>+</u> .01 (6%)	.027 <u>+</u> .002 (7%)	125+5 (4%)	3.0 <u>+</u> .3 (10%)	
NBS Certified Certificate Value or recommended Interim Value	.055	1.1 <u>+</u> .1	193 <u>+</u> 10	.18	.027 <u>+</u> .002	130 <u>+</u> 10	3.2	

H LI BE NA MG C C C C OL BL BL K CA SC TI V CR IMN FE CO INI CU IZN GA GE AS SE KR															
LI BE NA MG C C CL OL BL K CA ISC ITI IV ICB IMN FE ICO INI ICU IZN GA GE AS SE KB	HE														Н
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CS EA LA HF TA W RE IR PT AU TL PB BI PO RN C C C C C C C T 5 C OL PO RN OL OL OL OL OL C C N N N	PO RN	PB BI C OL	TL 5		AU T	PT .	IR		RE	W c	TA C	HF C OL	LA C OL EL	BA C OL	CS C OL
FR RA AC CE PR ND PM SM EU GD TB DY HO ER TM YB LU OL BL PA U NP PU AM CM BK CF ES FM MD NO LW	TM YB LU MD NO LW	ER 1	HO ES	DY CF	TB	GD		Si/) Ph	2 NE	PP.		AC	RA	FR

Figure 2. Elements retained during LTA.

tissue. Metallic salts of these four metals were chosen to study as mordants (chemicals which serve to fix a colorless surface active coupling agent on the surface of enamel or dentin) because ions of these metals form more stable chelates with organic completing compounds than calcium ions, which are dominant in tooth tissue. Improved bonding might be obtained if appropriate metal cations are placed on the tooth surface, and in effect replace or supplement calcium ions as sites for bonding by means of chelating or coupling agents.

The results obtained in this study will be published later in cooperation with the ADA-NBS.

3. Analytical Research Applied to Specific NBS Programs

a. The Use of Neutron Activation Analysis in the Evaluating of Sampling, Storage and Analysis of Samples for the National Banking System*

T. E. Gills and L. T. McClendon

When reviewing the literature for trace element concentrations in similar biological materials the range of the results are enormous. Several orders of magnitude in analytical data are not uncommon, even from laboratories using comparable (though not ridgidly standardized) techniques for biological materials. The need to make accurate and precise measurements of selected metals in biological samples such as human, plant and animal tissue is important to agencies who have the responsibilities to monitor trace element pollution trends and establish pollution guidelines. The National Bureau of Standards (NBS) has been cooperating with the Environmental Protection Agency (EPA) in the evaluation and improvement of methods for sampling and storage of environmental and biological materials. This research is in support of the proposed National Environmental Specimen Bank (NESB). It is the purpose of NESB to preserve samples for long periods so that retrospective analyses can be made.

One important part of this research is to gain a better understanding of trace element variations within whole organs or tissue biopsies. Since many of the biologically active elements are present at the ultra trace levels, the utmost in analytical sensitivities are necessary to study and evaluate many of these trace elements. Neutron activation has been extensively used in this study due to its sensitivity and specificity for many elements in a variety of matrices. A typical analysis of a fresh whole bovine liver compared to an NBS SRM (liver matrix) is given in table 1 as an illustration of elemental variation within a tissue compared to analytical variability.

b. Prompt-Gamma Activation Analysis

R. M. Lindstrom, W. B. Walters*, and W. Zoller*

In the past year, a cooperative venture was initiated between the University of Maryland nuclear and atmospheric chemistry group and the Activation Analysis Section to design and evaluate a prompt-gamma-ray counting facility dedicated to chemical analysis. The intense, wellthermalized neutron flux available in the National Bureau of Standards Reactor (NBSR) makes it likely that this prompt-gamma facility will be the best ever operated for analytical usage.

After examining several possible ports, it was decided that a vertical beam tube in the reflector could offer the best combination of high flux ($\sim 2.2 \times 10^8$ /cm²sec) good thermalization (CR_{AU} ~ 20) and low flux of scattered gamma-ray. A thimble and shutter have been designed by R. S. Carter and J. Sturrock for insertion into V5 and is currently being fabricated in the NBS Shops. An order has been placed for a large computer-based multiparameter pulse height analyzer, and designs for the target holder and beam stop, primary Ge(Li) gamma-ray detector and splitannulus coincidence detector are being completed. Initial experiments are planned for early in 1977.

c. A Study of Lithium Uptake and Location in the Brain Using the Nuclear Track Technique

B. S. Carpenter, D. Samuel, I. Wassermann and A. Yuwiler It is well established that lithium salts are a most effective treatment for some mental disorders, particularly for mania and forms of depression. In spite of their widespread use, both therapeutically and prophylactically, their mode of action is still poorly understood. One aspect of lithium treatment in man seems to be its mono- and bi-polar forms. An understanding of the neurochemistry and psychopharmacology of this disease may help in the understanding of other forms of mental disorder. In spite of research on the effect of lithium electrolytes, on various neurotransmitters and other biochemical systems, little is known about their mode of action or their distribution in mammalian brains. One reason is the absence of a suitable, sufficiently long-lived radioactive tracer which would enable the site and rate of turnover release of lithium to be studied by classical biochemical techniques of labeling, autoradiography, etc. A second problem is the difficulty of studying, in vivo, metabolic processes in the brain, and the dearth of suitable animal models of affective mental disorder.

