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THERMOANALYTICAL CHARACTERIZATION OF ELECTRONIC MATERIALS

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ABSTRACT

The diverse demands of the electronics and communications industries require a very wide range of materials, e.g. metals, ceramics, semiconductors, polymers, etc. Essential to the ability to create or develop new materials and devices, is the power to characterize and understand the critical properties and processes involved. A wide range of thermoanalytical method frequently perform a crucial role in gaining these insights. A broad survey of applications is described in a general manner to illustrate the versatility of thermal analysis in this field and a few prominent examples are selected and discussed in greater detail.

INTRODUCTION

There are many crucial aspects involved in the success and progress of the electronics and communications industries. Certainly one of the most important factors is the creation of new materials and processes. An essential step in this chain is the ability to adequately characterize these materials and processes. Many methods are usually required to achieve this goal and thermoanalytical techniques frequently play a prominent role.

It is well beyond the scope of this brief paper to attempt to extensively review the enormous overlap between the fields of thermoanalytical methods and material science as it applies to the electronics industry. Instead a brief survey is made to show the great versatility of thermal analytical techniques and the general areas where productive applications have been made. A number of these applications are then selected and discussed in somewhat greater detail but avoiding the specific example used in earlier reviews.[1,2] The selection of examples and references is somewhat arbitrary and by no means meant to represent the definitive choices.

In describing this rich area of overlap between thermal analysis and materials of interest to the electronics industry, it is possible to view it from either side, i.e., by sequentially considering the individual thermoanalytical techniques or from the standpoint of classes of materials. The latter format is adopted herein but an effort will be made to describe the full range of the most widely used thermoanalytical methods. Table I lists some of the common classes of materials and general areas in which thermal analysis can contribute. Each area of application is equally applicable to each class of material.

METALS

The structural uses of metals are obviously numerous within the electronics industry but these uses are

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TABLE I

Examples of the Applications of Thermal Analysis and Classes of Materials of Interest to the Electronics Industry

Class of Material	Areas of Application
Metals	Phase and Structural Relationships
Ceramics and Glasses	Composition/ Impurities, and Defects
Semiconductors	Synthesis
Polymers Plastics	Thermal Stability and Reactivity
Simple Inorganic, Organic, and Organometallic Compounds	Thermal and Physical Properties

common to industry as a whole and the thermoanalytical applications are better left to a review on metallurgy in general. The use of metals as spring type contacts in relays and switches, however, does place unique demands upon the mechanical properties and fatigue. These demands are generally met by the careful design of alloys and control of microstructure. Phase diagrams play a fundamental role in such designs and thermal analysis, DTA in particular is most useful in phase studies.[3]

Electrical conductivity, magnetic properties, and a high degree of corrosion resistance are the attributes most relevant in the specific context of this review. Virtually all electrical devices depend upon conduction through metals in the form of wire; thick film derived from fired pastes or electroplating; and/or thin films formed by evaporation, sputtering, chemical vapors deposition, etc. Usually a continuous electrical path within a device is not practical and frequent interconnections are required. A common means of connections is simply a press or spring loaded fit between metallic contacts. Preservation of this electrical contact requires a high degree of corrosion resistance and gold plating is often used for critical connections. Evolved gas analysis (EGA) has been shown to be a valuable thermoanalytical method to indicate the nature of impurities trapped in the film during the plating process and how the quantities of these impurities vary with plating conditions.[4]

