From the Director’s Desk

Technical Articles
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* Confocal microscopy – A new tool in Soft condensed matter

AWARDS & HONOURS

The Story of Fast Breeder Test Reactor

As Fast Breeder Test Reactor (FBTR) enters the 20th year of its successful operation on 18th October 2004, it is appropriate to recapitulate the history of this flagship of our centre. FBTR had its humble beginnings in 1968 in the Fast Reactor section of Reactor Engineering Division, Bhabha Atomic Research Centre (BARC). Memories of this section are even today found as rubber stamp marks in some of the books in our Centre’s library. It was due to the vision of Dr. Vikram Sarabhai that a decision was taken to establish an exclusive centre named Reactor Research Centre (RRC), subsequently renamed as IGCAR, dedicated to the pursuit of fast reactor Science & Technology. A sodium cooled fast test reactor (originally christened Indian Fast Test Breeder Reactor – IFTBR) was to be the hub of this centre, to serve as a test bed for irradiation of fuels and materials and to gain experience in the handling of large volumes of sodium and operation of a fast reactor. The excellent rapport between the French and Indian governments resulted in signing of a MOU between DAE and French CEA for (a) transfer of the design and drawings of Rapsodie fast reactor (b) training of Indian team in design and operation and (c) transfer of manufacturing technology of critical components.

The scientist must be free to think and put forward whatever ideas he considers right
- Dr. Homi Bhabha

INDIRA GANDHI CENTRE FOR ATOMIC RESEARCH
Kalpakkam, located between Mahabalipuram, the historic port city and once the capital of the Pallava kings, and Sadurangappattinam (shortened as Sadas), once a port city and stronghold of Dutch in India was chosen to be the place for setting up of RRC. Between these two historic places, history was again being created by Department of Atomic Energy in the first ever massive indigenous effort to build two Pressurised Heavy Water Reactors, christened Madras Atomic Power Project (MAPP). The availability of land within the exclusion zone of MAPP, low population density, assurance of water supply from Palar river, low seismicity and proximity to MAPP which would facilitate sharing of infrastructural facilities were the major reasons for the choice of Kalpakkam as the site for FBTR.

The design team went to France in 1959. The team included reactor physicists, engineers & draughtsmen. This was followed by members from operations and construction. But for their theoretical background and passion for success, the design team had not much of practical experience - no sodium loop experience, no construction or operating experience; probably they had seen sodium as sticks boiled up under kerosene in the laboratories. The first batch of engineers from BARC Training School came to Kalpakkam in June 1971. They had their offices in Engineering Hall I, which was the only building existing at that time. Central Design Office (CDO) was just being built. A thatched shed under the coconut trees near the present CDO was their canteen. Wooden planks nailed to stumps of casuarina and palm trees acted as dining furniture. Shri N. Swaminathan Iyer, Administrative Officer, who was the Chairman of Canteen Committee used to serve tea and snacks himself to the Scientists and Engineers, setting an example of teamwork. With their memories of BARC still fresh in mind, some of the officers were cherishing a fond hope of going back to Bombay (now Mumbai) six or seven years later, after building FBTR. They came to build FBTR, but most of them remained at Kalpakkam, spending the rest of their careers in designing & mastering the challenging fast reactor technology for subsequent development of commercial Prototype Fast Breeder Reactor (PFBR).

The project was steered by Shri N. Srinivasan, Project Director, RRC with Shri. S.R. Paranipe as the Principal Design Engineer and Shri N.L. Char as the Principal Project Engineer. Shri C.R. Ramamoorthy, Chief Engineer (civil) was looking after all civil activities including MAPP, township and RRC. Shri R.P. Nagaraj, reporting to him, was Project Engineer (civil). S/s D.S. Ahluwalia, N.M. Venkataraman, A. Venkateswarlu, M.C. Sabherwal, K. Raghavan, R. Jeevanandam, A.D. Raghuvir, A.T. Vasudevan and R. Chandramohan played key roles in the project construction, as also many others who are still in service. Shri G. Mallikarjunan headed the Quality Control & Inspection team. Difficulties in the manufacture of reactor assembly components resulted in the formation of a special Task Force for Block Pile, with engineers drawn from design and procurement wings. Shri S.B. Bhoge was the convenor of the Task Force. French engineers also had periodic stints at site as consultants. They include Mr. Gourievidis, Chouard, Pacton, Paziaud, Mesnage and Alari. The excellent training manuals of FBTR owe their credit to Mr. Mesnage and Mr. V.S. Justin. A Technical Liaison Mission was also set up in Paris, with Shri T.K.Ghosh as the Liaison Officer (followed by Shri P.G.S. Mony and later by Shri. K.C. Khandelwal). Dr. Placid Rodriguez, who in later years steered FBTR through several milestones as Director, IGCAR, was constantly guiding the construction team in the welding metallurgy of stainless steels & high alloy steels, choice of filler metals to the stringent FBTR specifications and heat treatment cycles for various components, along with his team of young and dedicated scientists and metallurgists.

When excavation for the project started, the French consultants
the project would be completed in a lifetime. "Now we know why you believe in God!" was their comment. Little did they realise that all the huge temple structures that dot the landscape of this ancient nation were built by men and not by machines. A small temple which fell within the original layout of FBTR had to be spared, respecting the sentiments of the local labourers, who were not willing to pull it down. It still exists within the premises of FBTR.