In order to determine the lithium concentration, rate of uptake and location in the brain, rats were injected interperitoneally with doses of 99.6% enriched ⁶Li₂CO₃ in an inert carrier (carboxymethyl cellulose). The doses of ⁶Li₂CO₃ varied from 50 to 100 mg/kg body weight. The rats were sacrificed at 0, 0.5, 1, 3, 5, 8, 12, 24, and 48 hours after injection. The brains were then excised, fixed, and two sections made: (a) a transverse section through the hypothalamus region and (b) a transverse section through the pons. These sections were sandwiched between dyed cellulose nitrate (CN) detectors and prepared for thermal neutron irradiation. The tissues were irradiated for 90 seconds (1.33 x 10¹³ $n \cdot cm^{-2} \cdot s^{-1}$) to produce α -tracks in the CN detectors from the ⁶Li(n, α)³H reaction. The tissues were separated from the detectors and the CN detectors were etched in a caustic solution. The entire area that the

brain tissues occupied on the detector was scanned, \sim 13 mm by 19 mm for each detector, by the use of an optical light transmission microscope.

Visual observation of the α -track density indicated that the lithium was dispersed between the cortical and the subcortical regions nonuniformly. The distribution of the track densities in the tissues is believed to be indicative of the fact that the Li is not bound chemically in the brain to plasma protein, but remains free in plasma water.

Microscopically, the results of lithium uptake study showed that Li reach its maximum concentration level in both the hypothalamus and the pons region of the brain 8 hours after injection of the Li_2CO_3 . Finally, from this study the biological half-life of Li in the brain was determined by assuming they followed a first order exponential decay 12 hours after injection. The Li biological half-life in the hypothalamus region was found to average 29.4 ± 2.9 hours. This value is in close agreement with the 30.9 hour value obtained from Ebadi, *et al*, data. The halflife in the pons region was longer, 132.5 ± 17.2 hours. There are no other results available on the biological half-life of Li in the pons region.

4. Analytical Support for the SRM Program

a. Standard Reference Materials

Much of the Section's efforts have been in support of the Standard Reference Materials Certification Program. A wide variety of SRM's were analyzed by NAA and the table below lists those in which activation analysis was responsible for a significant fraction of the trace element certification data.

Table 1

	Matrix	SRM/Reference Sample Name
(a)	Copper	Copper Benchmark
(b)	Food Grain	∫Wheat Flour Rice Flour
(c)	Body Fluids	Serum
(d)	Water	Toxic Metals in Water
(e)	Botanicals	Spinach Tomato Leaves Pine Needles
(f) (g) (h)	Biological River Sediment Rock	Brewer's Yeast Sediment Obsidian Rock

b. SRM Feasibility Studies - SRM Mixing

R. M. Lindstrom, B. S. Carpenter and S. S. Fine The intent of the project was to establish and test procedures by which an analyst can prepare from two different SRM's a standard sample of known intermediate composition.

The first pair of SRM's examined was pine needles and tomato leaves due to their similar particle size and density and their dissimilar trace element content. Mixing known (weighed) proportions of these two materials and analyzing each of nine mixes resulted in a satisfying predictability of concentration.

Similar mixtures have been made from pine needles and river sediment, a pair of materials that might be expected not to mix well nor stay homogeneous once blended. These experiments should give an indication of how well the simple procedures used here can be expected to predict the composition of mixtures. The final output of the work, to be completed during the transition quarter 1976, will be a manuscript describing the experiments and detailing expected variations in resultant compositions.

5. Service Analyses

In addition to analyses on SRM's, a large number of analyses were made on non-SRM Materials. These include cooperative analyses, where the analyses were made on samples furnished by other laboratories in the past year. The following analyses were done:

- 1. Mercury in Soil and Calcium Chloride G. J. Lutz, H. L. Rook
- 2. Mercury in Sea Water G. J. Lutz, H. L. Rook
- 3. Trace Elements in Antimony Trioxide R. M. Lindstrom, S. Fine
- 4. Platinum in Iron Oxide T. E. Gills, H. L. Rook
- 5. Impurities in Lithium Hydroxide T. E. Gills
- 6. Trace Elements in Raney Nickel R. H. Filby
- 7. Trace Elements in Fiber Samples R. M. Lindstrom, S. H. Harrison
- 8. Titanium in Sapphire G. J. Lutz
- 9. Analysis of Gold Films S. H. Harrison, R. M. Lindstrom
- 10. Trace Metals in Polyethylene Inplant Standards T. E. Gills
- 11. Trace Elements in Dental Materials L. T. McClendon, T. E. Gills
- 12. Boron in Silicon Detectors B. S. Carpenter

6. Facilities

Two major enhancements to our ability to analyze large numbers of samples were added in the past year. First, our two best Ge(Li) detectoranalyzer systems were equipped with automatic sample changers (Ref: Shideler and Suddueth, in prep.), allowing unattended counting of many samples over evenings and weekends. The two sample changers are electrically similar in operation, but mechanically different so as to handle 55 mm x 12 mm petri dishes with one system and 20-ml scintillation counting vials with the other. The first system offers the ability to count samples at 5 to 10 cm from the detector face, as well as at 1 cm.
NON-RRD NBS PROGRAMS

The second major strengthening of the analytical system is the addition of several additional capabilities to QLN1, the activation analysis computer code written and maintained for this Section under contract by H. P. Yule of the NUS Corporation. These features include the quantitative as well as qualitative use of the code's multiplet resolution abilities, and the estimation of upper limits for components sought but not found. The code will now handle up to 4096 channels of data per spectrum.