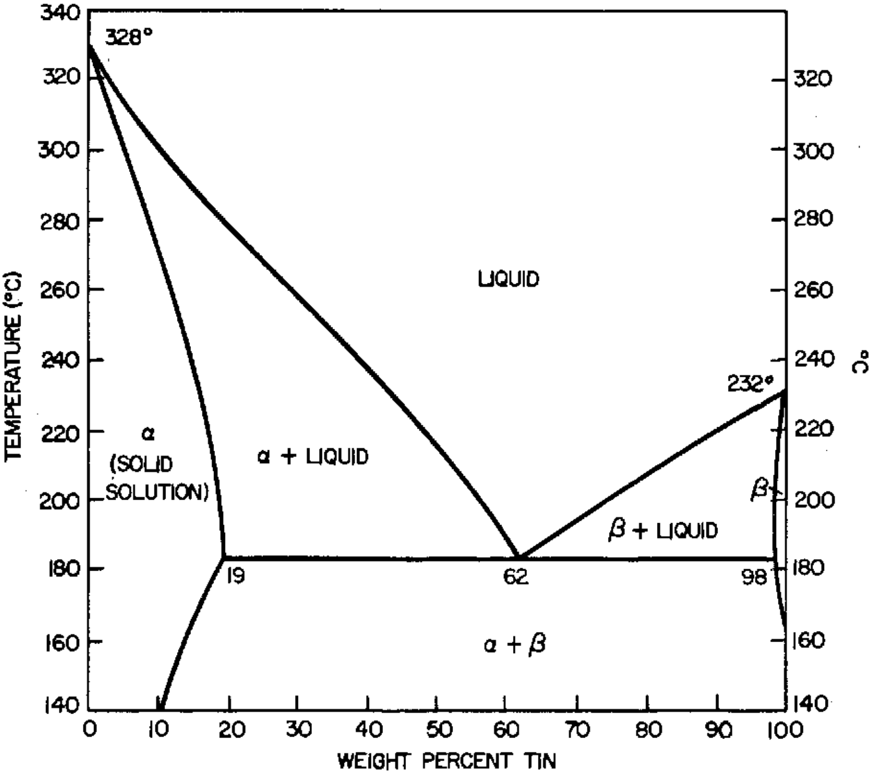
Certainly one of the most important means of electrical interconnection, however, is by means of soldering. A fine example of the value of thermal analysis in this field is the recent study by Kuck[5] showing how relatively fast simple DSC experiments can be made to determine the composition of common Sn/Pb solders. The study arose from a failure analysis of poor reflux-flow solder connections. In this application a Sn/Pb solder was electroplated onto metallic pins and the solder made to flow onto the mating part with the aid of heat transferred by means of a refluxing, organic, liquid that boils (215°C) about 30°C above the desired melting point of the solder. Occasionally poor contacts with a rough sandy appearance were obtained rather than the smooth appearing good contact. This was traced to the solder composition. The Sn/Pb contents of the plated solder were established from a comparison of the observed liquid temperature with that of a carefully established phase diagram of the Sn-Pb system derived from DSC measurements made under identical conditions. Samples were preheated to 350°C to melt them and then cooled rapidly prior to the measuring cycle at 5°C min^{-1} in N_2. Besides simple thermal hysteresis induced by a temperature lag, one must also contend with the common tendency of a liquid to supercool and the tendency on melting to lag the equilibrium situation because the last trace of solid has a melting point higher than the liquid with which it is in contact. Consequently, is is a common practice to average the temperature of first appearance of solid on cooling and the temperature of disappearance of the last solid in heating to determine the liquidus temperature. The resulting phase diagram is shown in Fig. 1. From comparisons of the observed electroplated solder samples with the phase diagram they, or at least portions of them, were found to be far from the desired eutectic composition. The convenience of this DSC technique permitted the further investigation of any inhomogenities within the electroplated solder or how the composition of the plated solder might vary with electroplating conditions. The Sn content, determined by this DSC method, is indicated in Fig. 2 for different locations about electroplated foils made at several current densities. It can be seen that higher current densities increase the Sn content in the plated material. The generally high Sn contents around the edge of the foils also indicate how the local current densities vary during electroplating. The value of thermal analysis to initial research in this general area and in quality control during subsequent manufacturing is clearly evident.

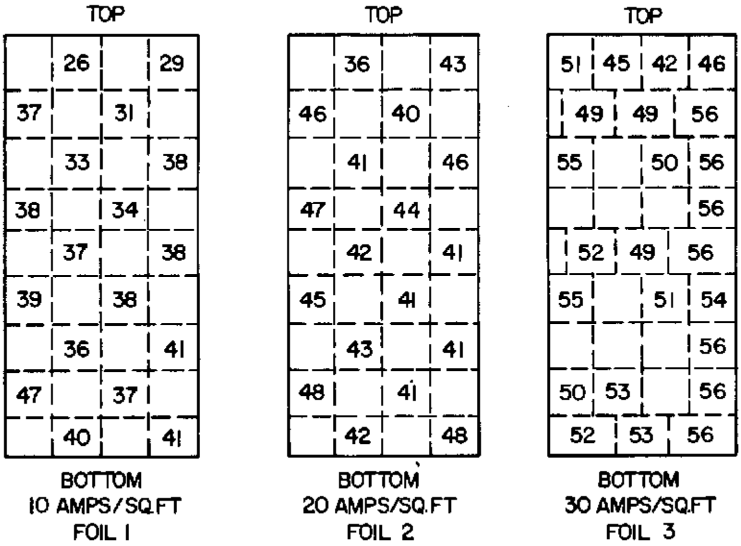
Some applications of thermal analysis associated with the magnetic properties of metal are 1) determination of composition based upon the temperature of the magnetic transition as determined by DSC, DTA, or thermomagnetometry (weight in a generally weak magnetic field), TM;[6] 2) utilizing TM to follow the appearance or disappearance of various magnetic phases during heat treatment;[7] and 3) evaluations of phase diagrams for magnetic alloys using DTA.[8]

CERAMICS AND GLASS

The technology of ceramics and glasses goes back to the earliest times, however, there has been a recent renisance in these materials and interest is rapidly burgeoning. Spearheading much of this progress are newer techniques of synthesis yielding greater homogeniety, purity, and controlled particle morphology. Some of these synthetic techniques rely upon the thermal decomposition of precursor compounds to prepare these improved materials. Obviously thermal analysis is an invaluable technique to follow the conversion of such precipitates or gels to, e.g. titanates,[9] niobate,[10] ferrites,[11] or glass.[12] Combined techniques such as DSC and x-ray diffraction[13] are particularly variable.

The use of glass films for the transmission of information on a modulated beam of light is leading to a revolution in the telecommunications industry. Present technology is largely centered on silica glass fibers using carrier wavelengths of less than 2 μm. A major thrust, however, is directed towards new glass systems which are transparent further into the IR, on the other side of the predominant OH absorption around 2.8 nm, where the Rayleigh scattering (with its λ^{-4} dependence) is much less of a factor and where absorption due to many of the common transition metal impurities is markedly lower. Determination of the range of glass formation and critical properties such as the glass transition temperature, T_g, and crystalization temperature are frequently measured by DTA, DSC, and thermomechanical analysis TMA. A wide range of halides[14,15] and chalcogenide[16,17] compositions are currently being investigated by thermoanalytical methods.