Though adapted from Rapsodie, FBTR had several design modifications. The steel containment of Rapsodie was replaced with RCC structure. A steam-water circuit having four once-through steam generator modules of Phenix type design was added along with turbine-generator and dump condenser. Though the motivation for this decision probably came from the energy crunch due to the middle-east oil crisis of seventies, in retrospect this is found to be a good decision, since steam generator is a critical component of any large fast reactor and without any experience in that front, there can be no mastery of the fast reactor technology. There were also several design changes in the reactor assembly and spent fuel storage. The unique small capacity Turbine Generator (16.4 MWe) employing high pressure and high temperature steam (125 bars & 753K) was designed and built by BHEL, Bhopal.

In line with the Department’s policy of self-reliance, the accent was on total indigenisation. With an indigenous component of more than 80 %, FBTR is in fact a symbol of our self reliance and indigenous capability. Except the raw materials for Nuclear Steam Supply System (NSSS), borated graphite, one control rod drive mechanism, one sodium pump, grid plate, bellow-sealed valves and sodium instrumentation, all other components were made in India. Indian industry in the seventies was just coming to terms with ASME Section III. FBTR standards being more stringent, there was reluctance on the part of many industries to undertake manufacture of FBTR components. Most of the major components were entrusted to public sector undertakings by persuasion. The steel vessel of reactor assembly was fabricated at Central Workshop (CWS), IGCAR, under the guidance of S/s B.S.Iyengar, N.Gunasekaran, R. Deivasigamani and K.S.Krishnamoorthy. Shri. Adimoolam of CWS was the first welder to qualify as per FBTR standards, followed by Shri. Kaliyamoorthy and others in FBTR.

Material and Personnel Airlocks were fabricated at Central Workshops, BARC. The rotary crane of Reactor Containment Building, first of its kind in India, was erected before construction of the dome. The China mosaic on the dome, for heat shielding, was the first of its kind.

Unlike pressure vessels, fast reactor components are thin walled, slender and long, with stringent shape tolerances. A classic characteristic of austenitic stainless steel during welding, viz. distortion, assumed almost notorious proportions due to the slender nature of components. Control of local and global distortions was a major agenda for the fabricator. Unlike conventional systems, fast reactor circuits cannot be flushed with water during commissioning. Hence, cleanliness during manufacture had to be strictly enforced. Most of the shop floors had to be equipped with nuclear clean halls. During site welding, the purge gas dams had to be meticulously numbered and inventory maintained, to ensure that nothing is left inside the systems. The arrival at site of the primary
sodium storage tank (which was required to be installed in the B4 cell of the reactor building, before casting its roof slab) with a cracked nozzle weld was an eye-opener to the team that due care should be taken during transportation. It was all a phase of collective learning for the industries as well as our engineers. Safety Evaluation Working Group (SEWG), with Dr. D. V. Gopinath as chairman and Dr. L. V. Krishnan as member-secretary, critically reviewed the design and safety of the reactor. Many of the technical decisions, which were at variance with Rapsodie were discussed and debated in depth before arriving at a consensus. The project progress was meticulously monitored and reviewed every month in Project Progress Review Meetings.

The psychological backdrop under which the project team had to work was no less daunting. The embargo in the wake of the Peaceful Nuclear Explosion in 1974 was casting a shadow on the fuel supply. It was in this context that a bold decision was taken to design, develop and fabricate indigenously the U-Pu Carbide fuel. Dr. Raja Ramanna, who was Chairman of the Council of the Centre in the early years steered the activities with remarkable vision. He along with late Shri P.R. Roy and Shri S.R. Panje, can be credited with the bold and visionary science-based technological decision to choose mixed carbide of unique composition as driver fuel for FBTR. When most of the reactor components had been manufactured, the news of sodium leak in Rapsodie and the decision to decommission the Rapsodie reactor were known. This called for several last minute design modifications in the primary sodium piping, warranting revision of the overall schedule for the completion of the construction. With the inspiration provided by Shri. N. Srinivasan, Shri. N.L. Char and later by Shri. C.V. Sundaram, the project team weathered all the storms with tremendous resilience to take the project to completion by 1984.

All the critical components were transported from the manufacturers’ works by departmental trailers, following the route-maps and security restrictions, which were personally decided by Shri. N.L. Char and speed limitations decided by the design group. Many components of over-dimensional nature (ODC) had adventurous journeys, as would be recounted for posterity by the transport crew comprising S/s T. Sampath, (Truck operator), P.B. Kunj Ahmed, L. Sebastian and K.Balan (riggers). The site erection of all the components, to the specified stringent positional accuracies was carried out by these riggers with dexterity under the supervision of Shri. P. Vasu and overall guidance of Shri. A.T. Vasudevan.

