

ON THE GEOCHEMISTRY OF METAMORPHISM^{1, 2}

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INTRODUCTION

This paper is not meant to cover the whole field of metamorphism, but rather to bring out some ideas about certain processes in metamorphism. These ideas are by no means universally accepted.

Metamorphism is a chemical, mineralogical, and structural adjustment of solid rocks to new physical and chemical conditions. The rocks can be considered to form an open system in which elements may enter and leave during the metamorphism. The geochemistry of metamorphism is concerned with the changes in the chemical composition of rocks during such metamorphism. If there are no changes in the bulk chemical composition of the rock, the metamorphism is said to be isochemical. Only mineralogy and structure are then changed. This type of metamorphism involves geochemistry only on a microscopic scale.

If there is a substantial addition and removal of elements during the process, it is called *metasomatic metamorphism*. Another process which may cause considerable chemical changes in the rocks is metamorphic differentiation. Both processes, metasomatism and metamorphic differentiation, involve migration of elements and proceed usually simultaneously.

How do we know that there has been change in the bulk chemical composition during the metamorphism? The best way, of course, is to compare the chemical composition of the metamorphic rocks with that of their unmetamorphosed equivalents. However, this is not always possible and many geologists rely on some other criterion like replacement textures. These textures can be seen in the field as well as under the microscope. The literature gives numerous examples of various replacements but these descriptions are concerned with small areas, and we have only a vague idea of the chemical changes involved if larger systems are considered, or of the laws which govern these chemical changes. It is known, for instance, that changes in temperature and pressure will cause recrystallization and that the chemical changes involved obey the laws of thermodynamics. This gives certain purely theoretical suggestions concerning the conditions under which the new minerals start to grow, but it is not yet known how the constituent ions migrate, where the introduced elements come from, nor where the removed ones go.

The literature on the geochemistry of metamorphism was rather scanty until recent years. The most complete discussions were those given by Harker (1932) and Eskola (1939). Eskola classified

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metasomatic processes and discussed them at some length. The recent textbook on *Igneous and Metamorphic Rocks* by Turner and Verhoogen (1951), *Theoretical Petrology* by Barth (1952) and *Geochemistry* by Rankama and Sahama (1950) devote long sections to the subject. Ramberg's new book (1952) on the *Origin of Metamorphic and Metasomatic Rocks* discusses the subject on the basis of thermodynamics. His conclusions are based on theoretical consideration rather than on the field evidence.

It is clear that any satisfactory theory of metasomatic processes must be capable of explaining the observed field, petrographic, and chemical data. The space problem may be cited as an example. It has been pointed out by Lindgren and later by Turner and others that metasomatic processes usually take place without change in volume. Therefore, if some elements are introduced, others must be removed in order to keep the volume unchanged.

CLASSIFICATION OF METASOMATIC PROCESSES

A short review of the various types of metasomatism gives a picture of many chemical changes involved in metasomatism. Eskola (1939) distinguished the following types: (a) alkali metasomatism, (b) iron-magnesium metasomatism, (c) lime metasomatism, (d) metasomatism with introduction of Si, Sn, B, Li, F, Cl, S. (e) hydrothermal alterations, and (f) carbon dioxide metasomatism.

Examples of various types of metasomatism mainly following this classification are given in Table 1.

TABLE 1.—CLASSIFICATION OF METASOMATIC PROCESSES

	Introduced	Removed
A.	Alkali metasomatism	
	K, Na	Mg, Fe, Al, H
	1. Slate→phyllite→mica schist→gneiss→granite	
	K, Na, Si	Mg, Fe, Ca
	2. Amphibolite→biotite gneiss→augengneiss→granite	
	K or Na	Na or K
	3. Exchange of alkalis (replacement of albite by orthoclase or vice versa)	
	K, Na	
	4. Gneiss→alkali granite (fenite) near alkaline intrusions	
B.	Aluminum-iron-magnesium metasomatism	
	Mg, Fe, Ca, Al	Si
	1. Quartzite or biotite schist→amphibolite→gabbro (→diorite)	
	Na, Ca, Al	Si
	2. Quartzite or biotite schist→biotite-andesine gneiss→anorthosite	
	Mg, Fe, Al, Si	Ca
	3. Limestone→skarn→amphibolite (biotite gneiss→) granite	
	Mg, Fe	Ca, Na
	4. Biotite-gneiss (leptite)→cordierite-anthophyllite gneiss	
C.	Lime metasomatism	
	Formation of calc-silicates in noncalcareous rocks near limestones and ultrabasic rocks	
D.	Metasomatism with introduction of Si, Sn, B, Li, F, Cl, S	
	Formation of minerals containing these elements	
E.	Hydrothermal alterations	
F.	Carbon dioxide metasomatism	
	Formation of carbonates in silicate rocks.	