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50	53		56
52	53		56

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Figure 1. The Sn/Pb phase diagram using DSC and alloys of known composition.[5].

Figure 2. The % of Sn as determined by DSC, in Sn/Pb alloys (1 mil thick) electrodeposited on stainless steel plates.[5]

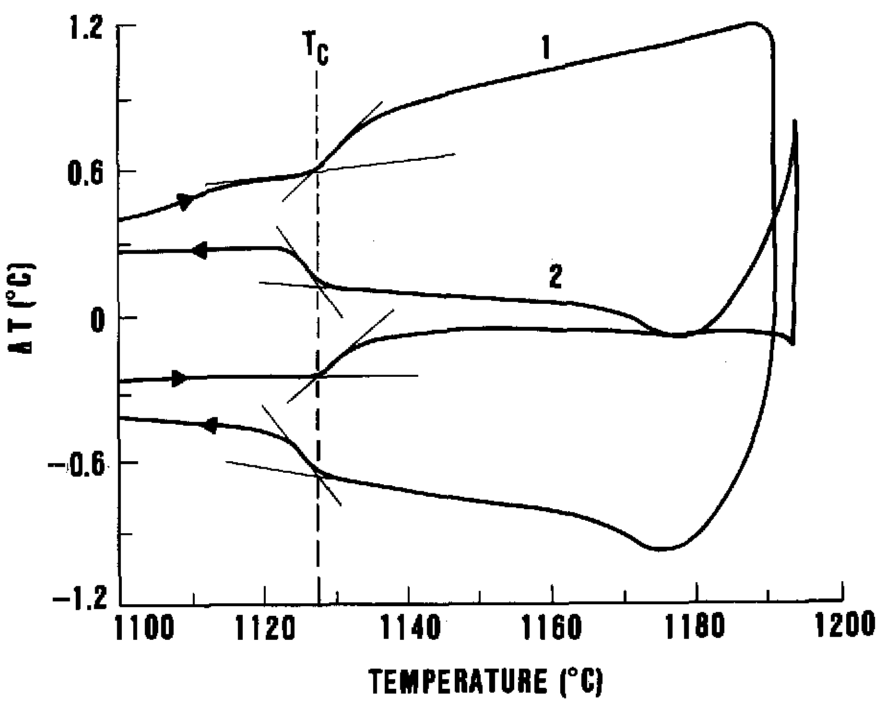
Besides the stable low-loss optical transmission medium, successful optical communication systems require many other devices of equal importance, e.g., coherent light sources, optical modulators/demodulators, and optical switches. Because of its desirable electro-optical coefficients and high Curie temperature (T_c), lithium niobate is a prime candidate for optical switching. There were, however, a number of uncertainties regarding its phase relationships that required confirmation or revision. These uncertainties arise primarily from the non-stoichiometry resulting from the fact that $LiNbO_3$ melts incongruently and consequently crystals grown by Czochralski techniques from the congruent composition are highly deficient in lithium and oxygen.

Questions then occur as the effects of these vacancies upon the electrical and physical properties of the crystal and upon a variety of possible chemical effects such as homogeniety diffusion rates, impurity incorporation, and second phase precipitation. Chemical analysis to determine the precise degree of non-stoichiometry is difficult and time consuming. It was noted, however, that the T_c of the material was quite sensitive to the degree of non-stoichiometry.[18] Because of the high temperature (>1100°C) and weak thermal effect (second order transformation) of the transition, it is necessary to perform careful DTA experiments. Dilatometric measurement may also be used to reveal the transition.[19]

The nature of the thermal effect is shown in Fig. 3. The value of T_c was taken from the extrapolated onsets of the departure from the base line during both the heating and cooling portions. The curves are unsmoothed and uncorrected for thermocouple calibration. It can be seen that the precision is relatively good and that there is little thermal hysteresis associated with the transition, even at 20°C min^{-1}. However, as the lithium and oxygen contents are increased towards the stoichiometric ($LiNbO_3$) composition, the value of T_c rises above the equilibrium solidus curve[20] and the weak transition is masked by the relatively large enthalpy change associated with the formation of the liquid phase. This is indicated in Fig. 4 where a greater range of the DTA curves is shown.

Fortunately it is possible to determine T_c for materials near stoichiometry by means of the change in thermal expansion coefficient associated with the transition. The effect, shown in Fig. 5, is much more pronounced along the c-axis than along the a-axis of the crystal. The problem associated with the volatilization of a Li_2O and segregation of Li-rich liquid phase can be seen in the lowering of the T_c during cooling. Since it was not necessary to go to as high a temperature for the congruent material, there is no significant change in T_c observed for that crystal.

The crystals as grown have the nominal composition associated with the congruently melting material[21] in order to obtain a homogeneous boule which resists cracking during cooling. The desired composition is then achieved by long time equilibration at elevated temperature in a controlled partial pressure of Li_2O.[22] Isothermal gravimetry has been particularly valuable in precisely define the composition by means of the change in weight between a sample before and after equilibration with the vapor. This "vapor phase equilibration" technique has been extensively described by Holman[23,24] and extended by others.[21,22,25] Similar high precision isothermal gravimetry[26] or thermogravimetry[27] is used to determine oxygen equilibrium with non-stoichiometric oxides such as the technologically important ferrites.



