Ruin marble: a record of fracture-controlled fluid flow and precipitation

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The ruin marble structure of the Cretaceous/Paleogene fine-grained marly limestone from the Outer Flysch Belt of the Western Carpathians has a non-tectonic origin, according to structural and sedimentological evidence. Distinctive offsets of coloured red-brownish ferric oxyhydroxide bands are not due to displacements along rock-cutting fractures, as they superficially appear to be. Evidences for shear movement along these pseudo-faults were not observed. Band offsets result from different velocities of pervasively diffusing fluids, precipitating ferric oxyhydroxides in corridors bounded by sets of mineralised systematic joints. During rock weathering, calcite-filled joints operated as barriers for lateral fluid diffusion, but enabled longitudinal diffusion along healed joints. Simple laboratory experiments have been performed to simulate the formation of natural ruin marble structure.

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INTRODUCTION

Spectacular ruin marble structure in banded porous calcareous rocks was described from classical Italian localities (Florence) in the last century. Stones showing this ornamental and decoratively coloured structure, named in Toscana “Pietra paesina”; used to be very popular in the manufacture of luxury furniture from Renaissance to secession times. Many rocks develop colour banding called Liesegang rings (Liesegang, 1896, 1913), which have nothing in common with primary sedimentary stratification (Civitelli et al., 1970; Dennis, 1972; Krug and Jacob, 1993; Krug et al., 1994, 1996; Jacob et al., 1994; Krug and Kruhl, 2001 and others). This is also the case for ruin marble, in which the colour banding results from periodic and rhythmic precipitation (Prager, 1956; Henisch, 1991) of impurities (usually ferric oxyhydroxides) due to infiltration of oxidising groundwater percolating through Fe-impregnated sediments during diagenesis and weathering. Calcite-filled, post-diagenetic joints divide the rock into numerous tiny blocks. The healed joints prevent lateral migration of the iron-bearing solutions, so that the banding extends in single blocks relatively independently (Shaub, 1953; Morawietz, 1958; Pivko and Marko, 1996), leading to an impression of apparent fault-related offsets along single blocks (Morawietz, 1958). This is the reason why the ruin marble structure is still frequently regarded as a product of faulting.

Civitelli et al. (1970) were the first who described details of a non-tectonic mechanism for the generation of Toscana’s ruin-marble structure. They showed that the limestone was pre-impregnated with homogenously distributed ferrous salts prior to the formation of the banding. Later on, capillary invasion of oxygen-rich meteoric water converted the colourless ferrous salt into banded brownish ferric oxyhydroxides by the process of periodic precipitation.

Buday et al. (1963) were the first who reported a “ruin marble” occurrence from the Western Carpathians. Having the opportunity to investigate in detail this rare phenomenon from a new Slovakian locality, we present observations and evidence corroborating a non-tectonic origin for this rock structure.

GEOLOGICAL SETTING

The new occurrence of ruin marble described herein is located in the neighbourhood of the village Horná Breznica near Púchov in Slovakia (Fig. 1). The ruin marble structure was observed in weathered blocks of fine-grained organodetritic...
clayey limestone embedded in the Quaternary eluvium (weathering residua). The limestone boulders come from 2–12 cm thick calcareous intercalations in a thin-bedded Maastrichtian-Palaeocene flysch, belonging to the Javorina Member of the Vlara Group (Potfaj, 1993). The flysch sequence represents the innermost part of the Outer Flysch Belt (the Biele Karpaty Unit) tectonically incorporated into the Pieniny Klippen Belt Zone.

**METHODS**

Structural observations were based upon macroscopic study of polished cross-sections of hand specimens under a binocular microscope. Thin-sections were investigated using a polarising microscope. Photomicrographs were taken using a CCD camera. Details of rock structure and variations in chemical composition across the colour banding were studied using transmission electron microscopy (TEM) of the polished rock surfaces. The limestone was examined by standardless energy dispersive (EDS) microprobe analysis. The homogenisation temperatures and salinities of calcite-hosted fluid inclusions within healed joints were determined using a LINKAM freezing-heating stage calibrated against phase transitions in synthetic fluid inclusions and inorganic solids of known composition.

