Chemical Resolution Mass Spectrometry: On-line Separation Chemistry for Radionuclides *inside* a Mass Spectrometer

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Chemical resolution mass spectrometry is a new approach to the removal of isobaric interferences in elemental and isotopic mass spectrometry. The approach is based on gas phase reactions of ions inside a mass spectrometer. Between the ion source and the mass analyzer, the ions are allowed to react with a reagent gas. Ideally, interfering ions react—changing their mass-to-charge ratio—while analyte ions do not. The reaction mixture is then mass analyzed and only the analyte ion signal appears in the mass detection channel of interest. This chemical resolution approach to interference reduction is more effective than high resolution mass spectrometry (MS) in that there is often little or no signal loss for the analyte of interest while the interfering species can be reduced by many orders of magnitude. Chemical resolution MS depends on the chemical properties of the ions; reagent gases with appropriate reactivity must exist for a given separation to be possible. High resolution MS is typically used for the separation of molecular ion interferences since most atomic isobars require extremely high resolution for effective separation, e.g., Sr-90/Zr-90 requires resolution of 29,600; Cs-137/Ba-137 requires resolution of 109,000. In contrast, many such separations have been demonstrated using chemical resolution MS. High resolution MS has the advantage of only depending on the mass difference between the species to be separated; it does not depend on the chemical properties of the ions being separated. However, operation of a mass analyzer at high mass resolving power typically costs an order of magnitude or more in analyte signal intensity. No such trade-off occurs in chemical resolution MS.

Chemical resolution MS is not a panacea, just as no other single analytical innovation solves every problem. However, it is extremely powerful and versatile. It capitalizes on the decades of work directed at achieving a better understanding of gas phase ion-molecule chemistry. The reaction rates for thousands of reactions are now available in an electronic database which greatly aids the design of chemical resolution MS separations.[1] For any given analyte/interfering ion pair of interest, the available reaction rates for various gases are compared to quickly identify a gas that reacts rapidly with the interferent, but not with the analyte. If no such reagent gas is listed in the database, a list of candidate gases can be quickly tested for their efficiency in effecting the desired reaction. A collision cell equipped ICP/MS is a nearly ideal instrument for this research purpose. Alternatively, reactions in which only the analyte reacts can be used, but these are generally a less effective approach since multiple reaction products are usually observed and the analyte signal is thus diluted over many mass channels.

Following our first reports on this work in 1996[2-4], over 200 publications and presentations have appeared capitalizing on this new approach. Nearly every ICP/MS manufacturer now offers a collision cell equipped instrument with which to carry out gas phase reactions for interference removal. Several chemical resolution MS Symposia have been organized: at the Winter Conference on Plasma Spectrochemistry as well as at recent meetings of the Federation of Analytical Chemistry and Spectroscopy Societies (FACSS). Now that commercial instruments are available, the number of application oriented publications is growing rapidly. We expect chemical resolution MS to find numerous applications in the elemental and isotopic analysis of biological materials since matrix effects and isobaric interferences can be severe with such complex samples.

While chemical resolution MS can be applied to both natural and non-natural isotopes, we have focused on examples where radioisotopes of interest are obscured by naturally occurring isotopes or by molecular isobaric interferences. We have previously demonstrated effective methods for achieving resolution of interferences in these systems: Sr-90/Y/Zr, Cs-135,137/Ba, Ca-40/Ar, Fe-56/ArO, Se-80/Ar_2, I-129/Xe.[4-6] Other groups have contributed significantly to this field by addressing other analyte/interference problems[7, 8] as well as by extending previous studies of collision cell operation and design.[9-11] In this paper, we will describe several of the above examples of past successes in our lab, briefly summarize results reported by other groups, and present new results for Se-79 (remove interferences by Br-79 and Ar_2H^+), Cs-135/137 isotope ratio measurements, as well as separations for other radioisotopes of interest.

References.

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