Mechanisms and Models of Soot Formation: Final Discussion and Perspectives

chaired by

Heinz Georg Wagner

Wagner: Now, we should try to determine the development into the future and I would like to take the chance to ask those of you who had certain perspectives concerning soot formation to take part and to contribute to this discussion. You have seen here different approaches to the soot problem. One is to determine the conditions under which you definitely do not get soot. That is information which is already accepted by the engineers and that is what they need. Some other people tried to look deep into the chemistry. There were also people who tried to build bridges across those parts which are unknown in chemistry and feed them into the models to describe everything. As you may remember, mankind got the fire about 10,000 generations ago and I sometimes imagine how God was thinking about which kind of fire should he give to these “animals” here on earth. If you had to make the decision you would decide to give them the diffusion flame. God did the same thing and he was of course absolutely aware of what he did. He preferred a complicated system with a high degree of intrinsic stability and therefore a small risk. So this is the reason why we have to face all these problems and have to make complicated models in order to understand what is going on in a diffusion flame. That was a little introduction and now I would like to start the discussion. Firstly, I would like to hear about your perspectives, where are the gaps of knowledge, and where do we need information urgently? Secondly, I would like to ask those of you who are willing to tell us about their further plans in their research so that we can get an overview of what is going on. I know that the last question is probably not very easy to answer. I see Dr. Sarofim is willing to start the discussion.

Sarofim: I would like to address a question to B. Moss. He gave us his rule of how many variables you can add to a turbulence model. In his own models of sooting he uncoupled and post-processed the kinetics from the turbulence and the flow field calculations. When dealing with pollutants which in many cases don’t influence the flow and temperature field why not have a complex pollutant model which is post-processed.

Moss: The straightforward answer is, for some pollutants I’m sure that this is true. The simplest case is to take the thermal NO\textsubscript{x}. Something which is thermally insignificant you can post process increasingly elaborate models. The issue would clearly be that you design your device ever better and the NO\textsubscript{x} concentration falls and the balance shifts from being thermal NO\textsubscript{x} to
prompt NO\textsubscript{x} or to fuel bound contribution or what ever. Then your simple model ceases to be simple. I'm not sure that it is possible really in the context of soot because of the strong radiative coupling. That is an issue that I. Kennedy has partially addressed and about which I have serious doubts. We have probably all flirted with trying to put detailed radiative calculations into even atmospheric turbulent flames and that rapidly becomes a very complicated issue. I believe that if we do offer more then two or three additional scalar variables whether you treated them in a very superficial way as I have done or whether you adapt the Kennedy, Kollmann, Chen formulation and go to a multidimensional scalar pdf. I suppose the greatest exponent in this area is S. Pope. Even there, even for the gas phase scalars, we are not prepared to go beyond four scalars at this point in time. The complexity of doubling that by adding the soot model is unacceptable at this point in time. We may be able to compute some very simple flame geometries but I can't see that there is a practical solution in which the soot problem has added more than two or three variables to the scalar dimension.

**Santoro:** I want to try to get things going by giving a summary of what I thought came out from this meeting that was an advance on the last meeting and where I see some controversies. If you give me a few minutes, I'll run through some points. I think at the last meeting we were just seeing in the preparticle chemistry area, the emergence of this idea of what is now called reactive coagulation. And at that time we were arguing about how do these things that are 1000 or 2000u stick together. At this meeting people are proposing reactive coagulation of 100 and 200u species and saying that this route may be, under certain flame conditions, the dominante way to soot as opposed to the acetylenic build up mechanism. So I see a challenge to further look at the kinetics of those types of isomerisations and dimerisation versus the Frenklach type of build up mechanism. I see that as an issue in my mind since, as much as I like the data, I haven't seen the physics that makes me say I'm comfortable with reactive coagulation of such smaller species. The second change has come in a subtler way and I think it deals with our focusing on the surface area question from a different perspective. We had a debate also in Göttingen about the Bockhorn and Harris flame results and what they meant. Now the active site argument has arisen, it is not a new concept, but what I think it does is make us focus much more on the nature of the soot particle surface. And that is something that I haven't seen in a while. There are other groups of people who presently do work in this area such as people who investigate additives for fuels or those in the carbon black industry. They know a lot about how to treat surfaces of particles. There are measurement techniques that we haven't taken advantage of yet. I'm trying to collaborate with workers in this area. The third point I'd like to make is, I firmly believe that the work that is going on in the measurements of the fractal aspects of soot particles is changing the way we are going to