A. 1. Introduction of alkali feldspars into slates and shales is in many instances a fixation of alkali by an excess of alumina. Barth (1952) has calculated the number of ions in a unit cell introduced and removed from the slates in Dutchess County, New York, when they were progressively changed to schists, plagioclase gneiss, and augen gneiss. He found that the slate steadily loses hydrogen, magnesium, iron, and aluminum ions. These ions are replaced by incoming ions of silicon, calcium, and alkalis.

2. Replacement of hornblende by biotite during the granitization of amphibolites is well known, as is the replacement of sillimanite and andalusite by muscovite. Many other similar examples show that alkali metasomatism is extremely common during the metamorphism.

3. Replacement of microcline by albite and the formation of myrmekite involve an exchange of alkalis.

4. Introduction of alkalis into quartz-rich country rocks in surroundings of intrusive alkaline rocks have been described from the Fen area in Norway by Brögger (1921) and from Iron Hill, Colorado, by Larsen (1942). These contact rocks, so-called fenites, are enriched in alkali feldspars, and the ferro-magnesian minerals in them are replaced by alkali amphiboles and alkali pyroxenes.

B. 1 and 2. In some siliceous rocks quartz is replaced by femic minerals like hornblende and biotite or by calcic or medium plagioclase. The author has studied this metasomatism in an area northwest of the Idaho batholith. Quartzites and biotite-garnet schists there seem to be transformed to hornblendites, gabbros, and diorites or on the other hand to anorthosites and garnet amphibolites through a metasomatic introduction of two or more of the elements: magnesium, iron, calcium, and aluminum and subsequent removal of silicon.

3. Limestones in the same area are converted to lime-silicate rocks because of introduction of iron, magnesium, aluminum, and silicon and removal of calcium.

4. A classic example of iron-magnesium-silicate metasomatism is the Orijarvi region in Finland. Quartzo-feldspathic fine-grained biotite gneisses, so-called leptites, are transformed into cordierite-anthophyllite gneisses with complementary removal of calcium and sodium. Common hornblende in the amphibolites is converted to cummingtonite because of introduction of magnesium and removal of calcium. The limestones are converted to andradite-hedenbergite skarns.

C. As an example of lime metasomatism, one can mention the transfer of lime from a limestone across the boundary to a silicate rock, where minerals like diopside, grossularite, and scapolite will crystallize. Introduction of lime into the country rock surrounding basic igneous intrusions is common.

D. Introduction of boron, fluorine, and chlorine can be seen in the development of the minerals containing these elements.

F. Among the examples of carbon dioxide metasomatism, the alteration of serpentines and actinolite rocks to talc-carbonate rocks,

and the carbon dioxide metasomatism in connection with gold deposits of the Mother Lode area are well known (Knopf, 1929).

GRANITIZATION

The metasomatic processes leading to granitization have been discussed recently by a number of petrologists. As rocks of widely different compositions may be transformed to granites of about the same composition, the substances introduced and removed in each individual case vary within a wide range. This fact has been interpreted in different ways.

Eskola (1950) concludes that because the product of granitization of rocks of widely different chemical composition is always the same, that is granite, the agent of metasomatic granitization is an igneous granite magma or granitic rest solution of simatic rocks.

The supporters of a true metamorphic and metasomatic origin of granites have a different approach. They assume that the substances added to the granitized areas were mobilized from ultra-metamorphic rocks at elevated temperatures. There are several theories about how this mobilization took place; these are discussed below.

Many of the supporters of this metamorphic origin consider that granite is a stable endproduct of metasomatic and metamorphic processes. Thus, the tendency of all migration of elements in the upper lithosphere would be toward the formation of rocks of granitic composition. Only rocks of granitic composition would be stable in the upper lithosphere. The uniformity of the chemical composition of the granites cited as a proof of their magmatic origin by many petrologists can also be considered as a proof of their metamorphic origin.

