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Standardization of an external (in air) PIGE methodology using tantalum as a current normalizer in conjunction with INAA for rapid and non-destructive chemical characterization of "as-received" glass fragments towards forensic applications

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An external (in air) Particle Induced Gamma-ray Emission (PIGE) method using tantalum (window material) as the external current normalizer was standardized for the first time for rapid compositional characterization of "as-received" sodalime (automobile) and borosilicate glass samples. It involves irradiation of glass fragments in air using a 3.5 MeV proton beam from the FOlded Tandem Ion Accelerator (FOTIA) and simultaneous measurement of prompt gamma-rays from proton induced reactions of isotopes of low Z elements (Si, Na, Al and/or Mg and B) and 135 or 165 keV from tantalum window (^{181}Ta). The results of external PIGE were compared with those obtained by conventional (vacuum chamber) *in situ* current normalized PIGE by analyzing sample pellets in a cellulose matrix. External and conventional PIGE methods were validated by analyzing both sodalime and borosilicate glass certified reference materials. The external PIGE method is found to be a rapid non-destructive method for discriminating sodalime and borosilicate glasses both qualitatively and quantitatively through their composition, which is important for forensic applications. Among the major elements determined by external PIGE, aluminium was found to be the only marker element for distinguishing sodalime (automobile) glasses. In order to find more marker elements for a grouping study as well as for forensic applications of "as-received" sodalime/automobile glass samples, Instrumental Neutron Activation Analysis (INAA) using high reactor neutron flux (~10^{13} n per cm^2 per s) from the Dhruva research reactor was utilized for bulk analysis to quantify thirteen elements (Na, Mg, Al, Ca, Mn, Fe, Sc, Co, Hf, La, Ce, Sm and Eu) at major to trace concentration levels. Samples were irradiated utilizing the Pneumatic Carrier Facility (PCF) as well as the self-serve facility of Dhruva reactor for short duration irradiation followed by high resolution gamma-ray spectrometry to obtain the elemental concentrations of interest including transition elements and rare earth elements (REEs). Preliminary grouping of sodalime/automobile glasses was carried out using the concentrations of marker elements (Al, Sc, Fe, Co, Hf, La and Ce) and concentration ratios like Al/Sc, La/Sc and Ce/Sc and total REEs, and all results indicated that the glasses fall into three groups. The grouping was confirmed by statistical cluster analysis (*i.e.* tree dendrogram) utilizing concentrations of transition elements and REEs, which indicated the potential of elemental concentrations particularly at minor and trace concentration levels for forensic applications.

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Introduction

Forensic science and its application often depend on authentic chemical characterization of diverse samples under investigation at major, minor and trace elemental concentration levels. Among the various objects under investigation by forensic scientists, glass objects are often studied due to their authentic as well as reliable forensic evidence because of their inimitable chemical and physical properties.[1,2] In earlier days *i.e.*, in the BC's these types of study were often carried out by archeologists in order to establish the trade pattern of glasses (glass beads or ornaments) through their chemical composition.[3] Glass traces/fragments obtained from the crime scene are important forensic evidence to relate the chronology of the

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