FILTERED BEAMS

R. B. Schwartz and I. G. Schroder (Nuclear Sciences Division)

A 136 cm long silicon filter was placed in through tube GT-2, viewing a graphite scatterer placed at the center of the tube. This



Figure 1. "Phantom" in position in front of the 2 keV beam.

arrangement produces an intense collimated beam of 144 keV neutrons. The addition of \sim 3 cm of titanium removed a low-energy contaminant peak, resulting in an essentially pure beam of 144 keV neutrons. The beam intensity is \sim 3 x 10⁶ neutrons/s, in an one-inch diameter beam. The gamma ray contamination is \sim 30 mR/hr. This 144 keV beam, together with the very pure 2 keV and 25 keV filtered beams, gives the NBS Reactor a unique facility for instrument and dosimeter calibration in the keV neutron energy range.

Typical uses for the facility include testing the response of albedo dosimeters by placing them on our anthropomorphic phantom and irradiating them under carefully controlled conditions in the various beams. Figure 1 shows the phantom in position in front of the 2 keV beam during such an irradiation.

INTERCOMPARISON OF FISSION RATE MEASUREMENTS: KARLSRUHE REACTOR PHYSICS AND LOS ALAMOS RADIOCHEMISTRY

D. M. Gilliam (Nuclear Sciences Division)

Fission rate measurement scales at two laboratories engaged in quite different branches of nuclear energy development have been intercompared via NBS initiative and cooperative execution. The Reactor Physics Group at GfK (Gessellschaft für Kernforschung) in Karlsruhe, Germany, submitted fissionable deposits for direct comparisons with NBS reference deposits, and the Radiochemistry Group at Los Alamos participated in joint fission rate measurements with NBS at Los Alamos. For the basic uranium fissionable isotopes, both laboratories are within 2% of the NBS fission rate measurement scale. This is marginally adequate for some programmatic goals; more serious, however, is a discrepancy between Los Alamos and NBS of nearly 6% for Pu-239 fission rate measurements.

With the cooperation of Dr. Werner Scholtyssek of GfK, fissionable deposits of U-235, Pu-239, and normal uranium were sent to NBS for comparisons with the corresponding NBS reference deposits. The mass assay of the NBS reference deposits has been described in detail by Grundl, et al.¹ The GfK deposits were received and maintained in sealed fission chambers of the type used at Karlsruhe. The NBS reference deposits were mounted in nearly identical gas flow chambers. Both chambers were of the parallel plate type with the deposit positioned flush against a thin The chambers were mounted so that the two deposits would aluminum wall. be back-to-back with only the thin aluminum walls separating them. In this configuration, the chambers were positioned so that the deposits were at the center of a 4-cm-wide beam from the thermal column. The angle between the beam axis and the deposit normal was not determined to high precision, but a very accurate 180° rotation was possible by alignment of the holder base against a fixed plate on the supporting table. The relative fission rates from the two orientations were averaged in the following way:

$$(F_{GfK}/F_{NBS})_{AV} = \sqrt{(F_{GfK}/F_{NBS})_{\theta}} (F_{GfK}/F_{NBS})_{\theta+180}$$

where θ is the angle between the beam axis and the chamber axis. A choice of θ = 55° was made for the final deposit comparisons, based on

Table 1. Variation of the orientation in the beam.

θ	(F _{GfK} /F	NBS) θ	(F _{GfK} /F	NBS) θ+180	(F _{GfK} /	F _{NBS}	5) AV
70°	1.3235	<u>+</u>	.0004	1.062	<u>+</u>	.001	1.185	<u>+</u>	.001
55°	1.267	+	.001	1.108	<u>+</u>	.001	1.185	<u>+</u>	.001
35°	1.2446	<u>+</u>	.0006	1.1334	<u>+</u>	.0005	1.188	+	.001
20 °	1.215	<u>+</u>	.002	1.136	<u>+</u>	.003	1.175	<u>+</u>	.003
0°	1.385	+	.001	1.137	+	.001	1.255	+	.001

the data of table 1 for enriched uranium deposits at a variety of angles. Consistency with \pm 0.25% is seen over the range 35° to 70°. The odd ratios at small angles are not at all surprising, since gross scattering effects from insulators, cables, support structures, and the gas line are expected at these angles.

Table 2 shows the results of the comparison and the estimated uncertainties. The largest uncertainties were the quoted errors (1.2% to 1.5%) for the NBS reference deposits themselves.¹ Other systematic errors contributed about 0.5% uncertainty or less.

Table 2. Comparison of results.

	Mass	Mass	Mass Scale
Principal	Determined	Determined	Ratio
Isotope	by NBS	by GfK	GfK/NBS
	μg	μg	
U-238	325.1 + 1.6%	325	0.9997
11-225	300 / 1 1 39	21.1	1 0052
0-235	JUJ.4 <u>-</u> 1.J%	JII	1.0052
Pu-239	28.62 + 1.3%	28.98	1.0126

The intercomparison of absolute fission rate measurements between NBS and the Radiochemistry Group responsible for weapons diagnostics at Los Alamos Scientific Laboratory (LASL) was carried out at LASL. After calibrations and temperature effects testing at the National Bureau of Standards Reactor, double fission chambers developed at NBS were operated in the BIG-10 critical facility at LASL simultaneously with the irradiation of an activation fission foil pack which was later destructively analyzed by the Radiochemistry Group. Table 3 shows the results of the comparison. The striking discrepancy in Pu-239 fission rates is receiving careful study by both groups.

