HYDROTHERMAL ACTIVITY AND HYDRO-EXPLOSIONS AS A CAUSE OF NATURAL COMBUSTION AND PYROLYSIS OF BITUMINOUS ROCKS: THE CASE OF PLIOCENE METAMORPHISM IN ISRAEL (HATRURIM FORMATION)

Arie Gilat

INTRODUCTION

This paper is an attempt to show that high-temperature fluid activity gave rise to Hatrurim metamorphism.

Metamorphic rocks of the Hatrurim Formation (Gvirtzman and Buchbinder, 1966) are well developed in central Israel (Fig. 1), almost always found in the specific stratigraphic position of the Ghareb - Taqiye formations of Campanian - Maastrichtian age (Fig. 2). They comprise a lower unit, 10 to 85 m thick, built of completely recrystallized low grade multi-colored metamorphic chalks and marls, and of three upper mapping units, characteristic of middle to high grade metamorphism, corresponding to pyroxene hornfels and sanidinite facies (calcite-spurrite- gehlenite- lizardite-, and larnite-bearing, Gross, 1977; Burg, 1990). They formed by processes involving heating, decarbonation and dehydration. The overall section contains secondary gypsum, aragonite and volkonskoite veins, iron oxides, quartz and uranium mineralization.

The generally accepted hypothesis for their genesis was proposed by Bentor et al. (1963) and may be summarized as follows: internal combustion of bitumens caused high-temperature, low-pressure metamorphism. The burning was intensive, and occurred close to the surface (where oxygen was available) in many local foci and continued for relatively short durations (Spiro, 1979; Matthews and Kolodny, 1978). The heating was then followed by retrograde processes in the metamorphosed rocks (hydration, carbonation and sulfatization). The progressive hydration caused development of lizardite-zeolitic rocks rich in hydrogarnets and hydrated calc-silicate rocks (Gross, 1977).

Kolodny and Gross (1974), Bentor et al. (1972) and Gross (unpubl.) have shown experimentally that metamorphism of the Ghareb and Taqiye formations is basically an isochemical process. They also postulated that the process took place in synclines (higher concentration of organic material) and that tectonic activity related to the Dead Sea Transform caused fractures along which oxygen was supplied. The mechanism responsible for ignition in many local foci is unknown.

Burg (1990) mapped the largest (~50 km^2) outcrop of the metamorphosed rocks in the Biq'at Hatrurim area, based on correlation between metamorphic rocks and protolith (Fig. 2); his K-Ar dating (with A. Heimann) of the metamorphic event yielded a Pliocene age (~3 Ma). Burg (1990) has also shown that in numerous cases the burning was clearly focused along cracks, which served as conduit channels; in many localities it occurred 10 to 200 m above the top of bituminous rocks (Fig. 3).

