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Efforts have continued in primarily the following two directions during the past few months: 1) The development of high-resolution solid-state ^1H and ^{13}C NMR imaging techniques, based on magic-angle spinning (MAS) and rotating gradients, that will be suitable for coal samples; and 2) the evaluation of direct polarization (DP) approaches, in place of cross polarization (CP), for the ^{13}C MAS analysis of coal.

The reliability of ^{13}C NMR spectroscopy for coal analysis, in terms of the number of carbon atoms that can be detected, has received a lot of attention during the past ten years; and this topic was the subject of a special "debate" in *Fuel*. The general conclusion of that debate was that the usual cross-polarization experiment is not able to yield quantitatively reliable results because of extremely complicated spin dynamics. Therefore, one should avoid this experiment if possible, performing the DP ^{13}C MAS experiment (DP-MAS) instead. There have been suggestions in the literature that 90-100% of the carbon-13 nuclei can be observed in coal by this approach, but definitive studies have not been reported previously.

It is interesting to note that, although usually all scientists agree about the uncertainty in the CP-MAS ^{13}C experiment, they typically do not avoid using it to determine the ^{13}C spin-lattice relaxation time, which is a crucial parameter in DP-MAS. This situation and our previous studies of

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coal structure encouraged us to perform detailed studies of ^{13}C spin-lattice relaxation on all eight Argonne Premium coal samples.

Because of the heterogeneity of coal structure, it is reasonable to expect that the ^{13}C T_1 value would vary with the CP contact time that is used in a cross-polarization experiment. For example, the carbons in condensed aromatic rings are presumably far away from protons, so they will be invisible in a ^{13}C T_1 experiment that employs CP contact times on the order of only one millisecond, which is a very common value in NMR studies of coal. This behavior was indeed observed among most of the Argonne Premium coal samples. Our studies show that the best contact time for determining the largest ^{13}C T_1 values of aromatic resonances is about 6 ms. The relation between the ^{13}C T_1 value and the CP contact time for aliphatic and aromatic resonance has been studied systematically. The ^{13}C T_1 values obtained by this approach are typically usually larger than values reported in the literature. We found, after analyzing these results consistently, that the number of ^{13}C spin detected by the direct-polarization experiment varies between 87 and 98%. These results are being summarized in a manuscript for publication in *Analytical Chemistry*.

In spite of uncertainty in quantitation of the cross-polarization experiment, it is very unlikely that it will be abandoned in coal science. Therefore, we undertook detailed studies of rotating-frame spin-lattice relaxation for all Argonne samples. Again, when relying on the CP approach, the observed rotating-frame relaxation times are found to depend on the CP contact time. We also found that the ^{13}C rotating-frame spin-lattice relaxation time is too small to be neglected in the variable contact-time equation, as is done essentially universally in the ^{13}C CP-MAS literature on coal.

We have already collected a large set of experimental data, which will require sophisticated numerical computer analysis in order to make marked progress in understanding the cross-polarization dynamics in coal. This effort will likely lead to substantial improvements in our knowledge of coal structure.

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