Crystallinity index of mineral phases in jaspers from the Eastern Rhodopes

Индекс на кристалинност на минерални фази в ясписи от Източните Родопи

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Introduction

Jaspers from the volcanic-sedimentary formations of Late Eocene and Early Oligocene age in the Eastern Rhodopes in Bulgaria are poorly studied. There are just a few publications on this topic, mostly about the mineral composition of the aggregates. One of the first reviews is made by Atanasov and Yordanov (1986). Since the jaspers are opaque aggregates build mainly of SiO_2 phases mixed with other minerals that give their distinctive color, the authors used Mössbauer spectroscopy to define the exact chemical composition of the pigments. Further on studies combined with Electron paramagnetic resonance (EPR) spectroscopy revealed the distribution and status of iron in the three most important impurity mineral phases of such SiO_2 aggregates – hematite Fe_2O_3, goethite α-FeOOH and celadonite $K(Mg,Fe^{2+})(Fe^{3+},Al)[Si_4O_{10}](OH)_2$, which are related respectively to red, yellow to yellowish-brown and green colour (Kostov et al., 2017). Another important method for mineral identification is powder X-ray diffraction. Not only mineral phases can be defined, but after studying a relatively large amount of samples, the idea was suggested for determining the crystallinity of a SiO_2-bearing sample using X-ray data (Barsanov, Yakovleva, 1979; Plyusnina, 1983).

Materials and methods

Seventy powdered samples from *in situ* and alluvial occurrences of different (mainly red, yellow to brown and green) colour varieties of jasper from the Eastern Rhodopes, collected in regions distributed along five of the main rivers in that area (Banska, Arda, Varbitsa, Byuiyukdere and Krumovitsa rivers) were studied on an X-ray diffractometer Bruker D2 Phaser equipped with Cu tube and PSD detector. The experiments were done according the following conditions: Cu/Ni radiation with wavelength of λ=1.542 Å, 2θ range 5–70°, 30 kV and 10 mA; 1782 s. The recorded patterns were interpreted with the help of a software EVA with database COD (Crystallography Open Database). The same method is applied for 70 powdered jasper samples from the Eastern Rhodopes. It is a fast method and does not require sophisticated work in order to obtain some results. For the purpose of determination of the ordering in the SiO_2 matrix, the ratio K_1 between the intensities of the reflections 110 and 102 of quartz, which occur in the range d=2.45 Å and 2.28 Å is suggested (Barsanov, Yakovleva, 1979).

Another method was proposed by Plyusnina (1983). It is based on the powder XRD pattern which occurs at 2θ=68° for the reflection 212 in quartz. The pattern is composed by two peaks – a strong- and a weak-intensity one. After dividing the values of the intensity for the weak peak by the stronger one and multiplying by 10, a coefficient C_1 is obtained.

An alternative method to calculate the index of crystallinity can be applied by using the corresponding software that calculates automatically both the percentage of the amorphous and crystalline phases. After dividing the amount of amorphous by crystalline percentage the coefficient C_2 can be obtained.

Results and discussions

After applying the suggested methods, two charts are created. On the charts one can clearly observe the ratio between C_1 versus K_1 (Fig. 1), as well as C_2 versus K_1 (Fig. 2). The red varieties of jasper, pigmented by the mineral hematite are with maximum distribution along the Y-axis on Fig. 1. This indicates that these aggregates are with the highest degree of crystallinity. The red jaspers composed of fine grained quartz are with a low crystallinity index, and those composed of chalcedony – with a higher one (Kostov et al., 2016). The position of yellow to brown samples, coloured by goethite is distributed approximately in the middle area between both of the axis. The samples with the smallest area of distribution between both axes are those coloured in different shades of green by celadonite.

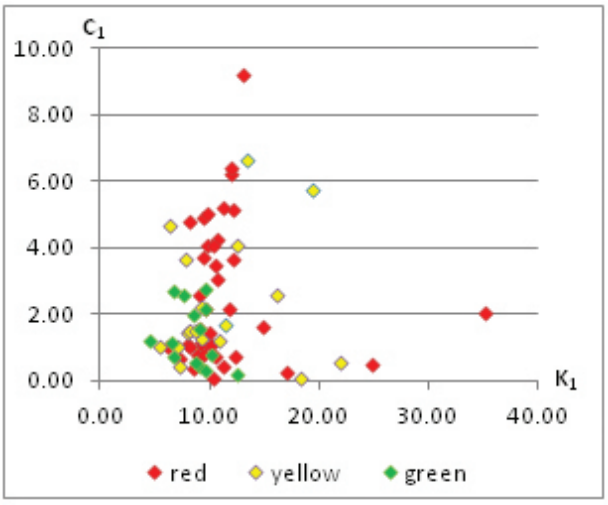

Fig. 1. Jasper samples of different colour: ratio K_1 vs C_1

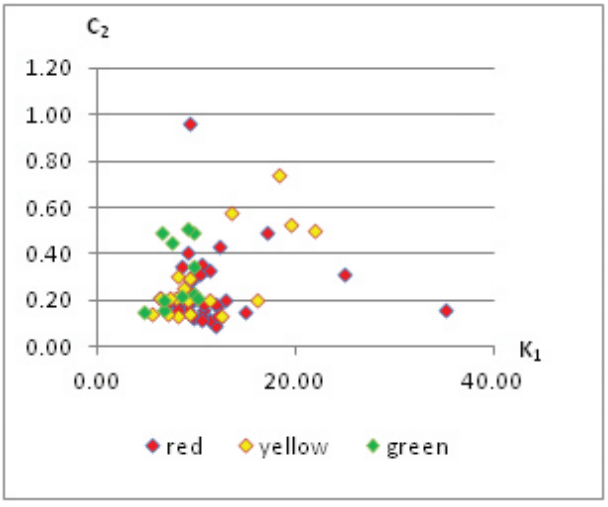

Fig. 2. Jasper samples of different colour: ratio K_1 vs C_2

After knowing the degree of crystallinity of a sample, it is possible for the SiO_2-bearing aggregates to be classified as different varieties such as jasper, jasper-opal and opal. This can be made after calculating the coefficient C_2 (Y-axis on Fig. 2). There is a certain trend which allows us to relate the SiO_2 aggregates to the appropriate variety. For values in the range 0.01–0.42, the samples can be defined as jasper, for values between 0.43–0.49 – as jasper-opal, and in the range 0.50–1.00 – as opal.

Conclusions

Both X-ray methods are reliable for determining ordering and index of crystallinity of SiO_2-bearing phases in jaspers. After comparing the results, it is clear that the data are more or less similar and the described methods can be used with relatively high reliability. The methods can be useful for a further on classification of jaspers and jasper-like rocks. The results show a tendency according which the red varieties of jasper are with the highest index of crystallinity, followed by the yellow to brown samples, and for the green ones the index of crystallinity is with the lowest value. This information, when correlated with the geological setting of occurrences of jasper samples and their host rocks can provide additional genetic information on how the aggregates were formed and what were the conditions during the mineral-forming processes.

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