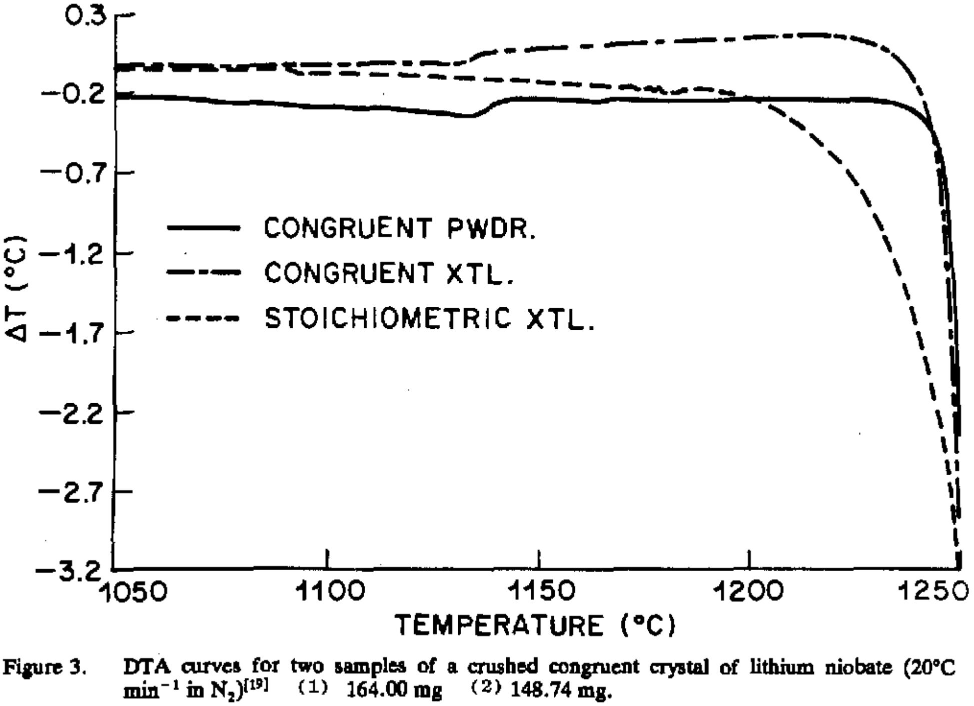
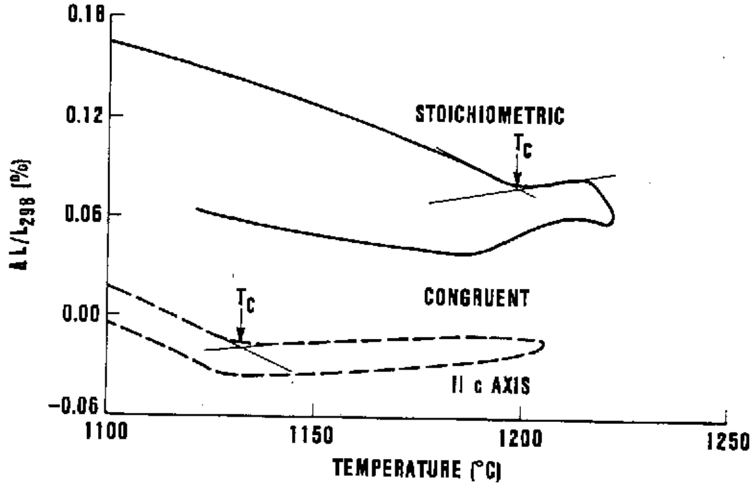


Figure 3. DTA curves for two samples of a crushed congruent crystal of lithium niobate (20°C $\min^{-1}$ in N_2)[19] (1) 164.00 mg (2) 148.74 mg.

Figure 4. DTA curves for congruent and stoichiometric lithium niobate (20°C $\min^{-1}$ in N_2)[19]



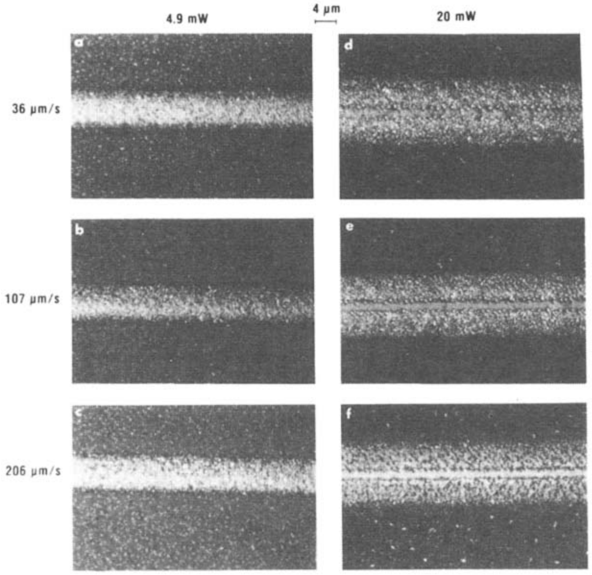


Figure 5. Thermal expansion along the C-axis for congruent and stoichiometric lithium niobate, above 1100°C (4°C min^{-1} in air)[19] ——— stoichiometric – – – congruent.