Commissioning of the systems was done in stages, as and when the components arrived under the guidance of S/s. M.K. Ramamoorthy and K.R. Karanth. Shri. R. Subramanian, Head, Reactor Chemistry Section, designed and erected a rig for purifying commercial grade sodium. When there was apprehension about the supply of NaK, he rose to the occasion by producing NaK required for one of the cold traps in the glove boxes of Radio Chemistry Laboratory (RCL), with no tools other than a few stainless steel kitchen ware cash-purchased from the local shops. Purified sodium was transported to FBTR in a specially improvised tanker and charged into the storage tanks in 1984. It was a credit to the meticulous care exercised by the Quality Control Wing in maintaining cleanliness so that uninterrupted sodium flow could be
established immediately in all the loops, with sodium purity unimpaired. Shri. V.A. Pethe, Head, Electronics Division, BARC and Shri. S.N. Seshadri, Head, RCnD, BARC were present during the commissioning of nuclear instrumentation and sodium pumps.

All the hardware required for the core were manufactured by Nuclear Fuel Complex (NFC) under the guidance of Shri. B.P. Varma. Fuel pins of MK-1 composition were fabricated by RMD, BARC, under the guidance of Dr. P. R. Roy and Dr. C. Ganguly. Quality surveillance for the fuel pins as well as for all the subassemblies was ably provided by Atomic Fuels Division, BARC, under the guidance of Shri. K. Balaramamoorthy. Fuel pins were assembled into subassemblies at FBTR by a team from NFC.

Loading of the non-fissile subassemblies into the reactor was done manually. The count-down for criticality began with the start of loading of fuel subassemblies on 2nd October, 1985. FBTR went critical on 18th October, 1985, with a small carbide core rated for 10.5 MWt. To Shri R. Shankar Singh and Dr. S.M. Lee, it was a moment of relief and satisfaction that the reactor went critical with 22 subassemblies, as predicted by reactor physics calculations. It was the joyous culmination of the sweat and toil of thousands of engineers and workers spread all over India.

Since the steam generators were not yet ready, reactor was operated at low power with the Steam Generator bypassed. Low power experiments were conducted during this period. The fuel handling incident in May 1987 was a setback, but under the sustained initiatives...
taken by Shri C. C. Sundaram and Dr. D. V. Gopinath, the reactor was back on stream in May 1989. The intervening time was also gainfully utilised for connecting the steam generators to the secondary sodium circuits and commissioning the steam generator leak detection system. Power was raised in a phased manner to 1 MW (in 1991), then to 4 MW(t) and subsequently to 8 MW (in 1993). After completing high power physics and engineering experiments, power was raised to 11.5 MW and TG was synchronised to the grid in July 97. The reactor power has been progressively increased, reaching the highest power of 17.4 MWt in 2002. Eleven irradiation campaigns have so far been completed, with the plant availability in the last two campaigns exceeding 80%. The reactor achieved a significant landmark with the fuel reaching a burn-up of 100 GWd/t in September 2002. BFTR was used as an irradiation facility for assessing the irradiation creep of Zr-Nb alloy being deployed in Pressurised Heavy Water Reactor. Presently, test fuel simulating PFBR MOX composition is under irradiation, with indigenous new generation clad and wrapper.

Hitting the headlines
The news of BFTR first criticality was widely acclaimed. Selected clippings from the first page of THE HINDU on 19.10.1985

All eyes on Kalpakkam
From G. K. Reddy
NEW DELHI Oct 18
After the巴拉克 registration, the first single event in India's nuclear development has achieved such great distinction in the foreign diplomatic and due to the successful fuel cycle that is being carried out by the country's atomic scientists. The presence of the fast breeder reactor in Kalpakkam, with the technical team of various nations, has been seeking the depart to the departmental technical direction about the remarkable achievement by developing nation, while the political experts and the country's atomic scientists are the various accomplishments of this spectacular breakthrough by India in nuclear development.

Ultimate intonation: What is considered really creditable is that the Indian scientists have succeeded in developing an indigenous fuel cycle with a plutonium and uranium core instead of enriched uranium which India has proposed to obtain from France on an arrangement for the experimental fast breeder project where the ultimate intonation is to be obtained by the FFTR to validate the data assumed and the codes used in the design and safety analysis of the reactor. The maximum annual activity released to atmosphere so far is a miniscule fraction of the permitted release. Cumulative occupational exposure so far is only 50 man-MSVs (man-rem). During the past 19 years, there has been no significant event of abnormal radioactivity release,
personnel or area contamination or personal exposure, thus confirming that sodium cooled reactors are ecologically way ahead of other types of reactors.

Unlike other reactors, which have fixed core configurations, targets and missions, FBTR has an evolving core, with a novel driver fuel. This calls for frequent reviews by the safety authorities, sometimes at short notice. The major milestones achieved by FBTR would not have been possible, but for the enlightened guidance and stage-wise safety clearances accorded by Safety Committee (SC), Safety Review Committee for Operating Plants (SARCOP) and Atomic Energy Regulatory Board (AERB). The Technical Specification Document, the book of do’s and don’ts for the operator, was reviewed by SARCOP under the chairmanship of Shri M.S.R. Sharma in several extended sessions. While being generally conscious of the difficulties of a new technology, SC, SARCOP and AERB have been exercising constant vigil over the safety of the reactor. There have in fact been a few occasions when they have clamped on reactor operation pending further investigations.

