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Invited review

Great new insights from failed experiments, unanticipated results and embracing controversial observations

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ABSTRACT

Experimental data and observations, whether telescopic or analytical, are never wrong, though data derived from such sources can be misinterpreted or applied inappropriately to derive conclusions that are incorrect. Given that nature always behaves according to the laws of physics and chemistry, rather than according to currently popular models and theories, experimental results should always be considered correct even when the results are far from those that one might initially expect. We discuss a number of cases where the results of experiments, even one carried out as a simple calibration measure, produced wildly different results that generally required many years of effort or contemplation to understand. On the positive side, exploration of the circumstances that produced the "errant" results often led to new and interesting insights concerning processes that might occur in natural environments and that were well worth the effort involved.

Specifically, we show how an experiment that "failed" due to a broken conductor led to experiments that made the first refractory oxide solids containing mass independently fractionated oxygen isotopes and to 1998 predictions of the oxygen isotopic composition of the sun that were confirmed by the analysis of Genesis samples in 2011. We describe a calibration experiment that unexpectedly produced single magnetic domain iron particles. We discuss how tracking down a persistent source of "contamination" in experiments intended to produce amorphous iron and magnesium silicate smokes led to a series of studies on the synthesis of carbonaceous grain coatings that turn out to be very efficient Fischer–Tropsch catalysts and have great potential for trapping the planetary noble gases found in meteorites. We describe how models predicting the instability of silicate grains in circumstellar environments spurred new measurements of the vapor pressure of SiO partially based on previous experiments showing unexpected but systematic non-equilibrium behavior instead of the anticipated equilibrium products resembling meteoritic minerals. We trace the process that led from observations of the presence of crystalline minerals detected in the comae of some comets to the 1999 prediction of large-scale circulation of materials from the hot, innermost regions of the solar nebula out to the cold dark nebular environments where comets form. This large-scale circulation was ultimately confirmed by analyses of highly refractory Stardust samples collected from the Kuiper Belt Comet Wild 2. Finally we discuss a modern and still unresolved conflict between the assumptions built into three well known processes: the CO Self Shielding Model for mass independent isotopic fractionation of oxygen in solar system solids, rapid and thorough mixing within the solar nebula, and the efficient conversion of CO into organic coatings and volatiles on the surfaces of nebular grains via Fischer–Tropsch-type processes.

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Contents

1. Introduction

Most non-scientists and even many science graduate students and post doctoral researchers imagine that scientific progress stems from well-executed experiments that measure new data required for building models of natural phenomena or that can verify model predictions concerning the behavior of artificial analogs of such systems and processes. What many fail to appreciate is the sometimes stunning insights that can be gained when seemingly simple experiments do not produce the expected results. Indeed, some of our laboratory's most interesting findings have stemmed from experiments that produced results that were initially incomprehensible or that were performed for completely different reasons. In this article we will discuss experiments that yielded the first laboratory production of mass-independent oxygen isotopic fractionation in silicates, the first production of single domain iron grains condensed directly from the vapor phase, and the recognition that carbonaceous coatings deposited onto amorphous iron silicate condensates (smokes) are better Fischer–Tropsch-type catalysts than are the surfaces of the initial smokes. Each of the aforementioned results was totally unexpected and stemmed from experiments performed for completely different reasons. We will also show that similar insights can come from comparing published observations against the current "standard wisdom" and looking for alternative explanations for phenomena that do not easily fit within the currently popular paradigm.

Since all of the results discussed below have been published and are referenced in this review, we have elected to tell the stories behind these research results rather than to describe these findings in the more traditional style of a scientific review paper where much of the "back story" would be (and was) left out of the manuscript. By fully exposing our confusion and complete lack of understanding of some of the results we have obtained in the past, together with the new insights gained once those results were understood, we hope to encourage younger scientists to examine the potential wisdom that might be found in confusing or unexpected results from their own experiments or observations. There is a quote generally attributed to Isaac Asimov (and pointed out to us by our referee) that "the most exciting phrase a scientist can say is not 'Eureka' but 'that is odd…'", though the source and exact wording of the quotation remain elusive (according to both snopes and Quote Investigator). It is this point that we hope to illustrate in this manuscript, based on our own experiences.

The general focus of our work over several decades has been to understand the formation and subsequent evolution of solids condensed from a hot gas in astrophysical settings ranging from outflows around dying stars to the products of energetic events such as lightning, collisions or shocks that might have occurred during the early history of the solar system. In previous reviews (Nuth et al., 1998, 2002), we have discussed experimental results carried out using our "dust generator" that generally produced the materials that we expected to find as products. The exception was our observation that condensation of iron-magnesium silicates from the gas phase only occurs at metastable eutectic compositions; this was an unexpected result that we still do not really understand, but which has profound implications for modeling the condensation of silicates in both circumstellar outflows as well as in the primitive solar nebula (see Rietmeijer et al., 1999; Rietmeijer and Nuth, 2000). While we have identified the phenomenology of the effect and have proposed related experiments to confirm that other refractory chemical systems will preferably condense at such metastable eutectics, we cannot yet identify the kinetic mechanism that would prevent $(SiO)_x$ clusters containing a magnesium atom from accreting iron as it continues to grow or a similar $(SiO)_x$ cluster containing an iron atom from accreting available magnesium from the vapor. We do not consider such chemical mechanistic ignorance to be problematic: it is merely an opportunity for more research with the potential for additional unplanned surprises, much as was done in the situations that will be described below.

2. Mass independent oxygen isotopic fractionation

We have collaborated with the research group led by Dr. Mark Thiemens since the mid-1980s when Mark requested that we send him various types of vapor-phase condensates for oxygen isotopic analysis. Over the next 30 years we must have sent several hundred samples to his laboratory, ranging from smokes that we produced in the course of routine experiments totally unrelated to the oxygen isotopic composition of the samples, to samples from experiments where we tried to induce fractionation based on specific hypotheses about how such processes might operate in a nebular environment. None of the simple samples that we produced in our other experiments ever showed evidence for mass independent fractionation and none of the specific experimental systems that we built and operated in attempts to produce such fractionated condensates ever yielded mass independently fractionated oxygen. Then one day our flow system broke during an experiment and we got a hint for how such samples might be produced.

In 1986 we began using a new flow system to produce larger, more uniform quantities of amorphous iron and magnesium silicate smokes. By flowing various mixtures of silane and pentacarbonyl iron in a large excess of hydrogen through a furnace where the input stream was mixed with oxygen or nitrous oxide to produce a high temperature flame and magnesium vapor was introduced to the gas from a graphite crucible in the hot furnace, we could produce gram-level quantities of highly amorphous condensates with average grain size ~10 nm in radius. The flame brought the temperature of the gas mixture above 1500 K at the flame front where the silane was converted to SiO and the iron carbonyl decomposed to produce iron vapor. This mix of iron and magnesium atoms, SiO, CO, OH and H_2O, still in a large excess of H_2, cooled rapidly and nucleated even before exiting the furnace to make very fine grained and highly disordered condensates (smokes) that were then used in subsequent experiments or studied for their own sake to understand the condensation process (e.g., see Nuth et al., 2002 for a review).