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Experimental Metamorphic Petrology

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EXPERIMENTAL METAMORPHIC PETROLOGY

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Introduction

Since Bowen and Tuttle's pioneering study of the system MgO-SiO_2-H_2O in 1949, advances in experimental metamorphic petrology have occurred steadily rather than in "leaps and bounds". The number and quality of papers published during the past quadrennium, 1975-1978, attests to the health of the science. Although the purpose of this report is to focus international attention on the U.S. effort in experimental metamorphic petrology, some papers published by foreign experimentalists have been included, especially where their contributions complement those made in the U.S. To keep the review current, abstracts of papers read at national meetings of the Geological Society of America and the American Geophysical Union in 1978 are included.

Coverage is generally limited to papers reporting experimental data acquired at pressures below 10 kbar and temperatures between 200°C and the solidus for the system in question. Several papers reporting calorimetric and experimental data for pressures and temperature outside these limits were included to demonstrate how experimental metamorphic petrology interfaces with calorimetry, low temperature geochemistry, and experimental igneous petrology.

Papers are grouped into nine categories and are cross-referenced where necessary. Papers reporting experimental data for silicates have been subdivided according to the structural group of the dominant phase or phases whereas papers dealing with non-silicates are grouped together regardless of structure or composition. Papers dealing with multicomponent chemical systems are grouped under the heading "Rock Systems". Papers dealing with experimental techniques or with the calculation of thermochemical parameters of minerals are grouped under the headings "Experimental Techniques" and "General Physical Chemistry", respectively.

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Discussion

With the realization that many important equilibria could not be investigated in the laboratory, experimentalists turned to thermodynamic theory in order to extrapolate experimentally determined equilibria to conditions not amenable to experiment, to calculate the effects of additional components, and to evaluate the consequences of varying intensive parameters. Unfortunately, many of the early experimental data are not sufficiently accurate for such calculations. In order to be useful for thermochemical calculations, phase equilibrium data must be reversed, activities of components in the fluid phase must be determined, and solid phases must be thoroughly characterized chemically, mineralogically, and structurally (order-disorder, vacancies, polytypism, etc.).

Several techniques developed or refined during the past quadrennium will aid the experimentalist in controlling fluid composition in hydrothermal experiments. The "Shaw apparatus" was redesigned [Frantz et al., 1977] because H_2-equilibration times in the original design resulted in f_{O_2} values lower than the equilibrium values [Hewitt, 1977]. Chou [1978] recalibrated the (FMQ), (MH), (MnO-Mn_3O_4), (NNO), and (Co-CoO) oxygen fugacity buffers; Hewitt [1978] recalibrated (FMQ). Although Hewitt's recalibration differs from Chou's by no more than 0.5 log units, the discrepancy must be resolved if the phase equilibrium data generated using (FMQ) are to be used for thermochemical calculations. The Ag + AgCl acid buffer was recalibrated by Chou and Frantz [1977]; Frantz and Popp [1977] modified this buffer technique in order to determine speciation in hydrothermal fluids. Hallam and Eugster [1976] developed a method for buffering fluids in the system N-O-H by combining the (Cr + CrN) nitrogen buffer with conventional hydrogen buffers. Modifications of a chloride electrode and a flow-cell calorimeter by Popp et al. [1978] allow accurate and precise measurement of chloride ion in the concentration range 0.01 to 0.0001 m in microvolume samples (1-10 μℓ).

Ziegenbein and Johannes [1977] designed a device which aids in the extraction and gaschromatographic analysis of fluids present in high pressure experiments. Schmid et al. [1978] developed a sensitive method for determining reaction direction in experiments where one of the reacting phases is a solid solution of two components. Navrotsky [1978] designed a microcalorimeter which can accept a cold-seal pressure vessel and is using the apparatus to measure enthalpies of aqueous solutions at pressures to 2 kbar and temperatures to 800°C.

During the past quadrennium, experimental studies were initiated to determine the compositional limits of micas [Hazen and Wones, 1978; Franz and Althaus, 1976], phlogopite [Robert, 1976a, b], brittle micas [Olesch, 1975], kornerupine [Werding and Schreyer, 1978], viridine [Abs-Wurmbach and Langer, 1975], and sapphirine [Bishop and Newton, 1975]. Successful synthesis of clinoptilolite [Goto, 1977], spodumene [Drysdale, 1975], carpholite [Mottana and Schreyer, 1977], and a triple chain silicate [Tateyama et al., 1978] was achieved.