Figure 6. Optical micrographs of gold features from 1.0 μm NH_4AuCl_4/pol;y(vinyl alcohol) film (58 wt.% Au) on SiO_2. Laser powder and scanning speed are indicated.[30]

SEMICONDUCTORS

At present silicon based devices dominate the semiconductor market. Besides silicon itself, there are many other materials and processing steps involved in VLSI technology. Thin films of insulators, conductors, capacitors, protective glasses, etc. are part of the device while other materials such as photoresists and masks are essential in the processing. The quantities of materials involved in each device are small and consequently sensitive techniques are necessary to characterize these films. X-ray diffraction of these films as a function of temperature and sometimes even DTA can reveal the amorphous to crystalline transitions frequently encountered as wellas reactions between the films themselves or with the substrate. Sometimes these amorphous to crystalline transitions have catostrophic results on device performance, e.g., IrO_2 films for displays.[28]

Mass spectrographic evolved gas analysis (EGA) is a particularly valuable technique to determine impurities which may be incorporated in the films during deposition. Hydrogen has a pronounced effect upon the electrical properties of polysilicon films and hence its determination by EGA is valuable.[2] Similarly, EGA revealed the presence of Ar in $TaSi_2$ films which were prepared by sputtering in Ar or by CVD in an Ar carrier gas stream[29] The release of this Ar at later stages in the device processing caused problems so that the use of EGA to establish the conditions of minimum Ar incorporation in the films was of great value.

It is not generally adequate to simply deposit a film of suitable composition and properties. It must also be deposited in the desired pattern or places. Usually this is achieved through masking and etching processes, however, it can also be achieved in a more efficient manner by selective deposition or directly writing the pattern. An example of the later is the scribing of gold lines by initiating the thermal decomposition (by focused laser radiation) of a gold compound, dispersed in polymer films[30]. Examples of such lines are shown in Fig. 6. The effects of scanning speed and laser power upon the quality of the Au deposit is obvious. Clearly, the nature of the thermal decomposition and the energy required for both the gold compound and binder are factors in establishing the optimum processing conditions.

Results of TG and DTA studies of a gold ink system in air are presented in Fig. 7. The gold compound decomposed in weak endothermic steps around 200 and 300°C while the organic binder burn-off is highly exothermic at higher temperatures around 400°C. Table II lists the results of DSC experiments on both the ink and a model organometallic assumed to resemble that present in the ink. These energy values are than used in model, thermal transfer calculations associated with the absorption, consumption and dissipation of the laser energy.

Semiconductors made from elements in groups IIIA and VA of the periodic table have arrived and seem to form the basis of the next generation of applications. These materials have a much greater electron mobility and hence are desirable for high speed computers. From the important standpoint of semiconductors lasers, such as might be used in telecommunication over optical films, unique hetero-structures of these semiconductors can be made to provide sufficient optical powder over an appropriate range of wavelength.

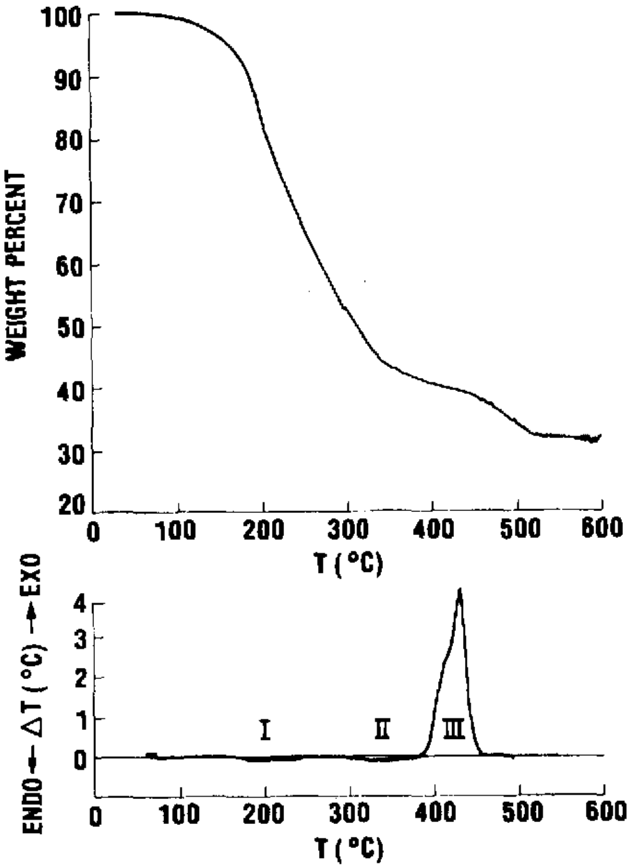


Figure 7. Thermal analysis of a commercial bright gold screening ink at 5° min^{-1} in air.[30]

TABLE II

Differential Scanning Calorimetry
(5°C/min in air)

Au p-ter-butylphenyl mercapitide	Au Screen Ink
173-225°C 21.7 cal/g	177-245°C 13.2 cal/g
263-270°C 21.6 cal/g	300-375°C 25.7 cal/g
	375-455°C -1,320 cal/g