Seismological studies show that there is a light granitic layer under the continents and under it a heavier shell probably of basaltic composition. As a granitic magma can be a differentiation product of a basaltic magma, it is usually considered that this granitic shell was formed by crystallization differentiation from the basaltic layer. The batholiths are considered to be formed by the lighter granitic magmas rising into geosynclinal areas. This poses three problems: (1) the amount of granites compared to basic igneous rocks, (2) the space problem, (3) granite batholiths occur exclusively in geosynclinal areas.

The first problem has led many geologists to believe that most granites are a result of granitization. The second problem, if studied in the field, will show that in many instances the older rock was carried away without disturbing the earlier structure when the granite came in. This cannot possibly happen unless the granite was emplaced by replacement, by a chemical rather than physical process. The third point may be answered if it is assumed that most of the material even in the igneous granites is of sedimentary origin.

What are the possibilities of a large scale granitization? The magmatists' point of view about the rising of granitic magmas into the upper lithosphere includes the fact that the granites have the

chemical and mineralogical composition which is in equilibrium in the upper lithosphere. As pointed out, there is a general tendency toward a granitic composition in regional metamorphism, and as soon as this composition is reached, the metasomatic rocks recrystallize as granular rocks with a granitic texture. The intrusive granites and the regionally metamorphosed rocks are usually side by side and, therefore, the temperature-pressure conditions are about the same in both cases. Because the rocks form an open system the substances start to migrate as soon as there are thermodynamic gradients, such as differences in concentration. Migration continues until equilibrium is re-established, that is, until the system is homogeneous. Thus, it is only natural that in the upper lithosphere, where granitic composition is predominant and in equilibrium, the tendency of all chemical processes would be toward a granitic composition.

Reynolds (1946) has suggested that granitization would always be accompanied by the introduction of elements into the surrounding rocks. She gave a name, "basic front," for these rocks which are usually enriched in Mg, Fe, Al and Ca and one or two of the minor elements like P or Ti. Barth (1952, p. 362) remarks that during metasomatic granitization, the rejected elements, that is, Fe, Mg, Al and Ca should migrate back to the place from which granitizing emanations came in order to keep the volumes unchanged. He admits, however, that it is possible that water vapor, which always travels away from the magma, would act as a "carrier" and bring along a part of these heavy cations which then would be precipitated into the surrounding area.

METAMORPHIC DIFFERENTIATION

The term *metamorphic differentiation* is used for processes which lead to segregation of certain elements from an originally homogeneous rock. Examples would be the formation of porphyroblasts and clusters, secretion pegmatites along cracks, accumulation of quartz or quartz and feldspars into blebs and veinlets (banded gneisses), and enrichment of the rock in certain minerals. The term was first introduced by Stillwell (1918) later discussed independently by Eskola (1932). Eskola's idea was that the material for the porphyroblasts is derived from the surrounding area in the rock by short distance diffusion. The quartzo-feldspathic veinlets were thought to be formed by a partial resolution and redeposition of the easiest soluble substances. The enrichment of a rock in certain minerals was explained as follows: If the solubility of a newly formed mineral in pore liquid is exceptionally slight, it will crystallize out and the pore liquid will be impoverished in the compounds of this mineral. Solutions will therefore bring more of these constituents from the adjacent layers, especially from those in which this particular mineral cannot form because of its incompatibility with the other minerals. Turner and Verhoogen (1951, p. 498) doubt this mechanism and suggest that all metamorphic differentiation would be a result of differential migration of components through the

metamorphic system under the influence of local gradients in chemical potential. Ramberg (1952) has expressed the same general idea. He emphasizes the importance of diffusion during the metamorphic processes.

MODE OF MIGRATION

The mode of migration of matter during metamorphism is much debated. The three following possibilities are usually cited:

- 1) The elements are transported by liquids and gases streaming through the rocks.
- 2) The ions are transferred by diffusion in solutions filling the pores and subcapillary openings in the rocks.
- 3) The ions move by diffusion in solid, that is, through crystal lattices.