Among the dehydration equilibria whose locations in P_{H_2O}-T space have been refined are reactions involving anthophyllite [Ravior and Hinricksen, 1975; Chernosky, 1976], phlogopite [Wones and Dodge, 1977], tremolite [McKinstry and Skippen, 1978], sillimanite + biotite + quartz [Hoffer, 1976], clinochlore [Staudigel and Schreyer, 1977], clinochlore + quartz [Chernosky, 1978], portlandite, and brucite [Irving et al., 1977]. Experimentalists have successfully reversed equilibria involving bicchulite [Gupta and Chatterjee, 1978], margarite [Chatterjee, 1976], lizardite [Chernosky, 1978], gehlenite [Huckenholz, 1977], and antigorite [Evans et al., 1976].

Considerable effort has been expended in determining the stabilities and phase relations of crystalline solutions. Such studies require that the compositions of the coexisting phases be determined. Determinative curves relating the unit cell parameters of a phase to its composition have been experimentally calibrated for piemontite [Anastasiou and Langer, 1977], Fe-Mg orthoamphiboles [Popp et al., 1976], orthoamphibole [Maresch and Langer, 1976], Fe-Mg-Al biotites [Hewitt and Wones, 1975], Mg-Al serpentines [Chernosky, 1975], nepheline-kalsilite [Ferry and Blencoe, 1978], OH-F amblygonite [Loh and Wise, 1976], edenite-pargasite [Hinricksen and Schürmann, 1977], pargasite-richterite [Braue and Seck, 1977], and grandidierite [Olesch and Seifert, 1976].

Among the crystalline solutions whose stabilities and phase relations have been determined are nepheline-kalsilite [Ferry and Blencoe, 1978], pyrope-grossular [Henisen, 1976], uvarovite-andradite and uvarovite-grossularite [Huckenholz and Knettel, 1976], grossularite-spessartine [Hsu, 1978], pargasite-richterite [Braue and Seck, 1977], richterite-ferrorichterite [Charles, 1975, 1977], actinolite-cummingtonite [Cameron, 1975], hastingsite [Charles, 1978], grunerite [Forbes, 1977], Fe-Mg amphiboles [Popp et al., 1977a], chlorite [McOnie et al., 1975; Fleming and Fawcett, 1976; James et al., 1976], serpentine [Moody, 1976], cordierite [Holdaway, 1976; Holdaway and Lee, 1977; Lee and Holdaway, 1976], calcium brittle micas [Olesch and Seifert, 1976], and pyroxenes [Warner, 1975; Lindsley and Dixon, 1976; Ikeda and Yagi, 1977; Herzberg, 1978; and Wood, 1978].

Experimental calibration of the temperature, pressure and compositional dependence of major and trace element distribution coefficients may enable them to be used as geothermometers and geobarometers. The partitioning of Fe and Mg between biotite-garnet [Ferry and Spear, 1978], garnet-phengite [Krogh and Råheim, 1978], olivine-spinel [Engi, 1978], and biotite-salt solution [Schulien, 1975] were determined experimentally. Rajamani [1976] investigated the distribution of Fe, Co, and Ni between sulfide and orthopyroxene. Jacobson and Usdowski [1976] investigated the partitioning of Sr between calcite and dolomite, Suvorva and Tenishev [1976] determined the distribution of sulfur isotopes between Mo, Pb, Zn and Sn sulfides, and Fournier [1976] studied the exchange of Na^+ and K^+ between water vapor and feldspar. Fluorine-hydroxyl exchange was experimentally investigated for topaz [Rosenberg, 1978], muscovite [Munoz and Ludington, 1977], and amblygonite [Loh and Wise, 1976].

Experimental data for reactions involving a C-O-H fluid phase have been obtained for sphene [Hunt and Kerrick, 1977], andradite [Taylor and Liou, 1978], grossular, and wollastonite [Shonulovich, 1977], rhodochrosite [Candia et al., 1975], siderite [Chou, 1978], and zeolites [Ivanov and Gurevich, 1975]. Novgorodov [1975, 1977] investigated the solubility of quartz in H_2O-CO_2 mixtures. Locations of key equilibria in multicomponent systems involving C-O-H fluids were experimentally determined by Slaughter et al. [1975], Hewitt [1975], Metz [1976], Zharikov et al. [1977], and Puhan [1978]. Devolatilization equilibria in graphitic systems were discussed by Ohmoto and Kerrick [1977].