Table 3. Fission rates (per nucleus) in BIG-10 test C-LASL.

	Ise	otope Studie	ed	
	U-235	U-238	N _D -237	Pu-239
NBS Measurement $(10^{-15} \text{ sec}^{-1})$	110.4±1.5%	4.14±1.8%	35.2±1.9%	132.3±1.6%
LASL Measurement $(10^{-15} \text{ sec}^{-1})$	112 ±1.6%	4.21±3.8%	35.3±5%	140 ±2%
LASL/NBS	1.014	1.017	1.003	1.058

 J. A. Grundl, D. M. Gilliam, N. D. Dudey, and R. J. Popek, "Measurement of Absolute Fission Rates," *Nuclear Technology* <u>25</u>, 237 (1975).

PRECISION MEASUREMENT OF THE WAVELENGTH OF NUCLEAR GAMMA LINES AND THE COMPTON WAVELENGTH OF THE ELECTRON

> R. D. Deslattes, E. G. Kessler,W. C. Sauder, and A. Henins (Optical Physics Division)

The wavelengths of nuclear γ -lines and the Compton wavelength of the electron are being measured in terms of the wavelength of a molecularly stabilized visible laser. The wavelength measurements use a two crystal transmission spectrometer equipped with angle-measuring interferometers to determine the crystal diffraction angles. The γ -rays are diffracted by nearly perfect Ge and Si crystals whose spacings have been accurately measured in terms of the above-mentioned stabilized laser. A more detailed description of the experimental procedure has been given in other reports.^{1,2}

Our activity over the past year has been primarily centered in three areas: (1) the reduction of systematic uncertainties associated with the vertical divergence correction, the angle interferometer calibration, the temperature sensitivity of the angle interferometers, and the crystal lattice spacing; (2) the irradiation of several

 γ -sources of Au-198 and Ir-192 and the measurement and data analysis of the wavelengths of the associated γ -radiations; and (3) the design and manufacture of a transfer cask for nuclear γ -sources and the design of a transfer cask for the Compton wavelength measurement.

1. Reduction of Systematic Uncertainty

Several new Soller collimators have been manufactured which allow the vertical divergence correction to be varied and be more accurately calculated.

The interferometers have been recalibrated under more controlled conditions to obtain an interferometer constant accurate to 0.06 ppm. The calibration procedure involves summing to closure measurements of the interfacial angles of a 72-sided optical polygon and provides absolute angle measuring capability.

One of the angle interferometer zeroes was found to drift with temperature. This drift resulted from a temperature sensitive dove prism in the interferometer which has now been replaced with good effect. An interferometer design which eliminates the dove prism is planned for the future. Temperatures are controlled to better than 0.01 °C so that sub ppm measurements are possible even with the present interferometer.

The crystal lattice spacing was determined by first measuring the spacing of a standard Si crystal by simultaneous optical and x-ray interferometry.³ Then, the ratios of the lattice spacings of this standard Si crystal and of the crystals used for γ -ray diffraction are measured. This transfer has been completed to the 0.5 ppm level. However, γ -ray wavelengths measured with Ge and Si crystals differ by 1 to 2 ppm as indicated below. An unknown systematic error of this magnitude is thus associated with the crystal lattice measurements or the γ -ray angle measurements. This systematic error is under investigation.

Three Au sources were irradiated with activities up to 4×10^3 Ci. These sources were used for measurement of the wavelength of the 412 keV Au-198 line. Measurements were obtained for diffraction with Ge crystals in two orders and Si crystals in three orders. Measurements in different orders are consistent. However, wavelengths measured with Si crystals are approximately 2 ppm larger than those measured with Ge crystals.

A 2 x 10^3 Ci Ir-192 source was produced and used for measurement of eight γ lines (205, 295, 308, 316, 468, 588, 604, 612 keV) by diffraction with Ge crystals in two different orders. Five of these lines were used in two tests of consistency by the combination principle (295 + 316 = 612 keV and 295 + 308 = 604 keV). Closure was found within 0.5 ppm. The wavelengths of four other lines (136, 416, 484, and 884 keV) were determined by use of the combination principle. The strongest four lines (295, 308, 316, and 468 keV) were also measured using Si crystals. Wavelengths measured with Si crystals are approximately 1.5 ppm larger than those measured with Ge crystals.

Although there is a 1 to 2 ppm unknown systematic associated with our wavelength measurements, our results are nonetheless 10 times more accurate than previous results.⁴ A detailed summary of all our γ -ray measurements will appear shortly in reference 5.

3. Design and Manufacture of Transfer Casks

A 41 cm diameter transfer cask for intense (several kilo-Ci) nuclear γ sources was designed and constructed. The cask allows convenient irradiation of samples from the vertical ports on the NBSR.

A large diameter transfer cask for a multi kilo-Ci Cu-64 source has been designed and is under construction. This source will serve as a positron emitter in the measurement of the Compton wavelength of the electron.

- R. D. Deslattes, E. G. Kessler, W. C. Sauder, and A. Henins, Atomic Masses and Fundamental Constants 5, J. H. Sanders, A. H. Wapstra, eds., Plenum Press, New York, p. 48 (1976).
- R. D. Deslattes, E. G. Kessler, W. C. Sauder, and A. Henins, Nat. Bur. Stand. (U.S.), Tech. Note 896, 114 (Jan. 1976).
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- 4. R. G. Helmer, R. C. Greenwood, and R. J. Gehrke, *Atomic Masses and Fundamental Constants 5*, J. H. Sanders, A. H. Wapstra, eds., Plenum Press, New York, p. 30 (1976).
- 5. R. D. Deslattes, *Proceedings of the School of Physics*, 'Enrico Fermi', Metrology and Fundamental Constants Course (1976). Preprints will shortly be available.