Liesegang rings similar to those observed in the rocks were experimentally created in the laboratory. Gelatine with a dilute solution of potassium dichromate was used as the matrix medium, which was poured on to the bottom of a Petri dish. Drops of concentrated silver nitrate solution were added to the centre of the dish, or poured around its periphery. Some experiments with the Liesegang rings were performed using laboratory test tubes to make visible their three-dimensional structure.

**OBSERVATIONS**

The rock showing ruin marble structure is a fine-grained porous clayey limestone composed of remnants of planktonic
calcareous organisms and about 15% clay minerals. Coccoliths prevail over foraminifera chambers, which dominate in the laminae. EDS data show dominant CaO, with minor amounts of SiO₂, Al₂O₃, MgO, Na₂O, P₂O₅, SO₃, TiO₂, K₂O, FeO and MnO. The limestone is homogenous, showing only rare silty laminae with cross-lamination, ripples and ball and pillow structures. Trace fossils referred to Chondrites sp., filled with organic material, and sand-filled Sabularia sp. are ubiquitous.

Structural study was focussed on the spatial and structural relationships between joints, healed joints, and colour bands, which create the “ruined” character of the rock.

JOINTS

The banded marly limestone is cut by several sets of fractures. In hand specimens, fractures making apparent offsets of colour bands are oriented perpendicularly or at high angles to bedding surfaces, creating a tautozonal system. The very frequent (several fractures per 1 cm), regular, macroscopically planar fractures are arranged into one or two conjugate systems (Fig. 2), often reported from the competent rocks of the Outer Western Carpathians (Mastella et al., 1997; Zuchiewicz, 1998; Mastella and Zuchiewicz, 2000; Konon, 2001). One set of the system is dominant. Most of these fractures are healed with 0.01–0.05 mm thick calcite fill. The fractures macroscopically resemble perfect shear joints, but are in fact tension gashes filled with blocky calcite (Fig. 3a). Healed joints responsible for apparent band offsets use to be non-penetrative (Fig. 3b) and are created by overstepping en-echelon-arranged single segments (Fig. 3c–e). At least these parts of joints could not accommodate shear movement necessary for a tectonic origin of the offsets. Offsets between intersecting conjugate joints have not been observed (Fig. 2a). In several cases, the “intersection“ of two healed joints has a non-penetrative character (Fig. 3g, h), so that the joints could not accommodate a fracture-parallel displacement. This is supported by observations of bedding surfaces, which also are not offset by crosscutting joints. Healed joints creating offsets of colour banding do not offset other sedimentation-related phenomena such as burrows (Fig. 3f) too. Rare unfilled joints do not cause offsets of the colour bands.

The frequency of macroscopically discernible joints increases towards the periphery of rock boulders, where numerous joints are enhanced by Mn-rich dendrites (Fig. 2a). Lateral infiltration of the Mn-rich dendrites from fissures into the host rock proves the porous character of the rock. A dense system of sub-parallel, tiny fracture cleavage planes can be discerned at larger magnification on bedding surfaces enhanced by weathering. The fracture cleavage planes are more closely spaced than the joints of the rock boulder interior.

MICRO-VEINS

Healed joints creating apparent offsets of colour banding are truncated by thicker (0.25–1.7 mm) micro-veins (Fig. 4a–c). The micro-veins propagated perpendicularly (Fig. 4c) and oblique to walls during calcite crystallization. The micro-veins are classic mineralised tension gashes, rather than recrystallization veins (sensu Mišík, 1998), because they cause offsets of older joints. Calcite crystal diameters tend to increase with width of the joints, as in those observed by Passchier and Trouw (1996). The micro-veins are arranged in conjugate, apparently coeval pairs, as evident from the lack of offsets at the intersections (Fig. 4a), but simultaneous propagation of both coexisting micro-veins could not occur. Direction of the micro-vein opening can be reliably deciphered according to the offsets of healed joints. It is clear from geometric analysis that the micro-veins creating apparent conjugate pairs cannot be coeval. Their con-
Fig. 3. Thin section (a) and details from polished surfaces (b–h) of the ruin marble from Horná Breznica

a — calcite-healed joint with offset of colour bands and irregular walls; the joint has an evident tensional character despite an apparently distinctive shear character in macroscale; transmitted light, crossed polars; b — diminishing “microfaulgs” (healed joints) responsible for apparent band offsets; c, d — en-echelon arrangement of healed joint segments; e — overstepping segments of healed joint and micro-vein; f — burrows unaffected by band offsets; g, h — apparently conjugate healed joints
Fig. 4. Details from polished surfaces of ruin marble from Horná Breznica observed using a binocular microscope

