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OXIDATION OF CARBON: WHAT WE KNOW AND WHAT WE STILL NEED TO KNOW

Osvalda Senneca*

Istituto di Ricerche sulla Combustione, C.N.R., P.le Tecchio 80, 80125 Naples, Italy

Abstract

Solid carbons include a very wide range of materials, form biomass and wood, with their disordered structure, high content of oxygen and hydrogen, to progressively more ordered and pure materials, passing through the large family of coals of different geological age and degree of graphitization, through soot, ending up with nearly perfectly organized materials such as graphene, graphite, nanotubes, fullerene (Fig.1).

The large variety of carbonaceous solid materials corresponds also to a large variety of applications. Beyond the numerous and strategic industrial applications, from the field of energy production to that of innovative materials, we can recall also the role played in natural and biological processes: atmospheric dust, for example, is responsible of several meteorological phenomena, carbonaceous nanoparticles are related to lung deseases, solid active carbon is employed in drug delivery etc...

We can probably say that oxidation of solid carbon is the reaction that mankind exploited earliest, since men learned to light up a fire. Over the ages combustion of wood first and of fossil fuels later, has continued to sustain the energy demand and the progress of human society and, despite the efforts of the scientific community to find alternative routes for energy production, a significant fraction of the global energy demand is likely to remain dependent on combustion of solid carbon also in the near future.

In the second half of the last century the need to reduce pollution from coal fired power plants, in particular soot, NOx and SOx emissions, motivated significant research on the chemistry as well as on the physical phenomena involved in coal combustion. The basis of our current knowledge in carbon oxidation was formed at that time, allowing some important scientific and technological advances in the field of coal to energy.

Over the last decade the environmental challenge and the rising concerns about climate change have become prime drivers of scientific and technological development in the field of solid fuel combustion and gasification. Studies in this field are targeted to the development of novel combustion concepts that make CO2 capture and sequestration inherently more economical and feasible. This goal is pursued by re-thinking combustion technologies in such a way that carbon dioxide is highly concentrated at the exhaust, possibly free of any contaminants, so that it can be more easily compressed and disposed of through the sequestration path. Chemical looping combustion and Oxy-combustion are two examples of these new generation “capture

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* Corresponding author: Tel.: +39 0817682969
E-mail address: senneca@irc.cnr.it

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ready" combustion technologies. In such innovative technologies carbon oxidation is carried out under more severe conditions than ever, for instance at very high temperature and pressure, in CO2 rich environments, or experiencing continuous temperature and oxygen swings. The knowledge so far available on coal combustion has proved unable to explain the behavior of coal oxidation under such conditions.

On a different, but parallel route, the current rush towards the development of innovative carbon based materials, for a multitude of industrial applications, has also entailed investigations on the affinity of solid carbons towards oxygen. The attention has been put, in this case, on ordered carbon materials such as graphene and graphite, carbon composites, carbon nanotubes etc. and on how their properties are altered by oxidation under mild oxidizing or even ambient conditions.

It is astonishing that despite the very long time research on coal, on the one side, and the more recent but massive research carried out on graphene on the other, there are still many unresolved (or poorly understood) aspects in the heterogeneous oxidation of carbons: : what are the bonding modes of oxygen on different solid carbons? What is the range of atomic equilibrium distance? What are the spin state of the absorbing site and of the absorption complexes? What is the energetic range of C-O bond? Is it a strong physisorption or a weak chemisorption? What are the kinetics of the absorption and desorption on the molecular scale?

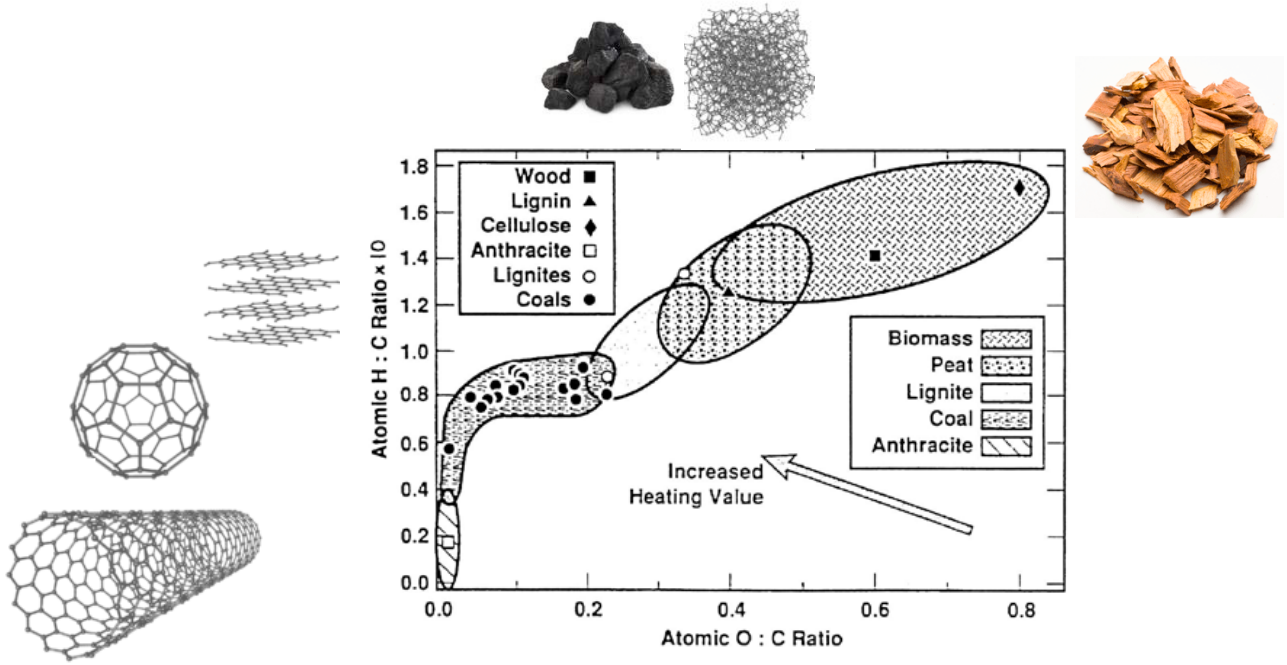

Fig. 1 Location of carbonaceous solid materials in the Van Krevelen diagram

Keywords: Carbon; Coal;Oxidation; Clean coal; Oxy-combustion; Chemical looping

1. Fundamental understanding of the interactions between solid carbon and oxygen

The earliest works on oxidation of solid carbon were produced in the context of coal combustion, although they were often carried out on model carbon materials such as graphite.

By the late 1950s the order of magnitude difference in coal combustion rate among various coals was clear enough to the combustion community and motivated extensive research on the rate of combustion of different coals. Notably in early kinetic models a very simplified approach was used and the rate of combustion was commonly expressed by means of power laws expressions.