Earlier it was thought that the chemical reactions take place only in the presence of liquids. Metasomatism was considered to be effected by reactions between the constituent minerals and chemically active liquid or gaseous solutions streaming through the pores of the rocks. This concept holds true if there are physical gradients which will press the liquids through the pores and if the permeability of the rock is great enough. It is fairly certain that during the orogenic periods and during the emplacement of igneous rocks such gradients will exist. Liquids and gases from crystallizing magmas or from recrystallizing sediments escape along the fractures, shear planes, and other planes offering least resistance. This mode of migration is perhaps most important because large amounts of new elements may be introduced over great distances within relatively short periods. However, the examples of metasomatism which are later than the regional metamorphism and deformation show that there must also be other ways of transfer of matter in the rocks.

Assume that there are no mechanical forces which would press the solutions through the rock and that the openings and pores are too small for any liquid to move through them. Pore liquids would then be stationary or moving only slowly. Assume further that the concentration of ions, or pressure, or temperature varies locally in this rock.

It can be shown, using simple mathematics and laws of thermodynamics, that the ions migrate from a place of high chemical potential (or high activity) to a place of low chemical potential (or low activity) (cf. Turner and Verhoogen, 1951), for instance from high to low pressure or from high to low concentration. According to Ramberg (1952) the mechanism of transfer of matter can take place as follows: Minerals go into a disperse phase (disperse phase may be a gaseous or liquid solution or a disperse phase of ions) in the places of high chemical activity (which corresponds to high chemical potential of Turner and Verhoogen). Dispersed atoms migrate towards the place of low chemical activity (or potential) and enter new crystal structures. This migration will cause a lowering of the concentration of the ions in the dispersed phase at the point of high

concentration. Equilibrium between a solid phase and a dispersed phase is disturbed. In order to re-establish the equilibrium new ions from the solid structures will disperse. At the point of low concentration (and low chemical potential) the equilibrium between a solid and dispersed phase is also disturbed because new ions were added into the dispersed phase. The equilibrium is re-established when ions enter the solid structures. Thus, the point of low concentration and low chemical activity or low chemical potential is a place of deposition.

This holds true for all reactions. As the partial vapor pressure of any component of a liquid or solid solution is defined as the partial vapor pressure of this component in the saturated vapor in equilibrium with this phase, the general conclusion holds as well for diffusion through gases, or liquids, or solids. Every chemical species in a rock tries to reach a place where its chemical potential or its molal free energy is lowest. Activity gradients are created by differences in 1) pressure, 2) temperature, 3) chemical composition of minerals, 4) size of minerals, 5) surrounding milieu. The last two are based on the surface energy and the free molal energy of the minerals. Thus, in the rocks where gradients in physical conditions (difference in pressure and temperature and concentration) cause gradients in chemical potential, the substances start to migrate in order to re-establish the equilibrium. The migration may take place in a gas or liquid filling the open channels, pores, and intergranular film or through the solid crystals. It remains to be decided in what way most of the matter is transferred.

In permeable rocks, one expects that the migration takes place through the liquids or gases streaming through the pores. In the deeper levels of the lithosphere where recrystallization and pressure have decreased the porosity and permeability, the diffusion along the grain boundaries (intergranular film) becomes probably more important, and in still deeper levels, diffusion through solid crystals may play a more important role than hitherto assumed.

On the other hand, the mode of migration must change during deformation and metamorphism. The permeability can be fairly high in the unmetamorphosed sediments but gets smaller as recrystallization advances because the open channels are filled by newly crystallized minerals. Thus, the velocity of the solutions and gases that stream through the pores of the unmetamorphosed sediments will be slowed down during the recrystallization until movement ceases. Assume that these solutions and gases carried chemically active elements that reacted with the earlier minerals, or that they carried compounds of minerals which were deposited in the rock. During this process, a certain amount of substances will be transferred through an area of 1 sq. cm. in a second and of course the same amount of old substances is extracted. This amount can be called a velocity of transfer of matter by moving solutions. On the other hand, as soon as there is a difference in a concentration of molecules, atoms, or ions, as there is between the rock and the loci

of imported elements, diffusion starts to operate. The ions from the imported liquid or gas start to migrate to the country rock along the cracks, crystal boundaries, and through the crystals. When the velocity of the liquid has become slow enough, it is possible that at certain moments, the amount of matter transferred by the movement of liquids or gases has become smaller than the amount transferred by diffusion. We can go even further; the advancing recrystallization of the rock will tend to decrease the porosity and a phase may be reached when the amount of ions transferred through the crystal lattices is larger than that through the liquids or gases filling the pores and other subcapillary openings.