E. SUMMARY OF REACTOR OPERATIONS

Operating productivity and fuel utilization efficiency for the past year were the best in the history of the facility. The newly installed heat exchangers functioned well. Otherwise, this operating cycle was uneventful. Four papers dealing with the operation of the NBSR were presented at the August 1975 meeting of the Reactor Operations Division of the American Nuclear Society.

A summary of the overall operating statistics for this and previous years is presented in the following table.

NBSR Operating Summary

	<u>1973</u>	<u>1974</u>	1975	1976
Reactor Operations to date, MWh	229,000	290,000	328,000	396,000
Reactor Operations for year, MWh	67,000	61,000	38,000	68,000
Hours Reactor Critical	7,000	6,600	3,900	6,900
Number of Days at 10 MW	279	254	157	283
On-line Time at 10 MW	77%	70%	43%	78%
Number of fuel elements used	28	24	16	24
Average U-235 Burnup	52%	54%	53%	55%
Number of Refueling Operations	7	6	4	6
Number of Unscheduled Shutdowns	14	6	0	2
Number of Irradiations	4,300	3,000	1,550	2,360
Irradiations, Total Hours	5,250	4,300	1,800	7,250
Hours per irradiation	1.2	1.4	1.2	3
Number of Visitors	3,000	5,000	3,000	7,000

ACTIVATION ANALYSIS PROGRAM OF THE FOOD AND DRUG ADMINISTRATION AT THE NBSR

J. T. Tanner and M. H. Friedman (Food and Drug Administration, Washington, DC)

1. Analysis of Chlorine in Japanese Rice Oil

During 1968 an incident in Japan (the so-called "Yusho" poisoning) resulted in the contamination of a large quantity of rice oil with polychlorinated biphenyls (PCB). Several people died and many were injured ppermanently as a result of consuming this contaminated cooking oil. The FDA has recently been reviewing regulations governing the amount of PCB allowable in foods. Since much of the data on the effect of PCB on man was gathered as a result of this accidental contamination incident in Japan, it was very important that there be an accurate analysis of this oil for PCB.

A sample of this rice oil was analyzed at the FDA headquarters laboratory for PCB's and compared with an earlier analysis of PCB done in Japan. The results differed by almost a factor of two. The analytical procedure used by FDA was the official method of analysis for PCB currently listed by the A.O.A.C. The Japanese data were based on earlier methodology and on neutron activation analysis for total chlorine.

In support of the pesticide analysis unit of FDA, the NAA unit analyzed the contaminated oil for chlorine. The NAA results supported the original Japanese analysis. Since control rice oil contained virtually no chlorine, the chlorine detected was assumed to be bonded to the PCB (approximately 50% chlorine) or another yet unknown contaminant.

To resolve the factor of two discrepancy, the pesticides unit then succeeded in separating the chlorine containing compounds by adsorption chromotography and GLC into two fractions. One fraction contained PCB and results from NAA chlorine determination and the A.O.A.C. PCB method were equivalent. The second fraction was a group of later components

from a GLC column and contained an equivalent amount of chlorine. This resolved the factor of two discrepancy between analyses at FDA and in Japan. To date, the other chlorine containing compound(s) have not been identified.

2. Analysis of Animal Waste Material

Recently it has become common practice to use animal waste as a feed ingredient for animal feeds. This practice has been approved by

Table 1. Elemental concentration of animal waste material $(\mu g/g)$

Element	Dried Cattle Manure (<u>low fiber</u>)	Dried Cattle Manure (<u>high fiber</u>)	Dried Processed Waste <u>Pellets</u>	Dried Poultry Waste (<u>with litter</u>)	Dried Poultry <u>Waste</u>	Dried Sewage Sludge
Fe	2394±444	4565±611	1890±125	895±84	1838±14	15414±444
Zn	109±11	79±10	69±3	165±3	140±12	1752±113
Rb	19±2	37±1	14.7±0.2	8.8±1.1	12.7±1.3	32±11
Co	1.7±0.5	2.2±0.5	1.1±0.2	2.0±0.3	1.2±0.1	7.1±2
ЅЪ	0.28±0.02	0.29±0.05	0.22±0.02	0.37±0.02	0.38±0.02	6.3±1.8
Br	29±4	29±2	56±3	9±2	20±2	30±3
Cr	20±7	31±9	5±1	6±1	4.9±0.3	280±105
Se	0.5±0.2	0.6±0.1	0.29±0.01	0.4±0.1	0.7±0.2	4.8±1.2
Ba	0.09±0.02	0.17±0.01	0.07±0.01	<0.2	<0.1	0.8±0.2
Sc	<0.7	<2	<0.4	0.1.9±0.02	0.3±0.1	
Cs	0.13±0.02	0.33±0.05	0.25±0.06	0.06±0.02	0.10±0.04	0.58±0.03
La	2.6±0.4	11±1	2.5±0.5	2.4±0.4	1.9±0.2	27±8
Eu	0.13±0.02	0.5±0.2	0.09±0.03	0.05±0.02	0.07±0.03	0.6±0.1
v	1.6±0.4	9±5	2±1	2.9±0.3	2.9±0.3	21±2
Ca	10500±700	10500±700	12500±2000	48000±3000	62000±7000	27000±6000
C1	7800±600	6050±1300	5200±400	3400±100	5300±400	570±100
Al	6300±300	12000±5000	2600±600 ⁰	1070±15	2400±100	19000±900
Mg	2300±800	2900±700	4000±500	3200±1000	3600±400	4000±2000
· Cu	<100	<100	<50	<50	<100	500±60
Mn	40±17	103±9	57±6	130±50	160±20	210±40
Na	5600±700	6800±1500	3300±900	1600±400	3500±900	3400±800
К	21000±8000	16000±4000	20000±4000	15000±3000	15000±3000	14000±7000
As	<1.4	<1.6	<1.2	<1.2	<1.4	4±1