Complications in the use and fabrication of these devices arise from the greater volatility and reactivity of III-V semiconductors at lower temperatures compared to silicon. Again the high sensitivity of EGA make it particularly well suited to investigate the threshold of decomposition[31,32] for such materials as well as ways to reduce this effect.[29] Similarly EGA techniques are valuable to evaluate the frequently premature interaction of these semiconductors with the metals used as electrical contacts.[33,34]

One of the principal methods by which the hetero-junction lasers are made is liquid phase epitaxial deposition (LPE). consequently the temperature of the liquidus curve as a function of composition must be accurately determined for the appropriate systems. Again the volatility and reactivity of these materials present problems in normal DTA or DSC studies. Osamura, et al.,[35] have surmounted these difficulties by designing a DTA apparatus which utilizes samples enclosed in quartz ampoules.

POLYMERS

Plastics and polymers are used in so many and diverse ways that it is hopeless to attempt to even scratch the surface in a few pages. One need only to scan the programs of major thermoanalytical conferences such as ICTA, NATAS, ESTAC, or this meeting of the Society of Calorimetry and Thermal Analysis of Japan to realize that this is one of the most rapidly growing areas of science. Fortunately, a very excellent, in-depth and up-to-date monograph on the thermal characterization of polymeric materials has been recently published.[36] Readers are referred to it in order to appreciate the many ramifications of this broad and important topic.

Among the most classic practical applications of DSC has been in studies of the oxidative degradation of polymers; in the context of this paper as it applies to electrical insulation of cable.[36] similarly the effects and nature of water in polymers upon the quality and stability of electrical insulation have been most profitably studied by thermoanalytical methods.[37] Virtually all of the key plastic and polymeric components, e.g., circuit boards, instrument housings, encapsulants, adhesives, insulation, connectors, binders or vehicles in tapes and inks, etc. have been extensively studied in some form by thermal analysis.

There has been recent interest in pyrolizing organic polymers to provide low temperature synthetic routes or products of somewhat unique properties. Some of these may be done in inert atmospheres to yield carbon, carbides, nitrides etc. If the polymers or organic precursors contain high contents of oxygen, or if the decomposition is performed in an oxidizing atmosphere, then stable oxides may be products. As an example consider the thermal decomposition of a film of an organosilicon resin to form a SiO_2 film.[38]

Thermogravimeteric curves are shown in Fig. 8 for platlets of the resin, about 1.2 mm thick and 16 mg, heated in various atmospheres at 10°C min^{-1}. In pure O_2 the specimen ignited around 480°C and gave rise to a rapid weight loss with undoubtably much self heating. In pure N_2 the sample decrepitated around 670°C, presumably due to the high internal pressures created by the trapped volatail products of the decomposition. The behavior in air was intermediate in that it neither ignited nor disentigrated prior to reaching the weight loss associated with the formation of SiO_2.

Selected results of the mass spectrographic EGA are presented in Fig 9 for a sample heated at 10°C min in a vacuum of $\leq 1 \times 10^{-7}$ *torr* (in the absence of any outgasing). Both the TG curve in ambient atmosphere, N_2, and the EGA in vacuum suggest basically a two stage decomposition process. The initial step centered around 300°C corresponds to curing of the resin while the methane evolved at higher temperatures corresponds to the decomposition of the organosilsesquioxane polymers. The dual nature of the evolution in vacuum is attributed to an initial breakdown near the surface and at a higher temperature due to the release of the internal products via cracks, pores, bubbles, etc. The IR results of isothermally treated film of the resin on Si are consistent with the general picture described above. The bands associated with Si-CH_3 linkages are at 2990, 1275, 780 and 410 cm^{-1} and they decrease in intensity continuously with increasing temperature until, after 30 min at 650°C, only the bands attributed to Si-O-Si linkages are left, 1080, 800 and 450 cm^{-1}.

SIMPLE INORGANIC AND ORGANIC COMPOUNDS

Sometimes the thermal decomposition of complex precursor compounds can lead directly to the formation of mixed compounds such as of those briefly mentioned earlier under ceramics. At other times, simple components of a controlled particle size, reactivity or defect structure may be desired for subsequent processing. Pigments or catalysts might be appropriate examples in other industries, but in the electronics industry iron oxides represent an ideal example. Obtaining the proper particle size and morphology in the conversion of precursor compounds to $\gamma-Fe_2O_3$ is of immense importance in the production of magnetic recording types. Similarly controlling the reactivity of the $\alpha-Fe_2O_3$ used to prepare ferrites is also necessary.