The road to success is not always smooth. True to its name as a test reactor, FBTR has given testing times also. There have been several major and minor incidents, from each of which we have returned wiser and stronger, and some have provided developmental spin-offs. The fuel handling incident led to the development of novel inspection techniques including air ultrasonics and under-sodium ultrasonics. The cutting tool developed by Central Work Shop (CWS), BARC, for cutting the guide tube, was a master-piece of mechanical design and its modified version is presently being deployed for cutting the coolant channels of PHWR for coolant channel replacement. The leaks in biological shield cooling coils, though chemically sealed by professionals, have led to a programme of indigenous development of chemical sealants. The seizure of main boiler feed pump has led to an indigenous pump of sturdy design. Core
temperature anomalies arising out of the stuck core cover plate mechanism led to the development of eddy current sodium flow meter for measuring flow through the subassemblies. The sodium leak incident gave us the confidence in handling radioactive sodium with minimum exposure or contamination and methods of disposal. More than 400 modifications have been carried out in FBTR, which have improved the safety and availability of the reactor.

The first reactor superintendent was Shri. M.K. Ramamurthy, with Shri. K.R.Karanth as the Operations Superintendent and Shri. P.R.Venugopal as Maintenance Superintendent. The reactor was later headed by Shri. S.B. Bhoje and then by Shri. R.P. Kapoor. Dr.V.M.Raghunath was the first health physicist of FBTR and his long stint as Member-Secretary of the IGCAR-CWMF safety committee will be ever remembered. He was conversant with all the safety related issues of FBTR, and would meticulously follow up the compliance to all regulatory stipulations.

All the directors of IGCAR have played major roles in steering FBTR through its challenges and milestones. Shri.N.Srinivasan, with his simple and unassuming but decisive and firm personality, played the key role during the construction phase, reviewing the cost and schedule and constantly motivating the team of young engineers by personal interactions in times of difficulties. The sight of Shri.N. Srinivasan, along with Shri.N.L.Char, taking their daily rounds of the project site, with safety helmets perched on their heads, is still cherished in the memory of senior personnel of the centre. Balance construction activities, commissioning of systems and first criticality were during the stewardship of Shri.C.V.Sundaram. He used to review the progress of commissioning almost on a daily basis, personally interacting with the commissioning group and inspiring them by his presence during every activity. His serene words of wisdom remain fresh in the minds of all who moved with him. During times of crisis, like the fuel handling incident, he used all resources at his command to resolve the problems. Shri.S.R.Paranjpe, fondly remembered as the father of fast reactor programme in India, was responsible for almost all the major technical decisions. Whenever there was a technical issue to be resolved, he used to perform hand calculations or mental arithmetic and give his spot verdict. However, Shri. S.R. Paranjpe insisted on further detailed and comprehensive analysis from all others associated with the design and development. Almost all safety clearances required for the plant were due to his genius. Dr. P. Rodriguez, who ushered in an era of confidence and high autonomy, fostered the managerial skills of this centre. He reposed full confidence in people and allowed them to naturally bloom to their levels of excellence, thus enhancing the productivity and successes. During his tenure FBTR saw all its major landmarks—vizz. nuclear steam generation, realising the target power of 10.5 MWe for the small carbide core, rolling and synchronisation of TG, MK-I fuel reaching a burn-up of 50 GWd/t and use of FBTR as an irradiation facility for the Zr-Nb alloy irradiation programme. Shri.S.B.Bhoje, who succeeded him is known for his tenacity of purpose, focus and emphasis on documentation. It was during his tenure that the reactor reached the highest power level of 17.4 MWe with MK-I operating at its design LHR of 400 W/cm and a record fuel burn-up of 123 GWd/t. PFBR test fuel using U^{235} for enrichment purpose was fabricated and loaded in FBTR during his tenure. Based on the successful performance of FBTR and the expertise developed over the years in the Centre, the Government had accorded its approval for the commercial phase of fast reactor programme in terms of the construction of 500 MWe Prototype Fast Breeder Reactor (PFBR) at Kalpakkam in September 2003.

Each of the successive heads had left his mark in terms of different milestones and leadership qualities. Thanks to them, FBTR has evolved a culture of well written operating procedures, respect for safe operating practices, adherence to specifications, meticulous planning, adherence to schedules, excellent documentation and system for introspection and collective leadership. When by the turn of this century a chain of fast reactors will be feeding power to the Indian grid, FBTR will no doubt be remembered as the mother of fast reactors in India.

[Baldev Raj]
Director
A Tribute

It is with profound grief and deep sorrow that we came to know on 24th September 2004 that Dr. Raja Ramanna, former Chairman, Atomic Energy Commission is no more amidst us to guide, nourish and motivate. Dr. Ramanna was an eminent nuclear physicist and an institution builder, par excellence. He played a colossal role in building DAE to an organisation of high national relevance and international repute. His science policies were directed towards encouraging creativity in basic research, leading to advances in innovative engineering and technology. He stood firmly for indigenisation and self-reliance in nuclear technology. The BARC Training School programme, which generated a wealth of trained human resources for various units of DAE, was his passion and pride. He was the father of Pokharan peaceful nuclear tests in 1974.

He along with Dr. Vikram Sarabhai sowed the seeds for the creation of Reactor Research Centre with a mission of developing fast reactor science based technology. The growth of our Centre is a standing tribute to Dr. Ramanna’s vision.