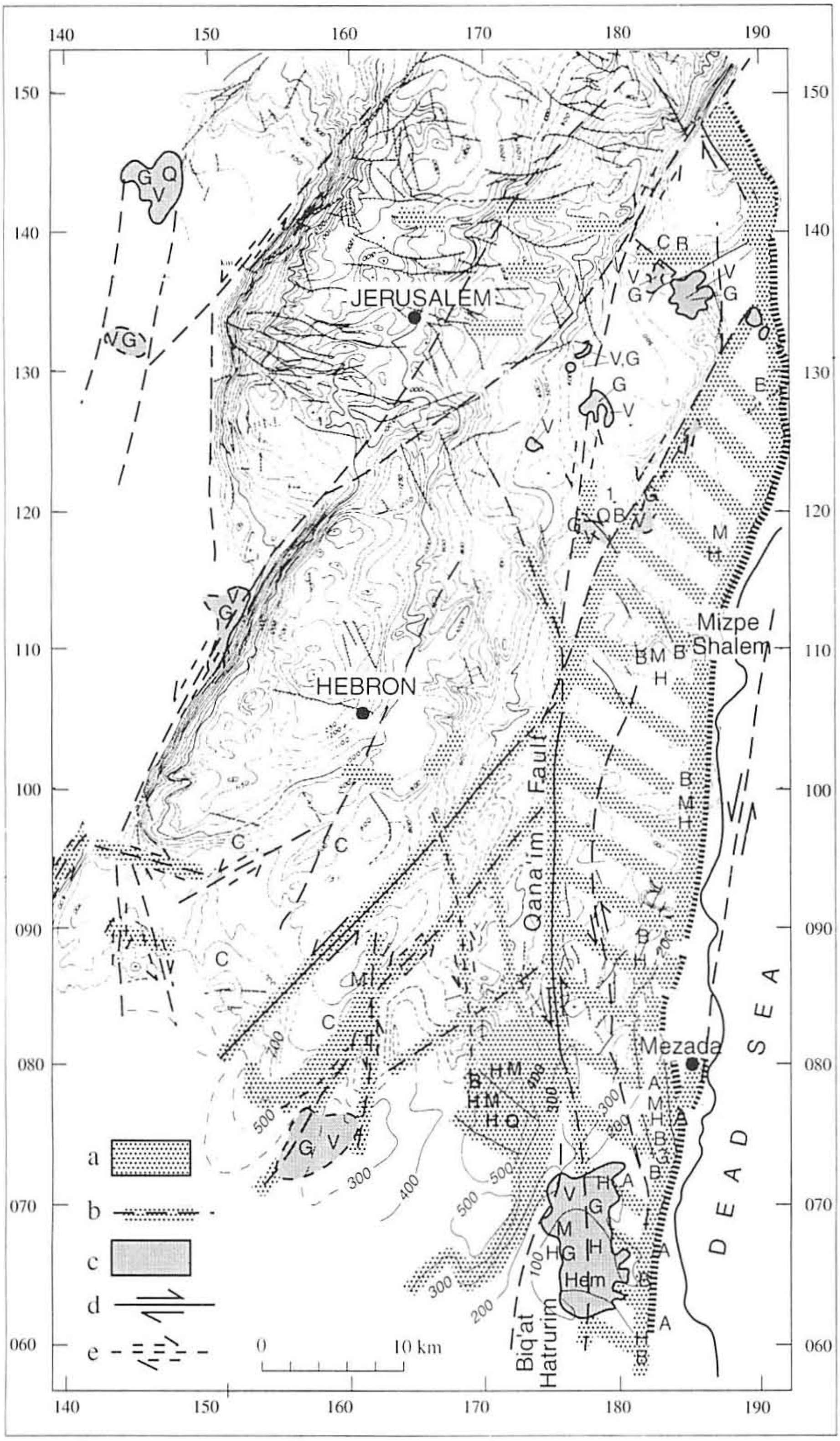
Among the other high-temperature events which took place in the Pliocene in the Judean Desert area, Raz (1983) and Magaritz et al. (1983) studied stable isotopes in epigenetic dolomites in the southern Judean Desert and came to the conclusion that the large scale dolomitization phenomena were caused by solutions with temperatures between 55 and 120°C. Gilat and Lang (1985) and Gilat et al. (1986) conducted fluid inclusion studies in post-dolomitizational hydrothermal barite mineralizations from the same area.

Gilat (1992) studied tectonics and epigenetic mineralization of the Judea region, in the course of which large scale hydrothermal-metasomatic processes related to strike-slip faults were mapped and investigated in the region bordering Biq'at Hatrurim. The relevant information is summarized below.

PLIOCENE-PLEISTOCENE HYDROTHERMAL-METASOMATIC ALTERATION AND MINERALIZATION IN THE VICINITY OF THE HATRURIM BASIN

Following are some of the characteristic features for hydrothermal-metasomatic alteration and mineralization in the vicinity of the Hatrurim basin:

1. All the alterations (and the Hatrurim-type metamorphism) developed in a specific structural position in the vicinity of shear zones or flexures (Fig. 1).
2. Large-scale dolomitization phenomena (Figs. 1-3) formed from solutions at temperatures between 55 and 120°C (Raz, 1983; Magaritz et al., 1983).
3. Calcite and barite veins are common (Figs. 1-3). Fluid inclusions in the barite indicate homogenization temperatures of 165-210°C (up to 400°C at Nahal



Key horizon top Bi'na Fm., contours at 100 m interval.

a area of the most intensive strike-slip faulting, block-rotation, circular dolomite bodies and massive dolomitization along fault-shear zones.

b massive dolomitization along major faults and flexures.

c low to high temperature - low pressure Pliocene metamorphism (Hatrurim Fm.).

d major strike-slip and oblique faults.

e major strike-slip and oblique faults, inferred.