This latter process has been extensively investigated.[39-41] Defining reactivity is a non-trivial

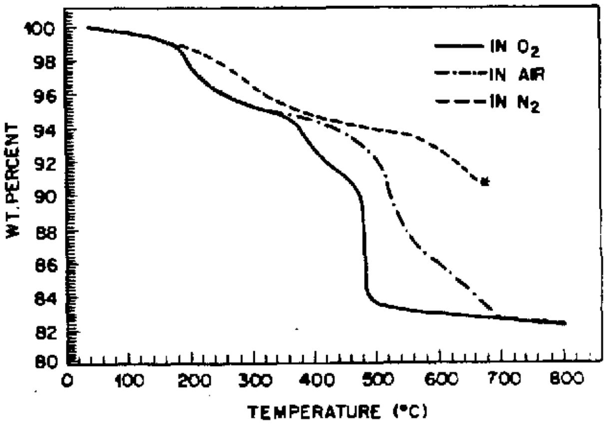
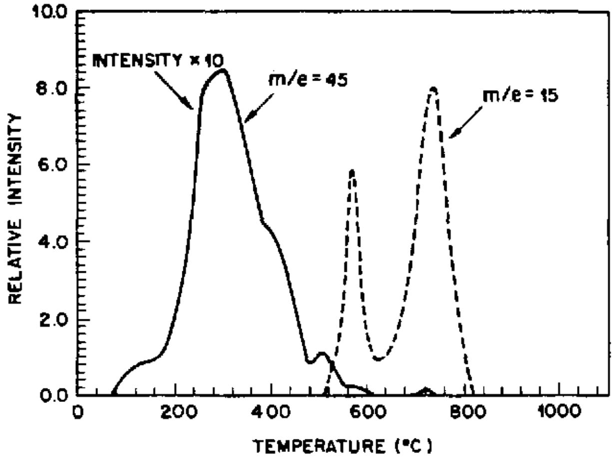


Figure 8. TG curves Owens-Illinois resin GR650 at 10°C min^{-1}.[38]



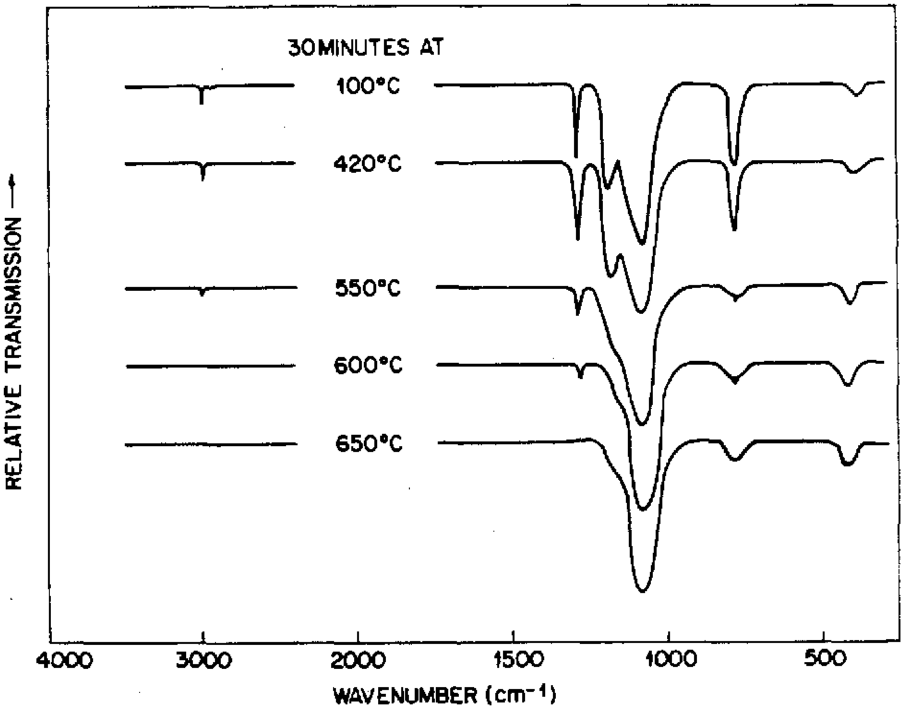


Figure 9. Mass spectrographic EGA curves for Owens-Illinois resin GR650 at 10°C min^{-1} and about 1×10^{-6} *torr*.[38]

Figure 10. Transmission IR spectra of 8000Å films of Owens-Illinois near GR650 after heat treatment in dry N_2 for 30 min at the indicated temperatures.[38]

matter. The temperature regime in which the experiments are performed has a very significant influence on the outcome of any evaluations. Comparison of numerous samples of $\alpha-Fe_2O_3$ prepared from many different iron salts by several different methods, indicated that the order of reactivity depended upon the test method. The order of reactivity at room temperature, as determined by the solubility of the oxide in an acid, is quite different from the order determined by TG for the reaction of $\alpha-Fe_2O_3$ with Li_2CO_3 or, at higher temperatures yet, by the ability of the $\alpha-Fe_2O_3$ to sinter to high density.

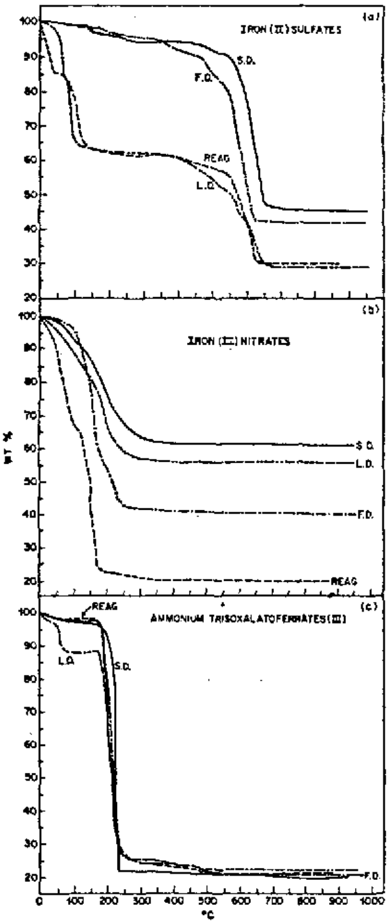
Thermogravimetry contributed to these studies in several ways. The point of conversion from the precursor compound to Fe_2O_3 could be determined from TG curves such as those shown in Fig. 11. The point of final weight loss was much different for each compound and somewhat dependent upon the nature of preparation. The reactivity at intermediate temperatures, the best indicator of reactivity as applied to the problem of ferrite production, was determined by following the reaction of the Fe_2O_3 samples with Li_2CO_3 as shown in Fig. 12. A summary of these results is given in Table III. The order of reactivity for the various samples of Fe_2O_3 was arbitrarily taken from the temperature at which the reaction with Li_2CO_3 was to 85% complete.