The stability relations of scapolite received considerable attention from experimentalists; Orville [1975] and Goldsmith and Newton [1977] investigated scapolite-plagioclase phase relations, Ellis [1978] investigated chloride and carbonate bearing scapolites, Newton and Goldsmith [1975] investigated the stability of meionite, and Goldsmith [1976] discussed the role of scapolite as a reservoir of CO_2 and sulfur in the lower crust.

Natural metamorphic fluids contain acids, salts, ions, and complexes as well as gases. The composition of a fluid in equilibrium with a particular metamorphic mineral assemblage can in principle be determined provided the assemblage contains fugacity indicators and appropriate buffer and exchange reactions [Eugster, 1977]. The success of this approach is limited because thermodynamic data for aqueous species at high temperatures and pressures is often unavailable. Although laboratory calibrations of equilibria involving multicomponent gases and aqueous electrolytes are uncommon, solubility studies continue to provide valuable information on the stabilities and thermochemical parameters of minerals and solutions. Solubilities of iron [Popp and Frantz, 1977], fluorspar [Malinin, 1976], wollastonite [Gunter and Eugster, 1978], mullite [Ostapenko et al., 1975], talc, antigorite, forsterite, chrysotile, brucite, and

enstatite [Hemley et al., 1977a, b] were measured. Frantz and Popp [1978] investigated speciation of aqueous $MgCl_2$ in the system $MgO-SiO_2-H_2O-HCl$ and found that at a total pressure of 2 kbar, associated $MgCl_2$ is the dominant magnesium species in the fluid at temperatures above 550°C whereas Mg^{2+} ions were dominant at temperatures below 400°C. Experimental data for solution-rock equilibria have been obtained for seawater-basalt [Mottl and Holland, 1978], seawater-peridotite [Seyfried and Dibble, 1978], seawater-andesite [Liou and Dickson, 1978], NaCl solution-carbonate rock [Radtke et al., 1978], and aqueous chloride solution-two mica schist [Vidale, 1975].

Textural relations in metamorphic rocks and ore deposits suggest that sulfides, silicates, and oxides often coexist stably. Experimental studies on sulfide-silicate phase relations remain uncommon despite the economic importance of sulfide deposits. A notable exception is the study of amphibole-magnetite-pyrrhotite phase equilibria by Popp et al. [1977] which hopefully will pave the way for future experimental work on sulfide-silicate phase relations. We must understand how and to what extent metals and sulfur dissolve and are transported in order to understand the ore deposition process. Experimental studies on the replacement of marble by sulfides [Howd and Barnes, 1975], on the mobility of metals in aqueous solutions [Govett et al., 1976; Giblin, 1978], on the solubility of chalcocite [Crerar and Barnes, 1976], scheelite [Foster, 1977], iron [Popp and Frantz, 1977], barite [Blount, 1977], and pyrite + pyrrhotite + magnetite [Crerar et al., 1978] provide the type of information required to understand ore genesis.

The processes of nucleation and growth (including diffusion and reaction rates) require further investigation before mechanisms by which metamorphic reactions proceed can be fully understood. Laboratory studies which enhance our understanding of diffusion and reaction rates include those of Hoffman et al. [1975], Wyart [1975], Kalinin and Shapovalova [1975], Seifert and Virgo [1975], Sipling and Yund [1976], McCallister and Yund [1977], Kay [1973], Sakai and Dickson [1978], Ildefonse and Gabis [1976], Lagache [1976], and Grandstaff [1976].

Methods for calculating reaction entropies and enthalpies from reversed phase equilibrium data were proposed by Gordon [1977] and Chatterjee [1975, 1977]; Anderson [1976, 1977] discussed uncertainties involved in such calculations and suggested that errors could be evaluated using a Monte Carlo method. Techniques for calculating and extrapolating equilibria in $P-T-X_{CO_2}$ space have been discussed by Skippen [1975], Skippen and Carmichael [1977] and Kerrick et al. [1976] and have been compared by Kerrick and Slaughter [1976] and Kerrick and Jacobs [1978].

Thermodynamic parameters for diaspore, margarite, pyrophyllite, zoisite, wairakite [Chatterjee, 1976], bicchulite [Gupta and Chatterjee, 1978], hydrous phlogopite [Wones and Dodge, 1977], talc, anthophyllite, enstatite, chrysotile [Zen and Chernosky, 1976], and andradite [Taylor and Liou, 1978] were extracted from phase equilibrium data. Thermodynamic properties of mullite [Ostapenko et al., 1975], barite [Blount, 1977], talc, antigorite, anthophyllite, chrysotile [Hemley et al., 1977a, b], and supercritical aqueous $CaCl_2$ [Gunter and Eugster, 1978] were extracted from solubility data.