Dr. Ramanna, as the first Chairman of the Council of our Centre, was instrumental in directing and guiding the development of the unique carbide fuel for FBTR in the face of embargos after Pokhran tests in 1974. This fuel has seen, so far a record burn-up of 130,000 MWD/t and is still going strong. The first criticality of FBTR was achieved during his tenure as Chairman, AEC and he was personally present in the FBTR control room during that historic moment. It was, at his initiative that our Centre (then called Reactor Research Centre) was renamed as Indira Gandhi Centre for Atomic Research by Shri Rajiv Gandhi, former Prime Minister of India, in December 1985.

Apart from his love for physics and mathematics, his other interests included philosophy and spirituality. He had passion for music and was himself an accomplished piano player. He was intensely humane and exhibited fatherly warmth and affection for the DAE fraternity and he was a source of great inspiration to Scientists and Engineers. His incisive remarks and humor are remembered by many of us, who had the opportunity to discuss with him.

He always seemed to have more than an answer to any challenging situation. However, he left the final choice to the concerned individuals for implementing the technology and creating new science, respectively. We would always miss the gentle fatherly figure that Dr. Ramanna was.

We pray God to give strength to the family to bear this major loss. Indeed, his family is very large, which includes all the personnel in DAE and many other associates with whom he came in contact during his life time in India and abroad. He was a proud son of India. IGCAR salutes Dr. Raja Ramanna and we realise that the best way to honour his memory is to rededicate ourselves to his ideals and vision.

Boldev Raj
(On behalf of IGCAR)

(1925 - 2004)

“...It is inevitable for a country of India’s size and resource base to consider nuclear power as essential for meeting the energy requirements of our expanding industry and economy... India has the industrial and technological base to sustain and support the entire fuel cycle activities needed for all peaceful uses of atomic energy...”
FBTR to go on Power with modified condensate system

Steam and water system forms the tertiary loop of the heat transport system of FBTR. It consists of three sub systems viz; condensate system, feed water system and steam system. In the old condensate system there were three direct contact type heaters viz; LP heater #1 (LPH #1), LP heater #2 (LPH #2) and deaerator with its integral hot well (Fig.1). Since free water level is maintained in LP heaters, pumps were provided at the outlet of these capacities to transfer water from one capacity to another. Level in the LP heaters was maintained by controlling the outflow of water from the heaters with the level control valve of each capacity. All the pumps viz; condensate extraction pump (CEP), condensate booster pump (CBP) and deaerator lift pump (DLP) had trips on low level in its suction capacity and high level in its discharge capacity.

The free hot well level of the capacities were very sensitive to mild process / power supply disturbances. These level fluctuations led to CEP trip and consequent reactor trip (on an average 3-4 reactor trips per year). Hence it was decided to replace the direct contact type heaters LPH #1&2 with surface type heaters.

Fig. 2 depicts the modified condensate system. In this system, CEP takes suction from main condenser/dump condenser (MC/DC) and discharges condensate to DLP suction. DLP discharges water into deaerator through LPH #1 and LPH #2. The new LP heaters are shell and tube type heat exchangers with condensate in tube side and steam in the shell side. Since there is no free water level, level control circuits of LPH #1 & 2 and CBP were dispensed with, thus simplifying the system. By the use of surface type heaters and necessary modifications in the logic circuitry of the pumps, the system is made more rugged and immune to process disturbances.

Retrofitting of the new heaters (fig.3) in place of old ones had many technical challenges. New heaters had to be designed to have size and shape similar to the old ones to place them on the same bed. Dismantling of old circuit was done in such a way to enable the replacement of the heaters with minimum modifications. This in turn saved the cost and time. Also utmost care was taken in the layout of equipment, valves etc., so that entire operation of the system can be done from turbine building ground floor. During erection many modifications in piping layout had to be carried out to suit the site conditions as the building was already occupied with many other pipelines and equipment. After careful study of new system and analysing expected operational requirements, transients and process disturbances, some more modifications viz; a bypass line across DLP to operate the system with CEP alone in the shutdown state of the reactor, an orifice in the recirculation line to drop the pressure before discharging water into LPFT, piping to let out the discharge of safety valve at DLP suction to prevent air ingress into LPFT through safety valve gland, auto startup of CEP on low discharge pressure to take care of pump cavitation etc., were implemented. In conventional power plants CEP takes suction from (MC/DC) and discharges water into deaerator. This arrangement makes the system more simple and rugged. But this could not be adopted in FBTR as the existing CEP with low discharge head (10 kg/cm²) has to be replaced with a pump of higher discharge head (20 kg/cm²) due to high
Development of a Unique Density Equation with Temperature Effect for Spent Fuel Reprocessing, Criticality Safety and Strategic Nuclear Material Accounting

In nuclear solvent extraction, prediction of exact density of aqueous and organic phases having multisolutes is required for several applications. The very first application is for proper design of solvent extraction contactors like pulse column, mixer settlers or centrifugal contactors, where difference in the density of phases plays an important role in post-mixing separation in the respective settlers. The flooding and dispersed phase holdups are strong functions of density of aqueous and organic phases. In the second case, the density for aqueous and organic phases is predicted for inter-conversion between molal and molar scale of concentrations as solvent extraction calculations are preferentially done on a solute-free basis to allow for the volume change during liquid-liquid extraction. It has been observed that during HA cycle extraction, volume of the organic phase swells by about 4% due to loading of U (VI) and Pu (IV). Another important prediction of exact density is in the area of criticality of solvent extraction equipment, storage-tanks and hold-up lines containing fissile material like Pu-239 and/or enriched U.