- **a, c** — spatial and structural relationships between healed joints and micro-veins, double arrows show direction of vein opening; I — earlier micro-vein, II — later micro-vein; 
- **b** — location of details a and c on the polished surface of the rock; 
- **d** — double concave shape of fluid front (dashed lines with arrows show direction of fluid front propagation); 
- **e** — half concave shape of the fluid front (arrowed); 
- **f** — cross-cutting healed joints showing corrosion; 
- **g** — records of fluids overcoming bridge (arrowed) among overstepping segments of healed joints; 
- **h** — healed joints with corrosion (dashed lines with arrows show direction of fluid flow)
jugate character results from successive opening of individual micro-veins. Early micro-veins (I) offset the older healed joints. Micro-veins opening later (II) propagated parallel to the strike of the earlier micro-vein system, thus resulting in the conjugate character of their intersection. Both micro-vein systems postdate joints and predate colour banding.

Crystal growth direction in micro-veins and healed joints could be deduced from the pattern of colour infiltrations. The infiltrations rim the micro-veins and healed joints only from one side, and never occur along both walls (Fig. 4f). This indicates the propagation of crystal growth from one side towards the other. Fluids probably infiltrated the micro-vein or healed joint with newly formed calcite crystals along microscopic fissures.

FLUID INCLUSIONS

Primary aqueous two-phase inclusions have been found in micro-veins healed with blocky calcite. The inclusions homogenized to liquid at 130–132°C. Ice melted between –0.9 and –1°C, thus indicating the salinity of the aqueous phase to be between 1.5 and 1.7 wt. % NaCl eq. (Bodnar, 1993).

The inclusion fluid properties observed in the micro-veins from the ruin marble are similar to those in other mineralized joints, which are ubiquitous in the flysch sequences of the Outer Western Carpathians. According to micro-structural and tectonic observations, blocky calcite in composite joint infillings is coincident with the regional collapse and uplift of the Outer Carpathians (Świerczewska et al., 1999, 2000). Homogenisation temperatures fall into the range of 92–137°C, which is characteristic of blocky calcite from mineralised joints in the Magura Nappe (Świerczewska et al., 2000). Similarly, the observed salinity also overlaps the commonly observed range between 0 and 3.2 wt. % NaCl eq., which can be interpreted as resulting from various proportions of marine and meteoric water components in the parental aqueous fluid.

Assuming homogeneous trapping, the recorded homogenisation temperatures of fluid inclusions in blocky calcite represent minimum possible trapping temperatures. Depth of the calcite crystallization—e.g. depth of burial is difficult to infer only from the fluid inclusion data. We do not have relevant information concerning the origin of the fluids, geothermal gradient or palaeoflow pattern. If we assume typical geothermal gradients for such regions and tectonic settings to range from 20 to 30°C/km (Hurai et al., 2002), the depth of calcite crystallization could be about 4.3–6.5 km.

COLOUR BANDING AND RECORDS OF FLUID FLOW

The colour banding (Liesegang rings, bands) consists of alternating brown and pale stripes, implying a perfect stratification. Individual bands exhibit contrasting colours, varying from ivory through beige to brown. A violet colour is less frequent. The brown colour results from enrichment in ferric oxihydroxides and the colour intensity depends upon their concentration.

A concentric spherical arrangement of the colour bands has been observed in several samples (Fig. 2a), where banding is generally sub-parallel to surfaces of rock boulders. This means that arrangement of the bands is shape-controlled. The shape of the rock boulders was developed during the disintegration of the rock strata and weathering, which preceded the formation of bands.

Healed joints crosscut colour bands and cause distinctive offsets of the banding in neighbouring segments rimmed by joints. The magnitude of the offsets varies in different parts of the joints (Fig. 2b), increasing generally towards the centre of the boulder. In contrast, the banding tends to diminish towards the centre of the sample (Fig. 2a). In several cases, the banding pattern does not extend into the neighbouring segment (Fig. 2b).

