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The role of shear movement on graphitisation of organic matter - In-situ analysis done by energy dispersive X-Ray diffraction and electrical impedance spectroscopy

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A well interconnected network of graphite can increase the electrical conductivity of rocks by orders of magnitude. Graphitic conductors high in electrical conductivity were detected in crustal rocks by magnetotellurics covering areas of up to tens of kilometres. These conductive zones often correlate with tectonic movement and it was speculated about its relation to former collision zones. From petrological studies it is known that temperature and strain influence graphitisation in metamorphic rocks. High-grade metamorphic rocks exhibit full graphitisation of former organic matter, while tectonic movements along thrust faults locally may rise rank, probably due to concentrated frictional heat. Rank is frequently higher in strongly folded areas than in unfolded areas, thus supporting the idea of graphite formation under the influence of mechanical stress. Anomalous high reflectance values of vitrinite in very narrow coal films in contact to thrust planes or within shear zones were explained as the result of local and short-term frictional heat generated during stick-slip faulting; thus graphite formation is facilitated to a considerable degree by strain and strain energy. Transmission electron microscopy (TEM) of tectonally deformed anthracite confirmed the assumption that strain is required to align the aromatic lamella to get an microtextured arrangement of carbon-planes. This was confirmed by the observation that shear strain is effective in promoting an increase in both, the maximum reflectance of the 002 X-Ray reflection and the anisotropy of vitrinite reflectivity. This supports the idea that graphitisation is triggered by mechanical stress and shear movement, thus being due to a considerable degree to strain and strain energy, but the physical and chemical causes

for transportation, precipitation and interconnection of graphite over greater distances are not well understood in view of pressure, temperature and dynamic conditions during metamorphism in the past. Graphitisation is the final step of a maturation process of cokes where adsorbed volatiles are removed and the degree of molecular ordering is increased. The formerly randomly oriented aromatic lamella of the carbon network are interconnected over larger distances and thus increase the electrical conductivity.

This process of ordering and reconnection of aromatic lamella was reproduced under in-situ conditions in laboratory experiments using impedance spectroscopy to follow changes in electrical conductivity and the nature of the conduction process, while energy dispersive X-Ray diffraction data were used to measure the increase in crystallinity of the carbon phase.

Graphitisation experiments were performed in the pressure interval 0.5 up to 0.7 GPa, and at temperatures ranging from 450˚ and 600˚C to study the influence of pressure, stress, shear movement and temperature on graphitisation of organic matter. Less ordered carbon is a rather poorly conducting phase, but well ordered graphite is as conductive as metallic compounds. Therefore the frequency dependence of the complex conductivity was measured on anthracite, brown coal and black-shale in the frequency range from 0.7 up to 100 kHz under in-situ p,T-conditions. Tectonics were considered by application of a) hydrostatic pressure, b) a stress field and c) under the influence of shear movement. The measurements were performed as a function of time due to the hindered kinetics of the graphitisation reaction. A decrease of the bulk resistivity was measured as a function of time (up to 6 weeks) covering about three orders in magnitude. At the same time the complex response exhibited a continuous decrease of the imaginary part of the impedance, thus indicating that "quasi-metallic" conduction dominates more and more the charge transport. The increase in conductivity was significantly enhanced when shear movement was applied instead of stress or even hydrostatic pressure at same temperature conditions. These results were confirmed by energy dispersive X-ray diffraction (EDX) measurements that revealed a low degree of ordering of the untreated starting material by showing a broad amorphous 002 graphite reflection. After p,T-treatment and shear movement nearly perfect crystallised graphite was detected by measuring a sharp 002 reflection peak. Differential-Thermo-Analysis (DTA) and Thermo-Gravimetry (TG) revealed a continuous loss of volatiles of anthracite in the temperature range 100˚C up to 850˚C (9.9 weight % at 450˚C).

To enhance the electrical conductivity two processes are required: a chemical one to remove adsorbed volatiles and a physical one to form a well interconnected network. It is essential to notice that shear triggers the graphitisation of less ordered carbon at temperatures as low as 450˚C.