In the classical literature on reprocessing and criticality safety, the density was handled in a sheer empirical way, often...
ignoring the relevant thermodynamic parameters. The equations, available in the classical literature-ARH-600, Maimon, Cauchetier, other French researchers, Sakurai and Tachimori etc. have dealt the density on two major assumptions- linear or power law behaviour of solute concentrations and often a complex relationship between the density and its temperature dependence. The databases used by various researchers were also limited mostly to their own data generated in-house.

Classically, density equations used in nuclear reprocessing are from ARH-600 (DOE Criticality Safety Handbook) or from Cauchetier (French work). Both forms of the equations are based on first-order dependence of the density on the concentration. A small thermodynamic analysis will reveal the defect of such first-order dependence based equations, since they fail to recognize the varying apparent molar volume of solute with varying concentration. The equations based on the first-order dependence predict a constant molar volume irrespective of the concentration that is theoretically as well as experimentally found to be incorrect. The variation of apparent molar concentration as a function of concentration is shown in Fig.1, where experimental data due to Kappenstien et al is compared with Masson’s equation. Recently Japanese researchers proposed a new polynomial based on complex equation, without rigorous theoretical basis and the current version of Japanese Criticality Safety Handbook uses that equation.

At Reprocessing Group, the importance of having a simpler and robust density equation was realized in late 1994, when work on development of PUREX solvent extraction simulation code SIMPSEX was under progress. Extensive literature survey was performed to collect the appropriate density data for uranium and plutonium nitrates in nitric acid from all the possible sources-solvent extraction literature as well as published data on criticality experiments.

For the density of the multi-component aqueous solution, we proposed the following correlation,

Where, a(T) and b(T) are coefficients determined from multiple linear least-square regression. r_a and r_b are the densities of the solution and the pure solvent (water in the case of aqueous solution). Although Eq.1 was formulated on the basis of Masson’s observation about partial molar volume of electrolytes in the aqueous solution, the novelty of approach is clear from the following two factors.

(a) Eq.1 has a non-dimensional form to bring the constant term in the equation to unity. Therefore, there is no base point error at zero concentrations of solutes (no offset at pure solvent conditions). This is important as recently French researchers evaluated their precise Pu solution density data with the following equation (derived from Eq.1),

(b) Temperature dependence of the density is already incorporated in Eq.1, whose related parameters are described by the following two equations:

In the Eq.1, r_a(0) is the density of water at temperature T in K. By using these systematic parametric equation forms, the usual empiricism of equations of ARH-600, Cauchetier and Sakurai-Tachimori, for temperature dependency of density, is successfully avoided. Our database consisted of 568 experimental points. The solute concentrations were uranium 0 - 604 g/L, plutonium 0-730 g/L and nitric acid 0-16 mol/L. The temperature range was 283.15-348.15 K. The mean and the standard deviations for the Eq.1 were 0.03% and 0.61% (an improvement over earlier reported value of 0.75% by us). Fig. 2 shows a plot of deviations. Maximum residual was only
0.037 g/mL and the maximum deviation was 2.8%. Even for the extreme Pu concentration of 730 g/L at 59°C, the predicted result had a very small residual of 0.00345 g/mL.

Recently, at ESARDA Symposium 2003, Tanaka and Hosoma of Reprocessing Centre, Japan Nuclear Cycle Development Institute, Tokaimura, reported benchmarking of Eq.1 (with constants as suggested by us) with the precise high Pu data. They reported that Eq.1 could predict density successfully for their Plutonium nitric solution with an accuracy of 0.2±0.5% and Plutonium – uranyl nitric solution with an accuracy of 0.3±0.7%. Their data were from two independent methods- vibrating tube density meter (VTDM) and Electric manometer and results from these two methods tallied well with in 0.1%. Due to near-perfect performance of the Eq.1 for the total data domain, they had used it to determine and evaluate possible sources of errors in their precise data. They had stated that the evaluation of possible sources of errors in measurements was a difficult task by conventional means. However with use of the Eq.1, the same could be done comfortably and with less effort. As per JNC, they had started this comparison in Plutonium Conversion Development Facility since 2001 at the most important flow key measurement point (KMP).

In the French work mentioned earlier, researchers pointed that during criticality calculations, improving the accuracy of density equation (i.e., improved prediction of H/Pu ratio) was more important than revisions in the calculational code or cross-sections. With the HANFORD benchmark experiments, they pointed that Cauchetier equation, used by them in criticality calculation, was fairly conservative and use of Eq.2 (based on Eq.1) had improved the density and H/Pu ratio prediction. Thus, it is expected that use of Eq.1 will remove the conservative nature of predictions and will help in suitably designing the appropriate safety margins.