Plio-Pleistocene hydrothermal mineralizations:

B - barite;
M - metallic;
H - halite veins;
A - asphalt penetrations;
V - volkonskoite;
Hem - hematite;
C - calcite
G - gypsum
(only the large mineralizations are shown).

Note: not all of the map area has been geochemically studied.

Figure 1. Structural map of central - eastern Israel showing areas affected by Late Miocene-Pleistocene hydrothermal-metasomatic and metamorphic processes (after Aharoni, 1976; Burg, 1990; Flexer et al., 1989; and Gilat, 1992).

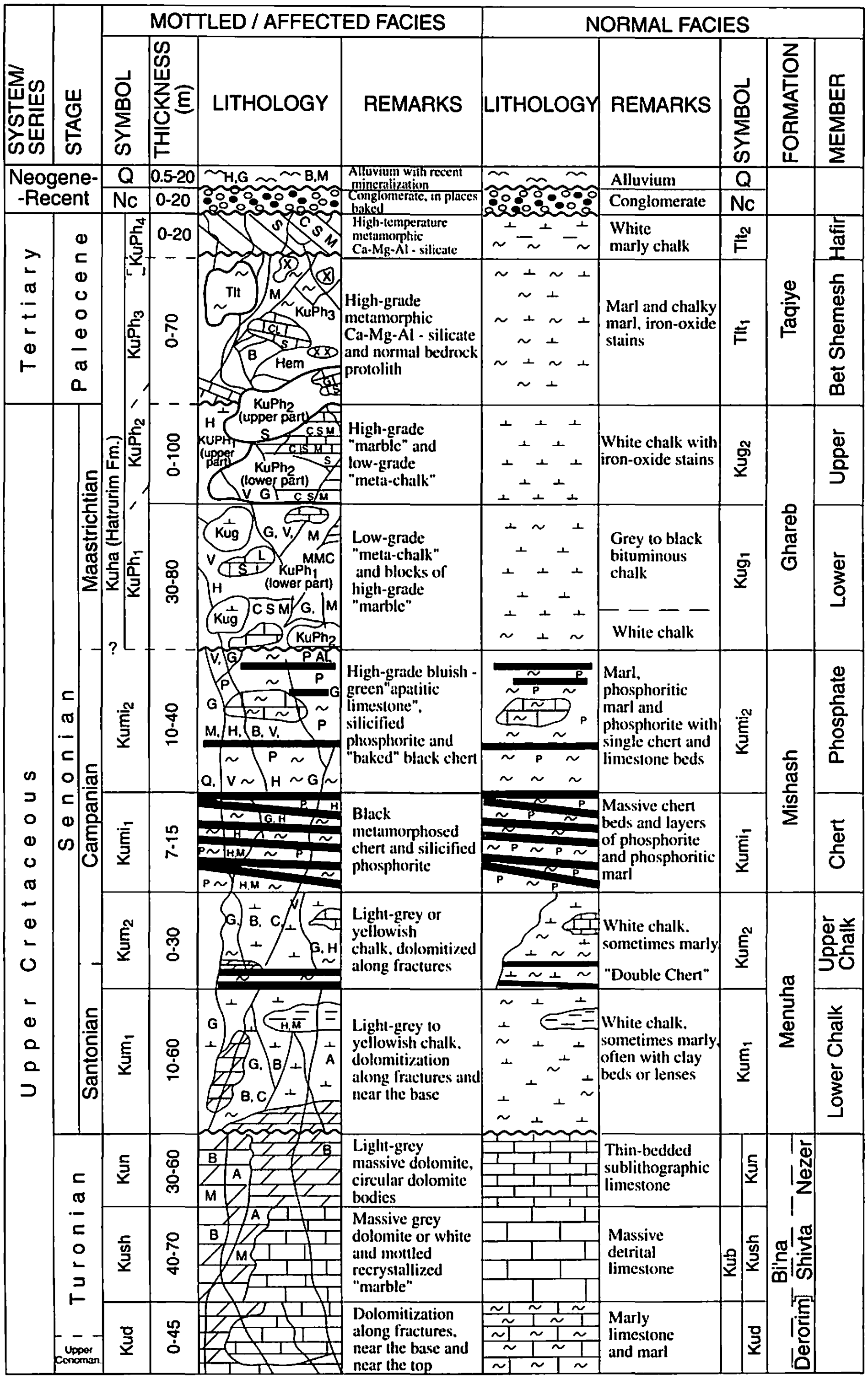


Figure 2. Plio-Pleistocene epigenetic mineralizations and metamorphic development in the Bi'na (Derorim - Shivta - Nezer), Menuha, Mishash, Ghareb and Taqiye formations (after Burg, 1990, and Gilat, 1992). Not to scale. Rocks (high temperature metamorphic facies): L - larnite; S - spurrite; CSM - calcite-spurrite marble; CL - calcite-lizardite rock; X - gehlenite; AL - apatitic limestone; MMC - mottled metamorphic chalk (low temperature facies). Plio-Pleistocene epigenetic mineralizations: V - volkonskoite; B - barite; G - gypsum; C - calcite; H - halite; Hem - hematite; A - asphalt; M - metallic mineralization.

Boqeq, Gilat and Lang, 1985), and 120-300°C at Nahal Hever (Gilat et al., 1986).

4. Hydrothermal mineralization with associated halite, gypsum and iron oxides, is characterized also by notable enrichments in U-Th, REE, trace elements and even noble metals, developed during the Late Pliocene-Pleistocene as an ongoing process related to periodic pulses of tectonic activity (Gilat, 1992). Eu, and locally Ce, are enriched relative to other REE. Enrichment in Cr is also typical for hydrothermal processes in the Judean Desert. The mean enrichment of the country rock in Cr in the vicinity of shear zones in Judea is about 400%, and its content in Fe-oxide, gypsum and halite-related mineralizations outside the Hatrurim Formation occurrences is much above average, reaching 0.2% (Gilat, 1992). Volkonskoite (contains 4.5% Cr, formed by hydrothermal alteration, Gross, 1977), has been found in the chalk of the Menuha Formation in the northern Judean Desert (Figs. 1-3). At each site of hydrothermal activity, the mineralizing agent was acid, sulfate-rich, sodium-chloride brine, probably driven by tectonic activity and compactive expulsion from the developing Dead Sea basin. As was shown by factor analyses applied to geochemical data from the Arad-West Hatrurim area (172 samples) and from the Mezada area (378 samples), the mineralizing brines carried mobilized elements from at least five different sources (Gilat, 1992).

5. Sub-horizontal fractures (Figs. 4, 5) are locally abundant in the lower part of the Hatrurim Formation (and are near-absent in the protoliths). These are slightly arched upwards, a few centimeters to several meters long. In places these are open fractures and are hollow or partly filled by gypsum and halite crystals. Metamorphosed country rock in the vicinity of these fractures is composed of loose angular fragments; its brecciated structure was not produced by tectonic movements.