Thus, the problem seems to be to determine the role of each of these three mechanisms at each moment during the advancing metamorphism. Under favorable circumstances this can be studied in the field. In an area northwest of the Idaho batholith one can distinguish several periods of metasomatic introduction of elements. During an early phase of metasomatism, the schists and quartzites were converted to quartz diorites or tonalites. In contrast to this, the resultant rocks during a later period of metasomatism were anorthosites and hornblende-garnet rocks or hornblende-biotite rocks occurring as separate lenticles. Approximately the same elements were introduced during the earlier and later phase, but during the later period the calcic plagioclase on the one hand and the ferromagnesian minerals on the other hand segregated in separate lenticles. Thus, there was a differentiation of the salic and femic constituents. There is no trace of such differentiation during the early period. A possible explanation is that the solutions which carried the introduced substances were forced bodily into the country rock during the early phase. Later when the mechanical forces ceased, the elements in the pore liquids migrated by diffusion. Probably the difference in the velocity of migration of constituent ions made the segregation of plagioclase from hornblende and biotite possible. Using a criterion developed from the study of textures in the field and under the microscope a sequence of introduction can be established. The biotite and hornblende are earlier than the calcic plagioclase. This may suggest that the velocity of Fe and Mg was greater than that of Ca. Part of the Al went with Fe and Mg to form hornblende and another part followed Ca to form plagioclase.

This is only partly in accordance with the concept that small ions are capable of migrating faster than the large ions. Ramberg has suggested that the type of bonding is the most important factor in determining the mobility of ions. He considers that velocity is unimportant and that the bonding energy is the deciding factor. Thus, for instance, K and Na which are loosely tied to feldspars are found to be highly mobile because they are easily detached from their structures. Perhaps both factors operate.

In conclusion, there are many problems to be solved before we can advance on a stronger foundation. We might solve some of the problems experimentally by determining the diffusion coefficients for

various elements in various silicate rocks, either dry or saturated with water or water vapor, and compare it with the porosity measured in the same rock or with the grain size, with the amount of cracks visible in the rock and in the minerals under the microscope, with the texture and orientation of minerals (fabric) in the rock. It is commonly believed that such mechanical agents as shearing stress and penetrative differential movements promote the transfer of matter and reactions during recrystallization. Probably ruptures which are formed during the deformation serve as avenues for migrating ions. It is also possible that during the deformation rock minerals attain such a preferred orientation that ions migrate in one direction faster than others.

Experimental data about the diffusion of elements through the silicates are very scarce. The work hitherto done suggests that diffusion through solids is very low compared to that through liquids and gases. Therefore, if the rock has a considerable porosity or contains cracks filled by aqueous solution, the number of ions migrating through the liquid or along the intergranular would be much higher than that through the solid. In solid rock, which has been recrystallized in an earlier stage of metamorphism, the porosity is reduced considerably and solid diffusion may become relatively more important.

Diffusion through solid crystal lattices is thought to take place in the following three ways (Barrer, 1941):

- 1) The clusters of pairs of atoms in close packed crystals are assumed to be capable of rotation.
- 2) It has been shown that crystal lattices are rarely ideal but that there are usually vacant positions (Jost, 1937). Small numbers of atoms in the crystal are assumed to be able to jump from one empty position to the other.
- 3) In a number of silicates there are openings and channels through which ions can migrate. The larger ions probably would tend to travel through the cracks, through mosaic fissures in the crystals, and along the crystal boundaries while the small ions could go also through the crystal lattices.

SOURCE OF THE INTRODUCED ELEMENTS

As has been mentioned earlier, the source of the elements which are introduced during the metasomatism is thought to be either an igneous magma or ultrametamorphic rocks.