The relationship between activity and composition for rock-forming crystalline solutions is required to calculate equilibrium conditions in complex natural systems using thermodynamic principles. Activity-composition relations for pyrope-grossular [Hensen et al., 1975], almandine-grossular [Cressey et al., 1978], muscovite-paragonite [Chatterjee and Froese, 1975], magnetite-ilmenite [Lindsley, 1978; Spencer and Lindsley, 1978], high albite-sanidine [Blencoe and Merkel, 1978] were extracted from phase equilibrium data using various solution models.

Procedures for estimating thermodynamic properties of minerals have been proposed by Chen [1975], Saxena [1976], Tardy and Garrels [1976, 1977], Tardy and Gartner [1977], Tardy and Vieillard [1977], Dibble and Dickson [1978], and Helgeson et al. [1978]. Ulbrich and Waldbaum [1976] emphasized the need for considering structural contributions to the third-law entropies of silicates. The thermodynamics of metamorphic fluids was discussed by Ryzhenko [1976], Eugster [1977], and Holloway [1977]; an equation of state for aqueous species at infinite dilution has been predicted by Helgeson and Kirkham [1976]. Fugacity coefficients for CO_2 have been calculated by Shmulovich and Shmonov [1975].

Among the minerals investigated calorimetrically are garnets [Kiseleva, 1976b; Kiseleva and Topor, 1976; Perkins et al., 1977; Kolesnik et al., 1977; Kiseleva, 1977; Newton et al., 1977; Charlu et al., 1975, 1978], pyroxenes [Navrotsky and Coons, 1976; Newton et al., 1977; Thompson et al., 1978; Charlu et al., 1975], spinel [Navrotsky and Kasper, 1976], gedrite [Kolesnik et al., 1976], sapphirine [Kiseleva, 1976a], gibbsite [Hemingway and Robie, 1977; Hemingway et al., 1977], kaolinite [Hemingway et al., 1978], muscovite [Robie et al., 1976], low albite [Hemingway and Robie, 1977], anorthite [Robie et al., 1978] and alkali feldspar [Hovis and Waldbaum, 1977]. Comprehensive computer programs designed to evaluate and correlate calorimetric and phase equilibrium data were written by Haas and Fisher [1976] and Helgeson et al. [1978]. During the quadrennium, two internally-consistent compilations of thermodynamic properties of rock-forming minerals were published; one is based on calorimetric data [Robie et al., 1978] whereas the other is primarily based on phase equilibrium data [Helgeson et al., 1978]. Although the thermochemical properties for many minerals in these compilations are similar, there are significant differences which the experimentalist is challenged to resolve.

The Future

One of the chief goals of the modern metamorphic petrologist is to determine the physiochemical conditions prevailing at the time a given metamorphic rock was recrystallized. Toward this end, experimentalists have explored the role of intensive parameters such as total pressure, water pressure, temperature, and oxygen fugacity in detail. Until recently,

however, the nature, composition, and role of the supercritical metamorphic fluid has not received much attention from experimentalists. Now that techniques for studying acid-base, dissociation, and solubility reactions at elevated temperatures and pressures have been developed, experimental work on the solution chemistry of metamorphic supercritical fluids will flourish.

The petrologic community is in need of experimental data for a wide range of topics: activity-composition relations for most rock-forming crystalline solutions need to be determined; diffusion coefficients for aqueous species and ions at metamorphic conditions are virtually unknown; the distribution of major and trace elements among most important metamorphic mineral pairs remains to be calibrated experimentally. Unfortunately, the current funding picture and the paucity of academic positions for scientists trained in experimental petrology preclude radical changes in the level of activity. Consequently, equilibria which provide the most useful information must be identified in order to maximize efficient use of equipment and time.

In the past decade it has become evident that phase equilibrium data can be used as input for thermochemical calculations. Consequently, considerable effort will be directed toward investigating relatively simple equilibria involving key phases even though such reactions may not be observed to occur in rocks. Although the traditional approach which involves mapping out the stability field for a particular mineral or assemblage will be retained, experimental techniques must and will be sharpened to the point that brackets 5-10°C wide will be routinely obtained. Narrow brackets are necessary if the thermochemical parameters of minerals are to be refined. Precise and accurate techniques for determining the composition of fine-grained material must be developed. The discovery of triple chain silicates in natural and synthetic phases [Veblen et al, 1978], for example, emphasizes the need to evaluate starting materials more critically. Interlaboratory calibration of hydrochemical equipment is necessary to insure internal consistency among data obtained in different laboratories.