*The above samples were collected from the feed lots (except sludge) and air dried. The sludge was also air dried.

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seven states for animals in intrastate commerce, but has not been approved by FDA for animals in interstate commerce. In order to establish base line information about trace metals in typical animal waste products, samples of dried waste from animals with a known diet history and a sample of processed waste pellets composed of a mixture of animal feed and dried animal waste were obtained for analysis. In addition a sample of sewage sludge, which is being tested as an animal feed ingredient on an experimental basis, was analyzed. The results of these analyses are given in table 1. The high levels of many metals in both animal waste products and sewage sludge indicate that the practice of using animal waste or sewage sludge as animal feed ingredients should be carefully evaluated before being fully sanctioned.

IMPROVED MATERIAL FOR SOLAR ENERGY CELLS

T. M. Raby

Neutron Transmutation Doped (NTD) silicon is beginning to assume a position of major importance in the semiconductor device field and may have wide ranging application in the development of solar energy cells.

It has been recognized in recent years that silicon is almost an ideal material for transmutation doping. Normally available silicon contains 3% Si-30 which transmutes to P-31 on the capture of a thermal neutron. This process produces a uniform concentration of phosphorus, a paramount factor in the material's performance and use.

The NBS reactor, with its extensive and versatile facilities and its intense highly thermalized neutron flux, is ideally suited for transmutation doping of materials. Over the past several months, together with the Oak Ridge National Laboratory, numerous silicon samples have been irradiated at the NBS reactor. Extensive research is currently being carried out to study the use of NTD silicon in power

devices and solar cells and to expand its application into other semiconductor fields. The success of this work could mark a new era in the use of reactor produced materials in important energy fields.

ATF'S FORENSIC ACTIVATION ANALYSIS PROGRAM

C. M. Hoffman (U.S. Treasury Department, Washington, DC)

Although the use of NAA for detecting barium and antimony in gunshot residues has been replaced by flameless atomic absorption spectrophotometry, NAA still plays a valuable role in the analysis of trace evidence. The method's sensitivity and non-destructive nature make the technique very attractive for the analysis of certain types of physical evidence commonly found at crime scenes.

During the past year, our Laboratory used NAA to examine evidence in 175 cases. This evidence consisted of hair, paint, glass, soil and metal particles.

The conclusions drawn by a Laboratory examiner concerning the relative origins of questioned and known evidentiary materials depends upon the analytical background data available for the type of material under examination. Characterization of the material requires that a significant quantity of data be available for this evaluation. Studies of various materials, such as paint and glass, are being conducted by our laboratory scientists to meet this need. For example, glass samples which cannot be discriminated by their refractive index, density or elemental composition as determined by x-ray flourescence, will be studied by NAA to assess the worth of this method as it applies to this type of problem.

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LUNAR SAMPLE ANALYSIS FOR 20 TRACE ELEMENTS

E. Anders (University of Chicago, Chicago, IL)

During the last year, numerous samples were analyized for 20 trace elements (Ag, Au, Bi, Br, Cd, Cs, Ge, In, Ir, Ni, Os, Pd, Rb, Re, Sb, Se, Te, Tl, U and Zn) by radiochemical neutron activation using the NBS Reactor. This is part of a continuing extensive program to determine the origin and constituents of lunar samples. The analysis involved trace levels as low as one part in 10^{15} and will be compared with those obtained from meteorite specimens.

ACTIVATION ANALYSIS PROGRAM OF THE U, S, GEOLOGICAL SURVEY

J. J. Rowe and P. A. Baedecker

(U. S. Geological Survey, Reston, VA)

Activation analysis is being applied by the Radiochemistry Project, Reston to the determination of trace concentrations of elements in geological materials. The technique is used for the analysis of rocks, minerals, waters and coals for geochemical studies related to ore formation, magmatic differentiation, hydrothermal alteration and transport, energy reserves and environmental problems.