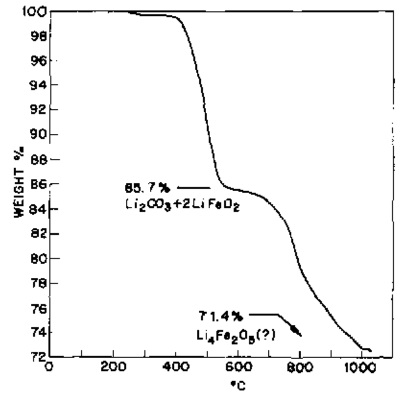
The particle size of the Fe_2O_3 can be controlled by the temperature and time used during calcination of the precursor.[39] It is an interesting question to ask whether there are variations in defect structure as well which might influence the subsequent reactivity of the powder. An isothermal gravimetric study[40] of the kinetics for the reaction of Li_2CO_3 with several samples of $\alpha-Fe_2O_3$ prepared by the calcination of $(NH_4)_2Fe(SO_4)_2$ at different temperatures would suggest not. Arrhenius plots of this study are presented in Fig. 13. The activation energy appears to be the same for each preparation and the increased activity as the surface area increases must therefore be reflected in the pre-exponential term. Activation energies do, however, vary somewhat with the precursor selection.

This high reactivity of the finely divided oxide products can have a pronounced effect upon the temperature of subsequent reaction, particularly if the reactants can be intimately mixed. An ideal example of such mixing occurs during the decomposition of a precursor compound which yields such an intimate mixture of fine powders. The preparation of $BaSnO_3$ from both a conventional mixture of $BaCO_3$ and SnO_2 and from the decomposition of $BaSn(C_2O_4)_2 \cdot 0.5H_2O$ has been studied using thermal analysis and x-ray diffraction.[42,43] The results indicate that because of the shorter diffusion paths present in the decomposed mixture of the precursor, the reaction to form $BaSnO_3$ proceeds without the detectable formation of Ba_2SnO_4 intermediates and at temperatures several hundred degrees lower than the convectionally mixed material.[43]

CONCLUSIONS

Thermal analysis is of great importance whenever information regarding the temperature or energy associated with various transformations or reactions, thermal stability, nature of the thermal decomposition products, or the effects of thermal history upon the properties of the material is required. The subsequent reactivity of powders prepared in several manners can be compared. In





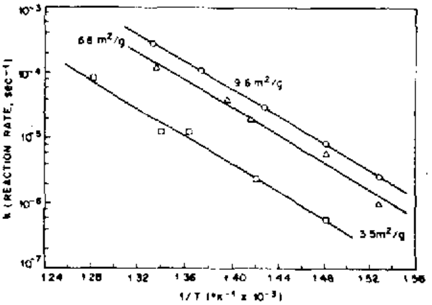


Figure 11. TG curves for various iron salts. REAG = reagent grade solid, S.D. = spray dry solution, F.D. = freeze dried solution, an L.D. = liquid dried solution.[39]

Figure 12. TG curve for $2Li_2CO_3 + Fe_2O_3$, 5 mg, 2.5°C min^{-1}, in air.[41]

Figure 13. Arrhenius plot for the reaction of Li_2CO_3 with Fe_2O_3 derived from $(NH_4)_2Fe(SO_4)_2$. The surface area of the Fe_2O_3 is indicated on the plots in m^2g.[40]

short, thermoanalytical techniques possess great power and versatility. The boundaries of their application to solve problems in the electronics and communications industries related to the characterization of materials and the understanding of both physical and chemical processes is constrained only by the limits of the investigator's imagination, time and budget.

TABLE III

Decomposition Temperatures of Iron-Lithium Mixtures

Iron salt	°C Calcined iron salt	°C 85% Decomp. Li_2CO_3
Iron (III) sulfate	700	535
Ammonium iron (III) sulfate	660	495
Iron (II) sulfate	720	580
Ammonium iron (II) sulfate	660	490
Iron (III) nitrate	390	570
Ammonium hexacyanoferrate (III)	670	600
Ammonium hexacyanoferrate (II)	480	840
Iron (III) oxalate	325	560
Ammonium trisoxalaftoferrate (III)	500	560
Iron (II) acetate		
Ammonium iron (III) citrate	580	800
Magnatite + ethylenediaminetetraacetic acid	525	615
Commercial iron (III) oxide		670
Lithium Carbonate		950

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