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Light scattering studies of randomly oriented polycrystalline fayalite micro particles as interstellar dust analogues

Manash J. Boruah [a,*], Ankur Gogoi [b,c], Bikash C. Nath [a], Gazi A. Ahmed [a]

[a] Department of Physics, Tezpur University, Tezpur 784028, Assam, India
[b] Department of Physics, Jagannath Barooah College, Jorhat 785001, Assam, India
[c] Institute of Biophotonics, National Yang Ming University, Taipei 11221, Taiwan

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ABSTRACT

Fayalite (Fe_2SiO_4), the iron end-member of the olivine group, is found in various extraterrestrial environments including the interstellar medium and meteorite. Since the iron rich silicates, i.e. fayalites, are not abundantly found in earth, there has been lack of sufficient experimentation and modeling leading to the unavailability of sufficient experimental data of fayalite for comparative analyses with computations and astrophysical observations. In this work interstellar fayalite dust analogues were synthesized in the laboratory using simple chemical route. Shape and size dispersed interstellar dust analogue models for laboratory synthesized fayalite particles were developed for performing theoretical computations of light scattering parameters (e.g., angular profiles of intensity and degree of linear polarization, geometric and single-scattering albedo, asymmetry parameter and cross-sections of extinction and absorption) using discrete dipole approximation (DDA). In order to demonstrate the validity of our models, phase function and degree of linear polarization were measured using a laboratory based setup and the results were compared with DDA computed theoretical values at three wavelengths 543.5 nm, 594.5 nm and 632.5 nm respectively.

1. Introduction

Computational light scattering studies of interstellar dust particles supported by laboratory simulations and experiments with analogue samples provide significant contribution towards understanding the unknown dust grains present in different astrophysical environments. Observations, e.g., interstellar extinction and polarization, absorption with the Hubble space telescope etc., indicated that these dust particles are mainly composed of amorphous silicate, graphite, amorphous carbon, carbonates, metal oxide grains, amorphous ice particles and nanodiamonds in the interstellar medium [1,2]. Most of the studies of interstellar and cosmic dust confirmed that the majority of interplanetary particles have highly irregular and complex shapes [3,4]. Computationally dealing with such complex and highly irregular shaped particles is a difficult problem due to the lack of adequate information about the shape, size, roughness, porosity and internal structures of those particles. Realistic modeling of such type of particles requires visual evidence which is very difficult to acquire with a few exceptions like the STARDUST mission [5]. The best approach is to try different advanced and efficient algorithms to model the irregularly shaped particles having a vast range of shapes and sizes supposed to constitute the complex dust particles. Recently, interstellar dust models have been extensively studied by various research groups with carbonaceous dust (graphite and amorphous carbon particles), silicates (olivine, fayalite etc.), polycyclic aromatic hydrocarbons (PAHs), etc. [6–8]. Despite the numerous computational, observational and experimental studies, the physical and optical properties of dust particles are not properly known till date [9,10]. Notably, the optical properties (e.g. absorption and scattering characteristics) of interstellar dust carries important information that is necessary to deduce the compositional, structural and chemical properties of their constituent particles which may in turn lead to a better understanding of the evolution of various astrophysical processes including the formation of the dust itself.

Silicates are among the most widely studied dust species due to their ubiquity in many different astrophysical environments including the solar system, interstellar dust clouds, intergalactic dust clouds, circum-planetary dust rings, cometary comae and tail, asteroidal atmospheres and aerosols of other planetary atmospheres [11–14]. Importantly, depending on their local environment, silicates possess different chemical composition and crystal structure. There have been numerous evidences for detection of silicate

* Corresponding author.
E-mail address: boruahmanash5@gmail.com (M.J. Boruah).

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species of dust both in amorphous and crystalline form (not negligible) with variable iron and magnesium contents in different ratios and compositions [15–18]. It is also reported that the observed spectra of interstellar dust could be better explained if one consider both amorphous and crystalline silicates to be present in the intervening dust [18]. Prominent mid- and far-infrared emission and absorption features observed by the Short Wavelength Spectrometer (SWS) of the Infrared Space Observatory (ISO) suggested the presence of olivine and pyroxene as two major crystalline silicate components of cometary, circumstellar and planetary dust. In addition, in situ data from the Cosmic Dust Analyzer (CDA) on board the Cassini spacecraft also revealed that the interstellar dust, as it passed by Saturn, is mainly composed of magnesium-rich grains of silicate and oxide composition, partly with iron inclusions [19].

Crystalline olivine is an important component of silicates in space and cover the range of isomorphous magnesium iron silicates having the general formula $Mg_{2x}Fe_{2-2x}Si0_4$ with $1 < x < 0$. Fayalite, Fe_2Si0_4 ($x=0$), is the iron end-member and forsterite, Mg_2Si0_4 ($x=l$), the magnesium end-member of the olivine group [20]. In this paper we concentrate on the iron rich species of olivine (fayalite) which is mostly found in interstellar medium and meteorites [21–23]. Although magnesium rich olivines and pyroxenes (forsterite, enstatite etc.) are studied extensively in various dust models [24–27], there has been lack of sufficient experimentation and modeling in case of iron rich silicates (fayalite). Importantly, the composition of the terrestrial silicates are mainly magnesium rich while iron rich silicates are not abundantly found in earth, due to which it is very difficult to conduct laboratory experiments on realistic analogue samples of iron silicates to simulate the physical and optical properties of extraterrestrial dust (which are extremely complex in nature and morphology). Not much available experimental data of fayalite is found for comparative analyses with computations and astrophysical observations despite its presence in several extraterrestrial environments.