Directions of fluid flow can be reconstructed from contrasting colours of Liesegang rings and infiltrations along joints. Some segments of dark bands rimmed by healed joints display concave shapes from one side (Fig. 4d). A half-concave shape is developed when related to only one of a pair of border joints (Fig. 4d). The concave shape of the bands is always oriented towards the direction of corridor widening (Fig. 2a) bounded by non-parallel joints. We suppose that the concave shape of the bands was formed in front of fluid flow driven by capillary forces in very narrow open fissures within healed joints. This mechanism accounts for acceleration and bending of the fluid front in the vicinity of joints. Schoonmaker et al., (1986) have described similar curvature of illite/smectite diagenetic fronts due to reverse faulting. Asymmetry or absence of curving observed in some corridors could be due to lack of capillary forces along thicker joints.

Longitudinal permeability of healed joints for fluids along contacts with wall rock is marked by distinctive dark infiltrations rimming healed joints (Fig. 4f). These infiltrations always occur only from one side of a corridor bounded by healed joints between rock wall and calcite fill. Dark drop- and flame-shaped infiltrations intruding the rock resemble corrosion effects. The corrosion is often accompanied by concave fluid fronts (Fig. 4e). It seems plausible that capillary forces operating along joints have been important. Interruption of joints has not stopped circulation of fluids. Migrating fluids have been able to overcome overstepping bridges among single en-echelon-arranged segments (Fig. 4g).

Corrosion along joints has played an important role in the origin of colour bands. Several lines of evidence indicate that development of the bands was triggered by lateral infiltration of oxidising fluids, migrating along the joints (Fig. 4h).

LABORATORY GROWN LIESEGANG RINGS

To simulate natural banding (Krug and Brandstädtter, 1999), artificial Liesegang rings propagating from the centre towards the periphery of a Petri dish (Fig. 5a) and vice versa (Fig. 5b) have been synthesized. The banding has developed in more or less narrow belts comprising red-brownish silver nitrate precipitates. The artificial Liesegang rings are characterised by bifurcations (Fig. 5c) and apparent dislocations (Fig. 5d, e). Three-dimensional spiral banding was observed in one test-tube experiment.

Several orders of banding can be discerned at higher magnification. Bands showing less contrast parallel to macroscopic bands of higher rank were formed during some experiments (Fig. 5c).
Fig. 5. Experimentally created Liesegang rings of silver nitrate in gelatine matrix at the bottom of Petri dishes

a — Liesegang rings propagated from centre to periphery of Petri dish; b — Liesegang rings propagated from periphery to centre of Petri dish; c — detail of band bifurcations; d, e — interruptions of banding developed during the process of precipitation; f — band offsets created during the process of precipitation due to interruptions — striae made in gelatine matrix
The role of barrier joints known from ruin marble structures (Krug et al., 1994, 1996) has been also studied. Impermeable joints have been simulated using narrow striated fissures at the bottom of the dish, which caused interruptions of cohesion in gelatine. Narrow channels operated due to absence of gelatine as barriers for diffusion, as in healed joints in marly limestone. Due to these fissures, diffusion fronts have propagated independently in individual corridors, resulting in apparent offsets (Fig. 5f).

Experimentally obtained banded structures are essentially similar to those observed in nature. These experiments have, of course, serious limitations compared to natural processes. First, they were undertaken in the “two-dimensional” space of the Petri dish. Only a few experiments have been conducted using laboratory test tubes. Second, the physical properties of gelatine, the diffusive medium used, are quite different from those of solid, porous, fine-grained sedimentary rock, in which the natural banded structures occur.

DISCUSSION

According to our observations and survey of available literature, a succession of events during the formation of “ruin marble” structure from the Horná Breznica locality can be reconstructed:

1. Pelagic deposition of calcareous mud with terrigenous material took place in the Magura flysch basin. The mud was composed mostly of coccoliths, locally with coccospheres and planktonic foraminifera. This quiet sedimentation was interrupted by deposition of horizontal or low-angle cross-laminated silt with fragmented foraminifera chambers. These silt laminae were probably deposited by bottom currents. Some cross-laminar ripples were then loaded onto underlying muddy beds, creating ball and pillow structures. At the same time, a sliding along slope deformed the laminae. After deposition and syn-sedimentary deformation, the sediment was bioturbated by trace fossils.

