

Chapter 3

Cleaner Energy Fuels: Hydrodesulfurization and Beyond

Neha Sareen

Banaras Hindu University, India

Subrato Bhattacharya

Banaras Hindu University, India

ABSTRACT

Today, SO₂ pollution has become a glaring problem especially in India and China. Thus, desulfurization of fossil fuels has become an essential area of research. Advances in experimental sciences to explain the desulfurization mechanism will be beneficial to the rational designing of more effective HDS catalysts. Several reaction pathways involving desulfurization of thiophene have been compiled. Also, a repertoire of the desulfurization pathways encountered in the author's laboratory has been provided. The results will not only throw light on some unusual mechanisms of desulfurization process but also break the popular belief that desulfurization is limited to C-S bond cleavage only. The reactions may also serve as a basis for engineering optimal catalysts for future applications.

INTRODUCTION

The air that we breathe is precious and life giving. However, we have taken it for granted for too long. In the worldwide zeitgeist of rapid economic growth and technological advancements, we have gambled away our very right to breathe fresh air. Since the beginning of the industrial revolution, widespread combustion of fossil fuels by humans has been adding a noxious cocktail of fumes into the atmosphere. Now, as the industrial setups encroach in our residential and agricultural areas, adverse effects on our environment and health have become evident. The world and specially the developing nations such as India, China and south-east Asia are shrouded in a blanket of smog, struggling for a breath of clean air. A state of emergency was declared during the *Malaysian haze* of 2005 when pollution levels (Air Quality Index (AQI)) sky rocketed beyond the permissible 500 mark. In 2013, the New York Times ran an article on Beijing's air quality describing it as "Crazy Bad". A recent report showed that a third of the urban population in India lives in cities where the PM₁₀ (particulate matter less than 10µm in diameter)

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levels were found to be “critical”. The gravity of the situation can be judged by the number of lives lost and the irreparable damage caused to our ecosystem. In 2008, nearly 3000 people died pre-mature deaths in Canada due to acute short-term exposure to air-pollutants. These excerpts distinctly reverberate the fact that air pollution, and particularly SO₂ pollution is an environmental crisis that demands immediate attention.

SO_x and sulfate aerosols are some of the major air pollutants responsible for incidents of smog. The Indo-Gangetic planes are especially vulnerable to a marked increase in the levels of black carbon and sulfate aerosols every year before monsoons, which adversely affect agriculture in this fertile area (Ramanathan *et al.*, 2005; Ramana *et al.*, 2010). A recent study by Burney and Ramanathan, 2014 showed that short-lived climate pollutants (SLCPs) such as black carbon, surface ozone and sulfate aerosols were responsible for about 36% decline in wheat production in India in 2010. If recent reports are to be believed, SO₂ emissions in India escalated by over 40% during 2005-2010 (Klimont, Smith, & Cofala, 2013). While China continues to be the world’s largest SO₂ emitter, these findings catapult India to a close second.

SO₂ is released into the atmosphere during volcanic eruptions and wild fires (natural sources) and by burning of fossil fuels at large stationary combustion units (anthropogenic sources). Fossil fuels’ combustion in coal-fired electric power and transportation sectors account for maximum anthropogenic SO₂ emissions. Coal burning electric utilities are liable for 73% of the total SO₂ emissions in US. (See US-EPA) The amount of sulfur in coal varies from less than 1% -over 12% (Calkins, 1994). SO₂ gets oxidized to sulphuric acid in the atmosphere which in turn gets deposited as “acid rain” (Ehhalt, 1999). Precipitation can be many times more acidic than natural rain with a *pH* of 3 or less and poses serious environmental and health hazards. SO₂ is also the prime precursor for anthropogenic aerosols. Sulfate aerosols are formed in the atmosphere by nucleation of gas-phase species such as SO₂ and H₂SO₄. In a seminal paper Ward, 2009 shows that scientists may have, until now, underestimated the role of SO₂ in bringing about the climate change. His work, which spans the effect of volcanoes on climate change over the past 46,000 years, shows that each of the episodes of rapid global warming were concomitant with episodes of extreme volcanism (a large number of volcanic eruptions within a short time span). The bulk of the paper is flowing lava of evidence arguing the role of SO₂, opposed to CO₂, as the key initiator of global warming and climate change.

By the turn of the 20th century, anthropogenic SO₂ influx had reached 150Mt of SO₂ per annum. (Smith, Conception, Andres, and Lurz, 2004; Smith, Pitcher, and Wigley, 2001) Eliminating acid rain and curbing sulfur emissions from fossil-fuelled power plants has since then become an environmental goal. The US EPA Clean Air Act had made it mandatory to keep the S levels in gasoline and diesel within 30 to 50 ppm since the year 2005. The permissible limit was further reduced in 2009 to 10 ppm -15 ppm. (Brunet, Mey, Pérot, Bouchy, and Diehl, 2005) For fuel cells, using gasoline as feed the limit is even lower at 1ppmw. (Hernández-Maldonado and Yang, 2004) Stricter environmental regulations to limit the sulfur content in coal, gasoline and diesel are steering us into a world of “zero” sulfur/sulfur free fuels. Traces of sulfur in diesel fuels act as poison for the oxidation catalysts in the emission control system thus reducing their efficiency to oxidize harmful carbon monoxide and hydrocarbons (Corro, 2002). In addition, rapidly declining resources of crude sweet oil makes it even more crucial to tackle the problem of sulfur removal from fuels. A commercially viable desulfurization process must be a catalytic process with a low energy pathway that involves cheaper and reusable catalyst. In order to develop efficient low energy pathways for fuel desulfurization, it is essential to master the underlying science. The reactions of transition metal and organometallic complexes in solution are often reproduced

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