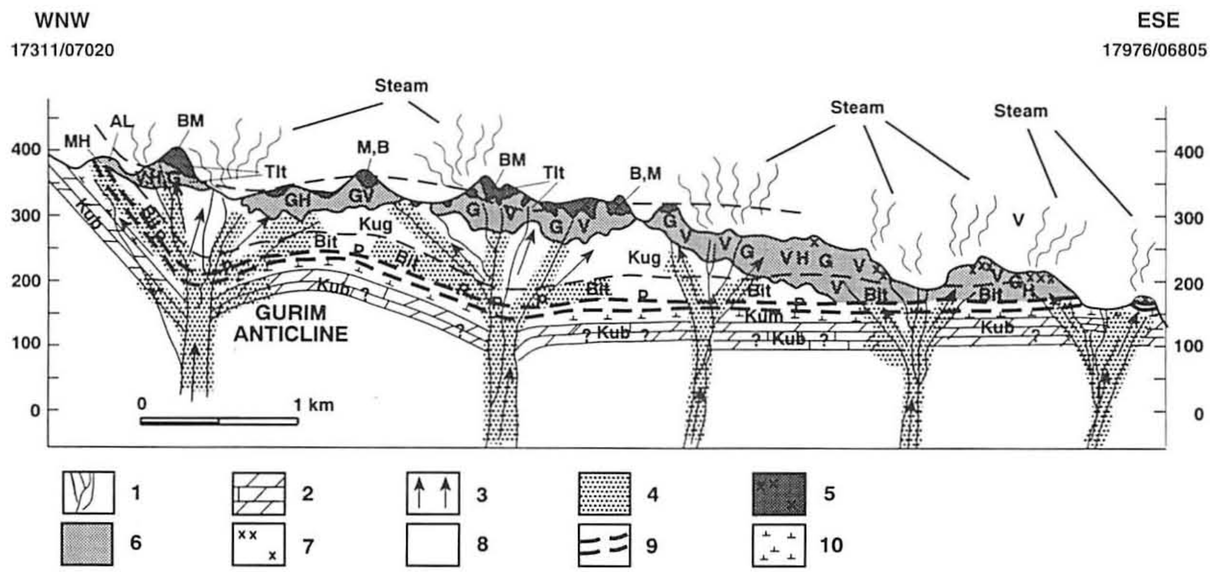


Figure 3. Cross section across the Hatrurim basin, showing field relations between the metamorphosed rocks of the Hatrurim Fm. and the non metamorphic Taqiye (Tlt) and Ghareb (Kug) formation rocks, separated by dotted line. The contact line between the metamorphic and the non metamorphic rocks, and the broken line which marks the top of bituminous section are schematic (from Burg, 1990). Some of the Plio-Pleistocene secondary processes are schematically shown on the cross-section: 1 - strike slip shearing; 2 - dolomitization; 3 - tectonic injection of heated and over-heated (up to 160°C) liquids enriched by hydrocarbons and/or H_2S; 4 - zone of hydrofracturing, hydro-explosions, underground spontaneous combustion and refining of organic matter in bitumens with ionized steam as an oxidizer and detergent; 5 - the near surface area of spontaneous combustion of expelled heated products of refining, which caused the high temperature metamorphic facies $KuPh_2$ - $KuPh_4$; 6 - the low temperature metamorphic facies ($KuPh_1$); 7 - gehlenite - larnite rock bodies, produced by crack-focused burning; 8 - nonmetamorphosed rocks; 9 - chert layers and phosphorite (P), belonging to the Mishash Fm;10 - chalk of the Menuha Fm; Bit - bituminous; B, V, AL, G, H and M - see legend to Fig. 2.

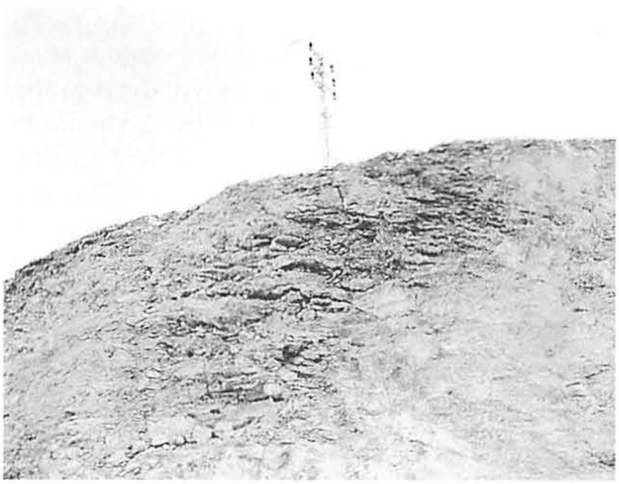


Figure 4. Unusual nonsystematic fracturization and brecciation presumably caused by hydro-explosions in mottled chalk of the lower part of the Hatrurim Fm. sequence ($KuPh_1$); road cut along the Jerusalem-Jericho road near coord. 1843/1358.

Figure 5. One of many large horizontal fractures, presumably caused by hydroexplosions, partly infilled by gypsum druses and halite mineralization exposed in the road cut ~200 m. NW of the Arad-Dimona-Zohar intersection (coord. 1736/0700). Unit $KuPh_2$ of the Hatrurim Fm., brecciated calcite-spurrite rock sequence.