In a short survey, for instance, of skarns which are common reaction products between limestones and silicate bearing solutions, igneous intrusions usually supply the surrounding rocks with heat, water, and other volatile compounds, which may carry appreciable amounts of heavy metals, probably as chlorides, fluorides, or sulfides. The primary magmatic gases are acid and react more easily with the limestones than with the silicate rocks. The limestones, therefore, serve as filters for the magmatic gases and are usually converted to skarns near intrusive rocks. Among the great variety of reaction

minerals in these skarns may be some which contain chlorine, fluorine, or sulfur indicating that these volatiles were present in the magmatic gases.

Many skarns, however, do not show any indications of these volatiles. This type of skarn is common in the regionally metamorphosed areas and it is possible that the elements which were introduced into the limestone and changed it to a lime-silicate rock were not from a magmatic source but that the silicon, iron, and other elements migrated from the adjacent silicate rock layers during high temperature metamorphism. The matter would be simple if presence or lack of the volatiles which are found to be common in the magmatic gases could be considered as an indication of the magmatic or nonmagmatic origin of metasomatizing gases or solutions. Unfortunately, it is also true that the volatiles may have been carried further and left no traces in the rocks through which they passed.

Concentration of minor elements in the surroundings of igneous bodies may serve as an indication of an igneous origin of the introduced elements.

In conclusion it should be pointed out that there is still a great deal to learn before the chemical processes involved in regional metamorphism can be understood. Large systems should be studied in order to find out what elements are introduced into and what removed from the system, and, on the other hand, what changes resulted from the rearrangement of elements within the system. It is natural to think that all metasomatic phenomena are parts of one global process and that the tendency of these chemical processes is toward equilibrium. It has been suggested that granitic composition would represent this state of equilibrium in the upper lithosphere.

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NEWS OF TENNESSEE SCIENCE

(Continued from page 285)

Atomic Energy Commission Oak Ridge Operations Office, has resigned to accept a position with private industry. Herman Roth, Assistant Director of the Division, has been appointed to the directorship.

The University of Tennessee is establishing an advanced degree program leading to the master's degree in engineering science. The program will emphasize research along broad scientific lines and will concern principles applicable in various fields of engineering and physical science research.

A new course in radioactivity for secondary school science teaching personnel will be offered by the College of Education at the University of Tennessee, Knoxville, for the period July 18-July 29, 1955. The course is intended to meet the special needs of the following groups: (1) those who are actively engaged in teaching science in secondary schools, (2) supervisors whose duties include those of working with secondary school science teachers, (3) representatives of departments of education of colleges or other workers in science education at college level whose interest is in college science courses. The course will include lectures, laboratory work, discussions, films, and field work related to the problem of presenting nuclear science in the secondary school program.

The Seventh Annual Biology Research Conference, sponsored by the Biology Division of Oak Ridge National Laboratory and supported by the Atomic Energy Commission, met in Oak Ridge April 19-21. The subject for discussion was "Genetic Recombinations."

Dr. Daniel Billen of the Biology Division, Oak Ridge National Laboratory, recently spoke before the Medical and Psychological Section of the Georgia Academy of Science, Dahlonega, on "Radiation Protection" and also gave a lecture before the Department of Botany, University of Georgia, on "The Effects of Ionizing Radiations on the Physiology and Cytology of Bacterial Cells."

Dr. Drew Schwartz of the Biology Division, Oak Ridge National Laboratory, recently gave a seminar on "Cytogenetic Studies on X-Ray-Induced Ring Chromosome Breakage" at the University of North Carolina, another on "The Effect of Oxygen Concentration on X-Ray-Induced Chromosome Breakage" at the University of Maryland, and another on "Studies on the Mechanism of Crossing Over" at the University of Pennsylvania.

Three members of the Biology Division, Oak Ridge National Laboratory, gave lectures during the summer, 1954, at the Duke University Marine Laboratory, Beaufort, North Carolina. Dr. C. W. Sheppard lectured in Radiation Physics, Dr. John R. Totter in Radiation Biochemistry, and Dr. Norman Anderson in Radiation Physiology.

RECENT PUBLICATIONS BY TENNESSEE AUTHORS

- Morgan, Karl Z. (ORNL). 1954. Maximum permissible concentration of radioisotopes in food, water and air and maximum permissible equilibrium amounts in the body. *Acta Radiologica*, 41:30.
- Moshman, Jack (ORNL). 1953. Critical values of the log-normal distribution. *Jour. Am. Statis. Assoc.*, 48:600.

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