The aim of this paper is to simulate the light scattering parameters of interstellar polycrystalline fayalite dust analogues using both experimental and computational techniques. The overall goals of this work were to,

- synthesize interstellar fayalite dust analogues in the laboratory using simple chemical route,
- characterize the morphology and composition of the materials using scanning electron microscopy (SEM), energy dispersive X-ray analysis (EDX) and X-ray diffraction (XRD) analysis (see section 2),
- design irregular shape and size dispersed interstellar dust analogue models for laboratory synthesized fayalite particles using BLENDER3D (see section 3),
- perform theoretical computation of light scattering parameters (e.g., angular profiles of intensity and degree of linear polarization, geometric and single-scattering albedo, asymmetry parameter and cross-sections of extinction and absorption) using the discrete dipole approximation verify the validity of our designed models by conducting laboratory based experiments using incident laser light sources of wavelengths 543.5 nm, 594.5 nm and 632.8 nm.

2. Synthesis and characterization of fayalite samples

2.1. Synthesis of fayalite

The availability of fayalite is relatively sparse in nature and therefore it is very important to synthesize such material in varying shapes and sizes as per the requirements for experimental studies [28]. For astrophysical dust studies, grains of sizes ranging from nanometer to a few micrometers are often required. To synthesize nanocrystalline fayalite (nanofayalite), a sol-gel technique originally described by De Angelis is adopted [29].

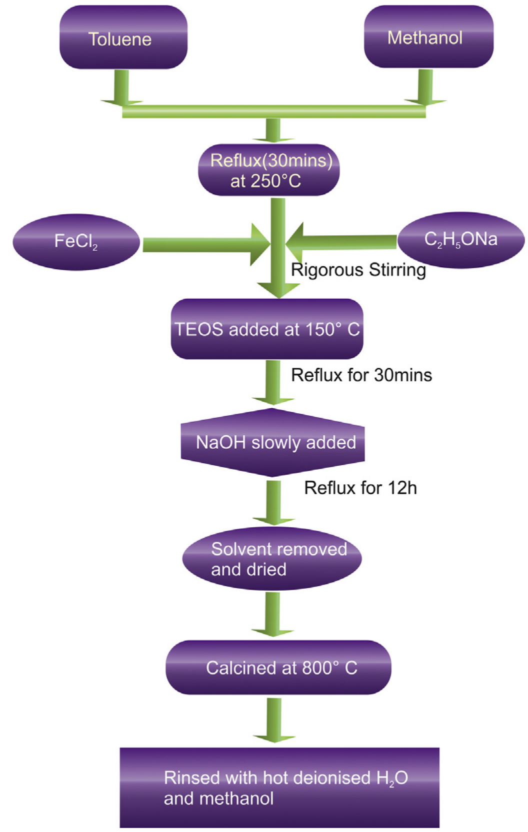

Fig. 1. Flowchart for sol-gel synthesis of fayalite.

In this method, Iron (II) chloride, sodium ethoxide, and tetraethyl orthosilicate (TEOS) were added in a sol-gel process to produce a precursor gel, which was subsequently calcined under reducing conditions to crystallize nanofayalite. X-ray diffraction (XRD) analyses indicate that the produced nanofayalite is nearly pure, with minor amounts of metallic Fe in some batches. The flowchart of sample preparation is shown in Fig. 1.

First 375 mL of toluene and 175 mL of methanol were added in a three neck round bottom flask. The solvents were refluxed to remove any dissolved oxygen at 250 ° C for about 30 min and then allowed to cool to room temperature. After that, 8.7 g of iron (II) chloride ($FeCl_2$) followed by 9.3 g of sodium ethoxide (C_2H_5ONa) was added while accompanied by rigorous stirring. The sodium ethoxide rapidly reacted with the iron (II) chloride resulting in a change in the solution color from being nearly-translucent light to an opaque, grayish-green color. Next, the temperature was raised upto 150 °C and at the same time 7.9 g of Tetraethyl orthosilicate (TEOS) was added. The solution was continuously refluxed for nearly 20–30 min, and 10 mL solution of 0.2 M NaOH was slowly dripped with a syringe to hydrolyze the iron(II) ethoxide and TEOS. The solution was then allowed to reflux for another 12 h. After reaction, heating was turned off and the solution was cooled down. The rotary evaporator was used for solvent removal and drying. The dried powder was calcined in a tube furnace under flowing N_2 gas for 4 h at 800 °C. After calcination, the nanofayalite crystals were rinsed using centrifuge process repeatedly with hot deionized H_2O to remove NaCl. After removal of all of the NaCl, the fayalite crystals were rinsed with methanol and dried with a rotary evaporator [29].