DISCUSSION

The combustion metamorphism theory for the origin of the Hatrurim Formation does not explain the following:

(a) What mechanism is responsible for pre-fire rock heating and eventual ignition in thousands of local foci?

(b) What type of fuel was burnt in the oxidation zone, sometimes at a distance of more than 100 m above the nearest bitumen?

(c) What caused recrystallization of the rock-sequence and the secondary mineralization? The present theory postulates that during retrograde metamorphism, cold waters somehow enter a hot (>100°C) rock-sequence. Under atmospheric pressure, this is unlikely since it would vaporize immediately.

Certain findings made during the last twenty years allow a "fresh look" at some characteristics of the Pliocene metamorphism.

(1) The Hatrurim Formation (Fig 2), has a typically hydrothermal appearance, both its lower recrystallized part and its upper high-grade veined rocks (the veins are related to liquids by definition); it developed in the vicinity of shear-zones and fundamental faults. An apparent difference in the Y-REE distribution patterns between high grade metamorphosed rocks and their non-altered protoliths, with preferential enrichment in Eu, is typical for hydrothermal alteration in the Judean Desert (Gilat, 1992). This feature can form only under conditions of high liquid-to-rock ratios (Cullers et al., 1975; Menzies et al., 1979; Michard et al., 1983). The Hatrurim Formation sequence, like the other hydrothermally altered country rocks in the area, is commonly enriched in Cr (volkonskoite is very common), iron oxides, uranium, halite, quartz and gypsum mineralizations.

(2) As shown by the detailed mapping of the Hatrurim basin (Burg, 1990), the combustion progressed along fissures and joints, often at a distance from the bitumens (up to 200 m vertically, Fig. 3). All samples from protoliths from far beneath or within the metamorphosed sequence show organic material contents of less than 2.5% (Burg, 1990). As shown by Spiro and Aizenshtat (1983), combustion affects an area of only a few meters from the source. In places, remnants of bitumen-rich rocks are present, surrounded by envelopes showing strong thermal effects. The few millimeters-wide margin between the highly affected zone (hot area), to bituminous rock (relatively cold area), is enriched in natural pyroproducts. It has a low content of hydrocarbons volatilized at temperatures lower than 250°C. The process probably is also related to the deposition of gypsum and may be a type of steam distillation under anoxic conditions (Spiro and Aizenshtat, 1983).

(3) As was shown above, Hatrurim-type metamorphism was not the only thermal event in the Pliocene. Large-scale dolomitization bordering the Dead Sea area formed in more than 100 km^3 of rocks, from a depth of more than 2 km (Buchbinder and Goldberg, 1980) and up to the surface, with temperatures of up to 120°C. This was followed by calcitization-baritization processes, with the temperatures up to 150°C and higher. The latter processes relate to the most dynamic period along the Dead Sea Transform, when not only overheated solutions, but also different hydrocarbons, produced during rapid thermal maturation of organic matter in the buried Dead Sea basin bitumen, reached the surface even on the Mezada horst. The apparent signature of these secondary hydrocarbons was found by Spiro and Aizenshtat (1983) in bitumen from the periphery of the Hatrurim and Nebi Musa metamorphic complexes (absent in the Ef'e basin). It is expressed by an addition of hydrocarbons volatilized at temperatures lower than 250°C (Spiro and Aizenshtat, 1983; fig. 1, the S_1 peak).

(4) The sub-horizontal open fractures and brecciated structure, which are unrelated to tectonics and are characteristic to the Hatrurim Formation sequence, are generally produced by hydro-explosions (S. Dzulinsky, 1991, pers. comm.), occurring when a fluid heated to >100°C rises to a spot of stress-release, becomes steam, with a corresponding volume increase of up to 1700 times.

(5) Another factor should be taken into account: the spontaneous combustion temperature for any inflammable compound is constant only under atmospheric pressure. In natural conditions, its P-T plot has a sigmoid form (Fig. 6); thus spontaneous combustion of intruded volatiles is very possible in normal sub-surface temperature conditions, under a wide-range of (hydro-) explosional pressure changes, from very high to vacuum.