2. After deposition, the sediment underwent burial diagenesis accompanied by a decrease in porosity and compaction at lithostatic pressure. This process must have taken place in water-saturated sediment under conditions of high pore fluid pressure, which plays an important role in the formation of joints in tectonically non-deformed sediments (Price, 1981; Černyšev, 1983; Price and Cosgrove, 1991; Dadlez and Jaroszewski, 1994). During diagenesis or sedimentation, the rock was homogeneously impregnated with a ferrous salt.

3. Later, increasing fluid pressure in already lithified sediments sucked the oxygen-rich water from the surrounding area into the rock interior. Some finely stratified and apparently faulted geological structures are likely Liesegang bands of ruin marble type, because it is impossible to restore a pre-faulting stage of this structures by simple length-line balancing. For example, laminated mudstone, apparently affected by conjugate normal faults (Oszczypko, 1997), shows distinctive offset fluctuations along individual “faults” (Fig. 6). This behaviour could not be caused by post-sedimentary normal faulting, or syn-sedimentary

Due to hypergene processes individual blocks of rock were saturated with oxygen-rich water, which reacted with the ferrous salt and colour banding was developed. The banding was strongly affected by healed joints and micro-veins, which caused apparent offsets of banding. The banding was formed due to diffusion/self-organization inside corridors bounded by single joints, which developed independently. Fluid fronts advanced in the individual corridors with different velocities and accompanying precipitation created different banding patterns. Capillary forces expelled these fluids along contacts between healed joints and walls, resulting in acceleration of fluid front infiltration at the borders of corridors. This resulted in bending fluid fronts in corridors as recorded by occurrences of infiltrations. Infiltration corrosion often intruded far into the rock interior, where distinctive banding was not yet developed (Fig. 2a).

5. Saturation of rock boulders with Mn-rich fluids was the last event recorded by Mn dendrites along tiny cleavage planes discernible only along narrow rims of the studied boulders (Fig. 2a). These cleavage-related joints are devoid of mineralization and did not affect the colour banding.

Some samples from Horná Breznica show concentrically zoned colour banding controlled by the shape of the rock boulders. This, together with observations of fluid flow directions, indicates that the fluids propagated and the ferric oxyhydroxides precipitated progressively from the periphery towards the centre of rock boulders. It seems thus plausible, that rock boulders sucked the oxygen-rich water from their surroundings. Two models could be proposed to account for the mechanism of fluid infiltration:

1. According to first model, the liquid infiltrated pervasively through interconnected pores along the whole surface of the rock boulders, but with variable velocities in individual corridors rimmed by healed joints. Differences in the velocity of fluid propagation and mineral precipitation between neighbouring corridors increased towards the centre of the rock boulder. The model supposes sucking of liquid from the surrounding area into already isolated, fluid-undersaturated boulders, so that a concentric pattern of colour bands could be created. These conditions could be reached during hypergene processes, after disintegration of sedimentary beds due to weathering.

2. The second model favours capillary force-driven local migration of oxygen-rich water through narrow fissures between newly formed calcite crystals and the rock walls. This kind of fluid migration accounts for precipitation of ferric oxyhydroxides along healed joints, resulting in corrosion. Colour bands could be created by lateral diffusion, extending from joints saturated with oxygen-rich water into the rock interior.

The absence of joints in many other worldwide-described banded structures is a serious argument against the second model. It is thus likely, that in the case of the “ruin marble” from Horná Breznica, both mechanisms could operate.

Some finely stratified and apparently faulted geological structures are likely Liesegang bands of ruin marble type, because it is impossible to restore a pre-faulting stage of this structures by simple length-line balancing. For example, laminated mudstone, apparently affected by conjugate normal faults (Oszczypko, 1997), shows distinctive offset fluctuations along individual “faults” (Fig. 6). This behaviour could not be caused by post-sedimentary normal faulting, or syn-sedimentary
faulting. The sediment is strongly laminated and contrasting thin-laminated markers exhibit distinctly different patterns on opposite walls of the “faults”, which thus appear to show conflicting offset-related kinematics. This is suggestive of a diffusion-related, rather than fault-related origin. Similar arguments could be used to account for origin of apparently faulted laminated Pleistocene sediments (Mandl, 1988, fig. 1.1–6).