Coals are being analyzed as part of a program to study the trace element content of U. S. coals. More than 1000 coal samples per year are being analyzed for 24 elements. This is possible through the use of automated counting equipment and computer data reduction. The system operates 24 hours per day, 7 days per week to collect gamma ray spectra onto magnetic tape. Samples are counted 4 days after irradiation and then again 6 weeks later. The data from the two counts are processed by computer to provide the final analytical report (Table 1). Through the use of epithermal neutron irradiations at the JEEP reactor, Kjeller, Norway we were able to instrumentally determine U, Sr, Mo, and Ni and

REPORT	
SUMMARY	
ANALYSIS	
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LIE JENKINS	10750 W1910760 W191076D 5 AL-7 AL-7 6 (A) (B)	12059 0.10753 0.12266
RIPTION: COALS	30 W1910740 W1910750 AL-5 AL-6 ()	85 0.10277 0.12059
M1910730 W1910740 AL-4 AL-5 () ()		0.13285 0.10277
0730 W19107 AL-4 8) AL-4		2126 0.132
0 W191072	AL-4 (8)	.2 0.1212
	W191073 AL-4 (A)	0.1067;
	W1910720 AL-3 ()	0.10574
	41910710 AL-7 ()	0.10780
	91070F W	1.10215
IRRAPIAT	W19 AL-	0.
	SAMPLE:	WEIGHT:

ERROR LIMITS ARE ONE STANDARD DEVIATION BASED ON COUNTING STATISTICS ALONE NUMBER IN PARENTHESES IS NUMBER OF INOIVIOUAL RESULTS, BASEO ON REPETITIVE COUNTS, AVERAGED TO YIELD EXPRESSEO RESULT

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108	NUMBER: AD-	-74	SUBM	ITTED 8Y: SM.	1TH		ANALYST:	BAEDECKER		
1 RR	ADIATION NUM	BER: 94,117			SAMP	LE DESCRIPTI	DN: VALLES M	ITNS . N.M.		
SAMPLE:	W176910 71-15 ()	W176911 71-16 ()	W176912 71-17 ()	W176913 71-18 ()	W176914 71-19 ()	W176915 71-20 ()	W176916 71-21 ()	H176917 71-22 ()	W176918 71-23 ()	76-50 ()
WEIGHT:	0.10684	0.11174	0.12156	0.11953	0*09830	0.12882	0.12996	0.11413	0.12181	0°19069
FE (2)	1.29	1.24	1.19	1.15	1.19	0.97	1.00	0.98	66*0	1.07
BA (PPM)	207 202	170	205	218	174	163	148	143	133	159
SIGMA(%)	8(1) 0-4	9(1)	7(1)	7(1)	10(1)	9(1)	9(1) 1-1 <	1-1 <	11(1)	7(1)
SIGNA(E)	11)2	(1)9	13(2)	13(2)	(1)2	0(1)	0(1)	(1)0	0(1)	31(2)
CR (PPM) SIGMA(2)	6.9 13(1)	3.5	6.2 18(1)	5.2	3.1 18(1)	1.4 < 10(1)	3.1 <	3.2 < 0(1)	3.1 0(1)	6.7 4(1)
CS (PPH)	9.6	10.0	11.0	10.2	10.6	10.1	10.0	10.4	10.3	6.0
HF (PPM)	3(2) 14°7	3(2) 13.8	13.12	3(2) 13.5	31 21 13 •5	12.3	12.1	12.3	12.6	7.2
SIGMA(2)	1(2)	1(2)	1(2)	1(2)	1(2)	(1)1	(1)1	(1)1	1(1)	120 1(2)
R8 (PPM) SIGMA(Z)	302 512)	313 5(2)	301 2(2)	314 2(2)	289 5(2)	342 4(1)	340 4(1)	334 4(1)	(1) 4 (1)	139 6(2)
S8 (PPM)	I.0	0.8	0.6	0.5	0.5 <	2.4 <	2.5 <	2.3 <	2 • 3	0.2
SIGMA(%)	16(2)	(1)6	10(1)	11(1)	12(1)	0(1)	0(1)	0(1)	0(1)	2 62
IA (PPH) SIGMA(%)	12.53	11.49	11.08	12.03	11.04	2(1)	2(1)	1(1)	(1)1	1(2)
TH (PPM)	. 39.3	35.9	36.1	36.9	35°1	44.7	45.3	45.4	44.5	13.7
ZN (PPM)	129	195 1	122 1(2)	127	111 1(2)	2(1) 89	85 2(1)	(1)2 62	82 2(1)	57 2121
SIGMA (%)	2(1)	2(1)	2(1)	2(1)	3(1)	3(1)	3(1)	(1)5	3(1)	2(1)
ZR (PPM)	338	377	346 7111	320	304	329 4111	279 0111	295 7111	298 7111	234 6111
SC (PPM)	1.02	0.94	0.87	0.83	0.87	0.54	0.55	0.55	0.54	1.36
SIGMA(2)	1(2)	1(2)	1(2)	1(2)	1(2)	2(1)	2(1)	2(1)	2(1)	72 1(2)
SIGHA(2)	1(1)	(1)1	1(1)	1(1)	(1)1	(1)1	101	(1)1	101	(1)1
CE (PPM)	136	133	139 1121	132	139 1731	109 2121	111 1/21	113	111 2421	114 1(3)
(MPP) ON	62	55	52	52	62	48	54	50	49	64
SIGMA(%)	6(3)	7(3)	7(3)	7(3)	7(3)	6(2)	6(2)	6(2)	7(2)	4(3)
SIGMA(Z)	10.00	2(1)	14.0	2(1)	1(1)	111)	2(1)	(1)1	1(1)	2(1)
EU (PPM)	0.12	0.10	0.09	0.17	0.10 <	0.06 <	> 90*0	0.06	0.06	0.31
SIGMA(%)	17(2)	13(2)	16(2)	16(2)	18(2)	0(1)0	0(1) 2 73	0(1)	34(1)	4(2)
SIGMA(2)	2(2)	2(2)	2(2)	2(2)	2(2)	4(1)	4(1)	3(1)	4(1)	3(2)
V8 (PPM)	13.1	12.7	12.2	12.9	12.6	12.3	12.0	12.2	11.6	4.5
SIGNA(%)	1.88	1(2) 1.69	1(2)	1.73	1.51	2(1) 1.61	2117	1111	1.59	0.64
SIGMA(2)	2(2)	3(2)	2(2)	2(2)	3(2)	5(1)	2(1)	2(1)	6(1)	6(2)