CONCLUSIONS

To explain all the pecularities of the Pliocene metamorphism in Israel, the following working hypothesis is proposed: the Hatrurim type metamorphism is a hydrothermal metamorphism, a complex process evoked by tectonically introduced overheated fluids, rich in volatile hydrocarbons. These fluids reached the pressure-release point, converted to steam, exploded and caused hydrofracturing ("hydro-explosions"). In addition, ionized steam acted as an oxidizer and detergent, causing combustion of volatiles, refining of organic matter in bitumens and expelling its heated products through cracks and fissures to the zone of oxidation, where the next stage of spontaneous combustion took place. The rock sequence through which the expelled hydrocarbons moved, could also have served as a chromatographic column. Exothermic reactions (alumo-silicification, etc.),

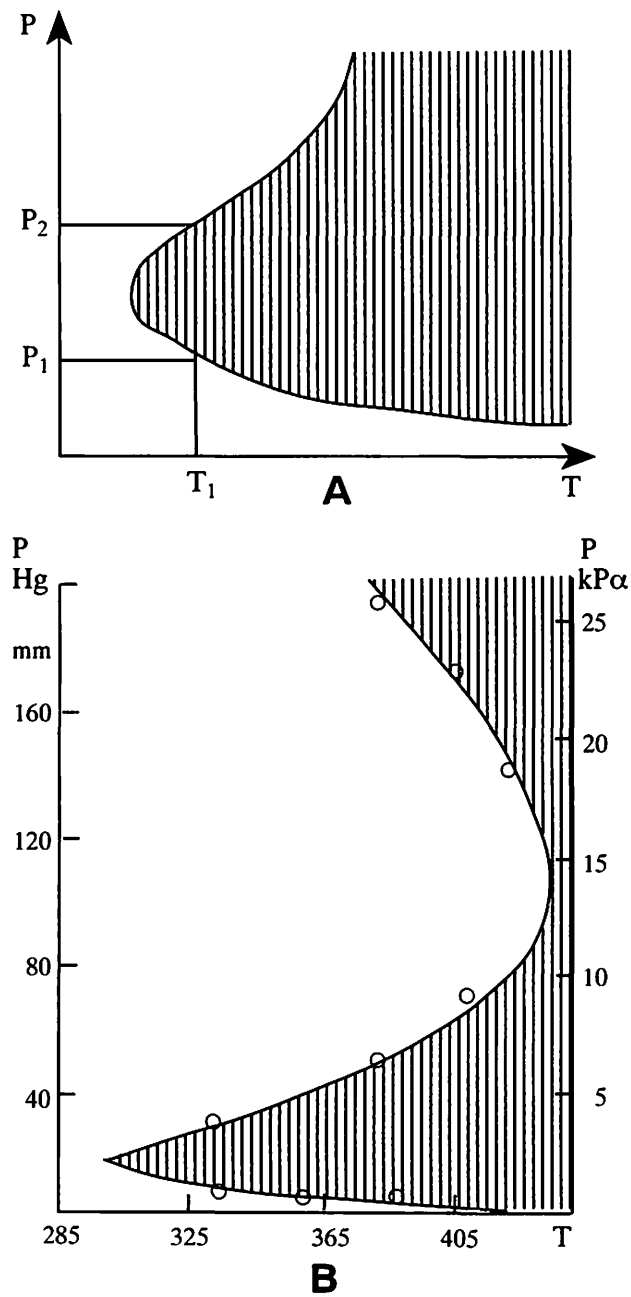


Figure 6. Limits of spontaneous combustion under different pressure/temperature conditions: A – theoretical; B – for hydrogen sulfide (H_2S) (from Panchenkov and Lebedev, 1974).

element redistribution, recrystallization of the rock sequence and enrichment in hydrothermal mineralization were probably occurring during all stages of metamorphism, the processes moving from one site to another. In very many large and small areas in Israel, with a stratigraphy similar to that of Biq'at Hatrurim and even a similar structural setting, no metamorphic process developed, probably because there was no intrusion of overheated fluids, rich in volatile hydrocarbons.

ACKNOWLEDGEMENTS

I would like to thank Dr. A. Vol (AMS senior physicist), Dr. Y. Nathan and Prof. Z. Aizenshtat for most fruitful discussions.