Banded carbonate from the Vel’ká Fatra Mts. could serve as another example of banding due to the interaction of chemical reactions, diffusion, precipitation and crystallization (Fig. 7). The sample shows alternating pale and dark bands, with white stripes composed of coarse crystalline calcite and dark stripes contaminated by clay and/or organic impurities. Banding is arranged as a tectonic SC mylonite structure (Kohút, 1989; Polák et al., 1997), thus implying a tectonic origin. In detail, however, single bands are not juxtaposed, as tectonic duplexes should be. Contacts between bands are diffusive, often very irregular. They are only locally superimposed by styloliths — pressure solution surfaces. The arrangement of the bands is very similar to that of banded siderite in zebra-type mineralization, interpreted as a Liesegang band phenomenon (Krug et al., 1996). These features seem to be reliable arguments supporting a non-tectonic origin of this structure, belonging genetically to lithogenetic self-organization processes.

Hitherto described examples of natural banded structures indicate that care must be exercised in the genetic classification of laminated and stratified fabrics, because diffusion and self-organization processes could be also responsible for the origin of such often apparently faulted structures.

Due to ubiquitous occurrence of banded Liesegang structures in rocks, the factors responsible for banding should be universal. In experiments, banded structures have formed due to diffusion driven by the homogenisation principle. It is known that diffusion flow is controlled by Fick’s laws, and always propagates from areas of higher concentration to areas of lower concentration, to establish a homogeneous distribution of matter (Turner and Verhoogen, 1960). This means that spontaneous natural processes lead to an increase of entropy and decrease in organisation of the system, but diffusion does not produce a homogeneous distribution of silver chromate in laboratory and of ferric oxyhydroxides in nature. The banded structure results from interaction between silver nitrate and potassium chromate, and ferrous salt and oxygen-rich water respectively. Elements are transported by diffusion, and chemical reactions during the Ostwald-Liesegang-cycle organize them into a banded structure. In addition to these methods of creating Liesegang banding, self-organising precipitation may be influenced for example by electromagnetic fields (Jacob et al., 1992, 1994).

CONCLUSIONS

A study of ruin marble from Horná Breznica corroborates a non-tectonic origin for this structural phenomenon. The structure is apparently a product of micro-faulting, but an “undeformed” stage of the banding cannot be restored by any kind of balancing. Fractures responsible for banding offsets represent healed joints and micro-veins, without any expression of shear movement, because:

— Some fractures creating distinctive offsets of banding do not-penetrate the whole of an observed rock mass, so they

Fig. 6. Structure redrawn from laminated mudstone affected by brittle discontinuities from the Outer Western Carpathians (Oszczypko, 1997)

Two distinctive marker horizons (thick and thin) are shaded; complex apparent faulting does not allow restoration of undeformed sediment; arrows express sense of apparent marker offsets

Fig. 7. Banded calcite vein displaying apparent internal tectonic S–C fabrics (Kohút, 1989)

Character of banding suggests a diffusion-related origin of banding created during crystallization

Ruin marble: a record of fracture-controlled fluid flow and precipitation
could not accommodate the displacement necessary for a tectonic origin. 
— Fractures apparently responsible for offsets of banding do not offset burrows, which always predate the Liesegang bands. 
— Magnitudes of band displacement are different in different parts of a single fracture, and tend to increase towards the centre of concentrically arranged bands. 
— Banding patterns in neighbouring corridors bounded by fractures do not continue exactly from one corridor to another.

The structure of colour banding in the ruin marble results from uneven precipitation of ferric oxyhydroxides from fluids percolating through porous rock in corridors bounded by calcite-filled joints. The healed joints operated as barriers to the fluids which, infiltrated the rock during hypergene processes. The process caused independent development of colour banding in individual corridors, thus implying a faulting-related phenomenon. Formation of the colour banding has occurred after lithification, and very likely also after disintegration of rock into individual blocks, because the concentric bands are arranged parallel to the outer shape of the rock boulders. Oxidising fluids propagated from the surface of the rock boulders into their interior, due to capillary forces in micro-fissures rimming sealed joints. Clear evidence of capillary forces affecting banding along sealed joints and micro-veins indicate that the rock had to be fluid-undersaturated prior to water infiltration.

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