ERROR LIMITS ARE DNE STANDARD DEVLATION BASED DN COUNTING STATISTICS ALONE Number in Parentheses is number of individual results, based on repetitive counts, averaged to yield expressed result

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improve the determination of Co, Zn, As, Se, Br, Rb, Cs, Ba, Sm, Tb, Hf, Ta, W and Th to supplement the thermal irradiations done at the NBS reactor.¹

The project has been analyzing a wide variety of geological materials for numerous programs of the Survey. Gold has been determined in sediments and igneous rocks using a fire assay radiochemical method.² Ruthenium and iridium were studied in chromites using a fire assay radiochemical method with collection into a platinum bead^{3,4} followed by Ge(Li) or coincidence counting.

The NBS reactor facility has been utilized to provide thermal neutron irradiations leading to the determination of 22 to 24 elements in many geologic matrices (Table 2). In some cases the thermal irradiations have been supplemented by epithermal irradiations to instrumentally determine tungsten, cesium, rubidium, barium, uranium and thorium when they occur at low concentrations.

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TRACE ELEMENTS IN THE ENVIRONMENT AND RADIOACTIVE DECAY STUDIES

W. H. Zoller, G. E. Gordon and W. B. Walters (University of Maryland, College Park, MD)

Our group has made extensive use of the NBSR for the irradiation of numerous samples for trace-element analysis using instrumental neutron activation analysis (INAA). We have had our computer-based Northern Scientific analyzer system and two Ge(Li) γ -ray detectors at the Reactor for periods of several weeks during the last year to count short-lived radioactive products. Most of the other samples are returned to the university for counting long-lived activities.

In our "local" air pollution studies we have applied INAA to particles collected in the stacks of coal and oil-fired power plants and municipal incinerators. We have also analyzed particles collected with airplanes in the plumes of the Vienna oil-fired plant, and the Chalk Point (Md.), Dickerson (Md.) and Four Corners (New Mexico) coal-fired plants. (The latter is a joint venture with Lawrence Livermore Laboratory and some of the analyses have been done there.) We have also analyzed ambient aerosols collected from several sites in Washington, D. C. during the summer of 1975. Briefly we find that:

(1) We can now account almost quantitatively for the sources of about thirty elements observed in the Washington atmosphere.

(2) Coal combustion does not appear to be the major source of most toxic elements, exceptions being As, Se and vapor-phase Hg.

(3) From previous years' work, motor vehicles are the major sources of atmospheric Pb, Br, Cl and, possibly, Ba.

(4) Oil-fired plants are major sources of V, Ni and possible Se.

(5) Municipal incinerators release little total mass, but the particles are so rich in many trace elements that they are often a major source of urban, ambient Zn, Cd, Sb and, possibly, Ag and Sn. Since Sb and Cd are clearly toxic, this finding raises a serious question about the proposed use of urban refuse as a supplement to coal in power plants.

Recently we have begun studies on additional sources: cement plants and steel mills.

We are also continuing our studies of global atmospheric particulate samples. We have analyzed additonal particulate samples collected from the very clean air of the South Pole. Since there is essentially total snow cover there, the particles have had to be transported long distances. The enrichment factors of many elements are the same as for urban air, so one might be tempted to assume that those particles originate from anthropogenic sources. However, one cannot assume that without investigating particles from natural sources, of which volcanoes are a major example. In order to obtain better information on volcanic emissions, we collected aerosols from the Jan./Feb. 1976 Mt. Augustine eruption in Alaska with the use of an airplane operated by the National Center for Atmospheric Research in Boulder. Preliminary analyses of these samples indicate that a number of moderately volatile elements are highly enriched on the volcanic particles.

In our decay scheme studies we have completed work on the $\gamma\gamma$ angular correlations in Rh-105 following the decay of 4.4-hr Ru-105 and published the results in Physical Review C. The results of earlier studies of the decay of Te-134 to levels of I-134 have also been published. New coincidence data have been taken for the decay of 100-min Nd-149 to levels of Pm-149, 12-min Nd-151 to levels of Pm-151, 5-hr and 20-min Pd-111 isomers to levels of Ag-111, and 3.4-hr and 2.6-hr Cd-117 isomers to levels of In-117.

THE NON-DESTRUCTIVE DETERMINATION OF TRACE ELEMENT CONCENTRATIONS BY NEUTRON-CAPTURE Y-RAY SPECTROMETRY

G. E. Gordon, W. H. Zoller, D. Anderson, M. Failer, and W. B. Walters (University of Maryland, College Park, MD)

In cooperation with Dr. Richard Lindstrom of the NBS Activation Analysis Section, efforts are underway to develop a neutron-capture γ -ray spectrometer for the purpose of non-destructive trace-element concentration measurements. The system is to include a split-annulus Compton-suppressed Ge(Li) pair spectrometer whose pulses are automatically collected and analyzed by an 80K PDP 11/34. Construction of the reactor beam tube is underway (with design help and some machining by the Reactor staff) and the fabrication of external shielding is being planned.

The computer system has been ordered and the development of the software system is advancing.

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