The ability to measure high temperature heat capacities of relatively small synthetic samples with a differential scanning calorimeter is a major development. High temperature heat capacity data for many rock-forming minerals and crystalline solutions will become available during the next quadrennium. The possibility of routinely measuring the high temperature heat capacities of phases used in starting materials for hydrothermal experiments now exists!

The development of computer programs for correlating and extrapolating phase equilibrium and calorimetric data challenges the experimentalist to insure that new experimental data are consistent with the corpus of existing phase equilibrium and thermodynamic data.

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VOLCANOLOGY

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Volcanology in the 1970s has expanded greatly in both size and scope. Comparison of the references listed here with those of the U.S. National Report to IUGG for 1963-6 (by R. W. Decker) and 1967-70 (by T. L. Wright)* illustrates many of these changes. The number of references has doubled, reflecting increased interest in active volcanoes as well as the overall increase in scientific literature. Kilauea has been at least twice as active as any other oceanic volcano during the last decade, and its references (48) continue to dominate work on individual U.S. volcanoes. Alaskan volcanoes, 33 of which have erupted in the last 100 years, continue to be the most neglected of U.S. volcanoes (total of 24 references). On the other hand, those of the western conterminous U.S., only one of which is known to have erupted in the last 100 years, have received far more attention in the 1970s (21% of the references here, compared with 14% and 7% in the '67-70 and '63-66 reports, respectively). Much of this increased attention stems from the search for geothermal power, and heightened awareness of volcanic hazards.

The most striking volcanological advances of the 1970s, however, have been in the investigation of deep sea volcanism. The recognition that most of the world's volcanic activity is submarine, matched with technological advances in marine exploration, have led to exciting new discoveries on the sea floor. Continental segments of the world's rift systems have also benefited from increased volcanological attention. Additional recent expansion in 1970s volcanology has come with interdisciplinary approaches: the application of physics and chemistry to eruptive processes, the meteorological effects of explosive eruptions, the geophysical delineation of subsurface magma bodies, and the increasing interaction with archeological problems.

Previous U.S. National Reports to the IUGG have reviewed volcanological findings at Hawaiian, Alaskan, and western U.S. volcanoes, and provided tabular summaries of the volcanic activity at each region during the report periods through 1970. The years since 1970, however, have all been reviewed in each January issue of Geotimes, and additional chronologic detail can be found in several references cited below (under "Surveillance"). U.S. volcanoes have produced an average of 3.4 eruptions per year (σ=1.8) during the last two decades, with no remarkable variation in the 1970s. Consequently, this review will carry no chronologic summary, although outstanding volcanic events will be woven into the review. The review will be organized around the main subject areas of volcanologic research, but the accompanying bibliography contains a letter code to facilitate rapid identification of references in major geographic areas. In a general way, the review will move from effusive rift volcanism, through intraplate activity, to the explosive volcanism of island arcs and continental margins, before treating meteorological and socioeconomic aspects of volcanoes.

Subaerial lava: flood basalts, flows, forms, & lava lakes

Flood basalts must be one of the most dramatic forms of volcanism, but they have never been witnessed by man. The most recent known are those of the Columbia Plateau, northwestern U.S., most of which erupted 15 million years ago. Recent work by Swanson, Wright, and others has shown that single flows measuring 700 km^3 have spread over 10^3 km^2 in a few days. This eruption rate (1 km^3/day per km of active fissure) is 2-3 orders of magnitude greater than sustained rates observed on Hawaii. The largest historic lava eruption was from the 10 km Lakagigar fissure in Iceland, when 10 km^3 covered 370 km^2 during 50 days in 1783 (Thorarinsson, 1969). Iceland also now holds the record for the smallest, historic lava eruption known, after 1 m^3 of magma was extruded from a Krafla drill hole in 1977.

Hawaii's largest and longest historic flank eruption ended in 1974, and provided many opportunities for the study of moving lava. The full course of the 5 year eruption is reviewed by Peterson et al. (1976) and its vigorous first 17 months, including dramatic lava falls into pit craters and the construction of the small shield volcano Mauna Ulu, is described in detail by Swanson et al. (1979). The formation of lava

*Volcanology was not reviewed in the 1971-4 report. Several important references appearing during this period, if not listed in the author's subsequent work, have been included in this bibliography and the 1970's are reviewed as a whole in the text.

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