Although, Eq. 1 was originally proposed for aqueous density of mixed electrolyte solutions, the same was successfully extended to cover a wide variety of systems i.e., non-electrolyte-aqueous, non-electrolyte-organic, organic-organic, electrolyte solvate-organic and the extremely dense systems of metal-amalgam (density ~ 14 g/mL), molten metals/ alloys and liquid metal systems. One particular example is worth mentioning. Fig.3 shows a plot of density residuals for Hg-Zn system. The Eq.1 was extended to cover organic solutions of interest to reprocessing. A single equation with given set of constants could predict densities of loaded and unloaded TBP/diluent solutions of various TBP concentrations (0-100% TBP). TBP was considered as a solute and diluent density was taken as the density of the solvent.

(Shekhar Kumar, Rajnish Kumar and S.B. Koganti, Reprocessing Group)
Confocal microscopy – A new tool in soft condensed matter

In the last decade, the traditional research areas of colloids, polymers, gels, surfactant systems, mem-branes, liquid crystals and biological macro-molecules have merged into a new research field “Soft Condensed Matter” or “Soft Matter”. The building blocks of the soft matter are aggregates of many millions of ordinary molecules and are characterized by structures having typical length scales between nanometers and micrometers. Due to the large structural length scale, the number density of their translational degrees of freedom is many orders of magnitude smaller than that for ordinary molecular materials. This and the interactions between the structural units, which is typically on the order of thermal energy $k_BT$, implies that these materials are easily deformable by external forces. For example, the shear modulus of a colloidal crystal with a lattice constant of 0.5 μm is approximately 50 dynes/cm², which is about 12 orders of magnitude smaller than that of an atomic crystal. The increased dimension of these basic building blocks essentially causes the dynamics in soft matter systems to be very slow (~ microseconds to several seconds). The information with regard to structure and dynamics at the lower end of length and time scales can be obtained using static and dynamic light scattering. In the last two decades, soft condensed matter program of Light Scattering Studies Section of Materials Science Division (MSD), has extensively used this technique and investigated structural ordering, dynamics and phase transitions in charged colloidal suspensions.

The higher end of the length scale of soft matter can be probed using digital video microscopy (DVM). However, the information from DVM technique has largely remained as two-dimensional (2D) due to limitation of loss of contrast arising due to light (scattered/fluorescent) reaching from objects outside the focal plane. The advent of confocal laser scanning microscopy (CLSM) has opened up new horizons for quantitative microscopy and is now available as non-destructive 3D probe. The light that does not come from the focal plane is rejected by CLSM and enables one to perform optical slicing and construction of 3D images. Hence, the same specimen can be subjected to multiple investigations. Since this instrument gives high contrast images at different depths (z-values) of a bulk sample, the output can be subjected for 3D reconstruction as well as further image analysis. The 3D data sets, which are not available through conventional microscopes, have created immense interest in large number of research groups to develop image-processing algorithms, catering to specific applications. The availability of such data sets has paved the way for 3D studies in the field of life sciences and soft condensed matter.

Confocal microscopes can operate in bright field as well as in fluorescence modes, hence are extensively used to image hundreds of micron-thick biological or transparent samples.

The basic principle of a confocal microscope is simple, in spite of the complicated electronic, mechanical and optical components associated with the commercial instruments. Fig. 1 illustrates the principle of a reflection mode CLSM. When a laser or any other light source illuminates a pinhole, light emerging from the pinhole passes through a beam splitter and is focused by an objective lens to a spot in the focal plane, where the sample is placed. Light reflected from this spot is partly reflected by the beam splitter towards a pinhole in front of the detector. Light reflected from any other parts of the sample, including parts above or below the focal plane, reach the edge of the detector pinhole and will not be detected. The objective lens forms an image of both the detector and the illuminating pinholes at the same spot in the focal plane, hence these are said to be confocal with each other. The selective rejection of light is responsible for sharp images and increased depth resolution of confocal microscopes. This depth discrimination property is the major reason for the advantages of the confocal microscopes.

Recently, a fast confocal laser scanning microscope for investigating structure and slow dynamics in soft matter systems has been commissioned at MSD. The scan speed of this microscope for an image size of 512 x 512 pixels is 7.3 frames/sec and is the only fast confocal microscope available in Indian research labs. Apart from its fast scanning speed, the microscope (Fig. 2) is capable of giving bright field as well fluorescence images in reflection mode with a lateral resolution of 160nm and axial resolution of 550nm, with two fluorescence channels and one transmission channel. CLSM is equipped with lasers: Argon ion laser and He-Ne lasers operating at 633 nm and 543 nm laser lines.

Charge stabilized mono-disperse colloidal suspensions are studied with great interest because of their technological applications such as optical filters, switches and materials with photonic band gaps. Under certain conditions, colloidal dispersions of monodisperse spheres self assemble into periodic structures, which are known as colloidal crystals. Further, these suspensions exhibit structural ordering similar to those found in atomic liquids and glasses. Because of this commonality, these systems are treated as convenient model systems for studying phase transitions at ambient conditions.