REFERENCES

Aharoni, E., 1976. The surface and subsurface structure of southern Judean Desert, Israel. Unpubl. Rep., 100 p. (in Hebrew, English abst.).

Bentor, Y.K., Gross, S. and Heller, L., 1963. Some unusual minerals from the "Mottled Zone" complex, Israel. Am. Miner., 48: 924-930.

Bentor, Y.K., Gross, S. and Kolodny, Y., 1972. New evidence on the origin of the high temperature mineral assemblage of the "Mottled Zone" (Israel). 24th. Int. Geol. Cong., Section 2, pp. 265-275.

Buchbinder, L.G. and Goldberg, M., 1980. Petrography of core 47, Massada 1. Isr. Geol. Surv. Rep. OD/3/80 and Oil Explor.(Investments) Rep. 81/2, 5 p.

Burg, A., 1990. The geology of the Hatrurim Formation. M.Sc. thesis, Hebrew Univ., Jerusalem, 118 p. (in Hebrew).

Cullers, R.L., Chaudhury, S., Arnold, B., Lee, M. and Wolf, C.W., 1975. Rare Earth distribution in clay minerals and in clay-sized fraction of the Lower Permian Havenswille and Eskridge shales of Kansas and Oklahoma. Geochem. Cosmochem. Acta, 39: 1691-1703.

Flexer, A., Gilat, A., Hirsch, F., Honigstein, A., Rosenfeld, A. and Rueffer, T., 1989. Cretaceous evolution of the Judean Mountains as indicated by ostracods. Terra Nova, 1: 349-358.

Gilat, A., 1992. Tectonics and associated mineralization activity, southern Judea, Israel. Ph.D. thesis, Ben Gurion Univ. of the Negev, 279 p. (in Hebrew , English abst.).

Gilat, A. and Lang, B., 1985. Fluid inclusions in barite megacrystals from Nahal Boqeq (southern Judean Desert). Isr. Geol. Soc., Annu. Meet., p. 5.

Gilat, A., Milsztajn, N. and Lang, B., 1986. Low temperature hydrothermal barite of Nahal Hever. Isr. Geol. Soc., Annu. Meet., p. 45.

Gross, S., 1977. The mineralogy of the Hatrurim Formation., Israel. Isr. Geol. Surv. Bull., 70, 80 p.

Gvirtzman, G. and Buchbinder, B., 1966. The Tertiary project, progress report on geological research projects. Inst. Petrol. Res. Geophys. Rep. 1018, pp.11-17.

Kolodny, Y. and Gross, S., 1974. Thermal metamorphism by combustion of organic matter: isotopic and petrological evidence. J. Geol., 82: 489-506.

Matthews, A. and Kolodny, Y., 1978. Oxygen isotope fractionation in decarbonation metamorphism: the Mottled Zone event. Earth Planet. Sci. Lett., 39: 179-192.

Magaritz, M., Issar, A. and Azmon, E., 1983. On the genesis of discordant dolostone bodies in the northeastern Negev, Israel. Chem. Geol., 39: 93-114.

Menzies, M., Seyfried, W.J. and Blanchard, D., 1979. Experimental evidence of REE immobility in greenstones. Nature, 282: 398.

Michard, A., Albarede, F., Michard, G., Minster, J.F. and Charlou, G.L., 1983. REE and uranium in high temperature solutions from East Pacific Rise hydrothermal vent field (13 N). Nature, 303: 795-797.

Panchenkov, G.M. and Lebedev, V.P., 1974. Chemical kinetics and catalysis. Chimiya, Moscow, 592 p. (in Russian).

Raz, E., 1983. Geology of the Judean Desert. Isr. Geol. Surv. Rep. 83/3/S, 110 p. (in Hebrew).

Spiro, B., 1979. Thermal effects in oil shales - naturally occurring kaolinite and metakaolinite organic associations. Chem. Geol., 25: 67-78.

Spiro, B. and Aizenshtat, Z., 1983. Natural combustion and pyrolysis of bituminous rocks at the margin of Hatrurim, Israel. Advances in Organic Geochemistry 1981., Proc. of the 10th Int. Meet. on Org, Geochem., Norway, John Wiley and Sons Ltd., pp. 799-807.