Knowledge of interparticle interaction is essential for understanding the phase behavior of colloidal systems. Though many investigations on bulk suspensions have indicated the possibility of a long-range attraction in the interparticle interaction of like-charged colloids, a direct evidence for its existence in the effective pair-potential $U(r)$ where $r$ is the interparticle distance) is lacking. Hence we have carried out detailed investigations using CLSM on very dilute suspension of highly charged polystyrene particles having a diameter $d = 600$ nm and surface charge density $σ = 2.7\mu C/cm²$. At very low volume fraction ($ε = 0.0001$) the many body interactions will be completely absent and effective interaction is truly pair-potential and hence it can be obtained using the relation $g(r) = exp\left(-\frac{U(r)}{k_BT}\right)$. The pair-correlation function $g(r)$, which gives the probability of finding a particle at distance $r$ given that a particle is at the origin, is calculated using the particle coordinates. So the experiment involves generalizing several thousands of images and obtaining particle coordinates by image analysis. The $U(r)$ thus obtained should correspond to only interaction between particles and should not be influenced by walls of the sample container and hence images should be recorded far away from the walls. This can only be done using CLSM as it can probe deep inside the sample. Fig. 3(a) shows a typical confocal image obtained at 600 μm deep inside the suspension and Fig. 3(b) shows $U(r)$ estimated using particle positions obtained from 4500 such images taken
Fig. 1: Schematic optical layout of reflection mode CLSM. (1): sample, (2): objective lens, (3): XY-scanner, (4): beam splitter, (5): focusing lens, (6): pinhole and (7): detector (photo-multiplier tube). The depicted beam paths show that light scattered from any other position on the sample, including those above or below the focal plane, is rejected by the pinhole, which acts as spatial filter. The illuminated pinhole is not shown in the figure.

Fig. 2: The photograph of the confocal laser-scanning microscope with dual monitors. The software controls are monitored on monitor one and the image is captured on the second monitor.

at different depths and at different times. The \( U(r) \) thus measured shows an attractive minimum, \( U_m \), at an interparticle distance of 1.5 mm. Since \( U_m > k_B T \) one is expected to observe bound pairs due to strong attraction which is long-ranged \( (R_c \approx 1.5 \mu m) \). Image shown in Fig. 3(a) clearly shows existence of several such bound pairs. These pairs are found to be stable over several seconds. These observations provide a direct evidence for existence of long-range attraction between like-charged colloids. When such attraction exists bulk suspensions of highly charged particles are expected to be inhomogeneous in the form of voids (particle free regions) with dense ordered regions under deionized conditions. Investigations using CLSM clearly showed coexistence of voids with ordered regions (Fig. 4). Existence of ordered regions is confirmed from the iridescence for visible light exhibited by the sample.

Present CLSM will also be used to study colloidal alloys, colloidal epitaxy and biological systems. CLSM is now finding application in investigating process and materials of technological importance viz., evaluation of fracture process in steels, in corrosion studies and in accurate measurement of residual strains in metal composites.

Fig. 3 (a) CLSM image showing bound pairs (marked by arrows). (b) Pair-potential \( U(r) \) vs. \( r \) calculated from 4500 such images.

Fig. 4. Iridescence due to Bragg diffraction of visible light from crystallized aqueous suspension of polystyrene particles of \( d = 104 \text{ nm} \), \( \epsilon = 0.4 \text{ pC/cm}^2 \). 3D reconstructed CLSM image from 20 optical slices revealing inhomogeneous nature of the suspension in the form of voids coexisting with crystalline regions.

B.V.R. Tota
MSD
Awards & Honours

- Dr. Baldev Raj has been conferred with prestigious MRSI-ICSC Superconductivity & Materials Science Award for the year 2005.

KALPAKKAM SCIENCE & TECHNOLOGY AWARDS

Basic Sciences

a) Shri M. Kamruddin, Shri P.K. Aijikumar, Dr. Sitaram Dash, Dr. A.K. Tyagi-2000
b) Shri K. Sankaran, Shri K. Sundararajan, Dr. K.S. Viswanathan-2001
c) Dr. A.K. Arora-2002

Applied Science

a) Shri. J. Jayapandian-2000
b) 1. Shri Anish Kumar, Smt. Vani Shankar, Dr. T. Jayakumar-2001
   2. Dr. M.D. Mathew
   3. Smt Vani Shankar, Shri M. Valsan

c) Dr. C. Anand Babu, Shri B.K. Sharma, Shri G. Mohanakrishnan, Shri A.K. Chandran-2002

- Dr. S. Venugopal has been elected as Life Fellow of Indian Institute of Metals.

- Dr. K. Bhanu Sankara Rao, Mechanical Metallurgy Division selected to receive G.D. Birla Gold Medal of The Indian Institute of Metals for the year 2004.

- Shri Anish Kumar, Non-Destructive Evaluation Division chosen for the “Young Metallurgist of the Year” award for the year 2004 by Ministry of Steel, Government of India.

- The paper entitled “Characterization of Microstructure in 9% Chromium Ferritic Steels Using Ultrasonic Measurements” by Shri Anish Kumar, Dr. B.K. Choudhary, Dr.K. Laha, Dr. T. Jayakumar, Dr. K. Bhanu Sankara Rao and Dr. Baldev Raj published in Transactions of The Indian Institute of Metals selected as the Best Technical Paper for the year 2003. The principal author Shri Anish Kumar has been awarded SAIL GOLD MEDAL and co-authors have been awarded Certificate of Merit by The Indian Institute of Metals.