Enrichment of helium from hydrothermal gases

Nisith Kr. Das, P. Barat, Prasanta Sen and Bikash Sinha*

Extraction of gaseous helium from natural sources is potentially important for its use in frontier technologies and as an essential ingredient for advanced research. In India, an attempt is being made to harness helium from hot spring gases, since India lacks helium-bearing natural gases or petroleum reserves. In this effort, we have developed and fabricated a helium-enrichment plant based on the principle of cryo-condensation. The plant has been commissioned successfully and is now in operation. Although the plant has been designed analytically, it is unique in the sense that it can enrich helium from 0.75 vol% to ~ 90.0 vol%. The basic principle, approach and philosophy of the plant have been discussed in this paper. The plant process has been computer-modelled to optimize and understand the relevance of the various process parameters . This model is capable of predicting the yield of enrichment efficiently for a pre-assigned setting of the process parameters.

THE second lightest element, helium, owing to its unique electronic configuration, is endowed with many extraordinary properties. As helium defies chemical combination with other elements and has a very low boiling point of 4.2 K, it stands out to be indispensable in frontier technologies involving space, atomic energy and in particular superconductivity. It is also widely used in many advanced researches including the gas discharge lasers for the transfer of energy to the lasing gases and behaviour of materials at very low temperatures. Its low cross-section for nuclear reactions leading to radionuclides under neutron bombardment and its high thermal conductivity make it suitable to use in nuclear power plants[1]. One of the most important applications of liquid helium is as a refrigerant for superconducting magnets, key to the superconducting cyclotron. In diverse fields such as nuclear magnetic resonance, magnetic resonance imaging and magneto hydro-dynamics studies, helium turns out to be crucial. It may well be used for superconducting power transmission cables[2]. Helium, therefore, becomes a commodity with wide potential applications in modern technology and has assumed considerable strategic significance.

The atmosphere is the prime source of all commercial gases excluding helium. Despite high solar and cosmic abundance of helium, it is extremely sparse on earth. The very lean quantity of helium (~ 5.2 ppm) in the atmosphere makes it uneconomical and next to impossible for large-scale extraction from this source. Helium is conventionally derived from petroleum gas fields, however, the geographic distribution of helium-bearing natural gas deposits is singularly uneven. The recognized helium-rich (> 0.3 vol%) regions include the middle eastern part of USA. Nearly 90% of the world's exploitation is concentrated there and the average helium concentration is approximately 0.8 vol%. Helium is also extracted in Poland, Russia, China, Algeria, Canada and the Netherlands. The average concentrations of helium in the fields of these countries range between 0.18 vol% and 0.9 vol%[3,4]. Since, in India such favourable gas deposits are not found yet, it seems logical to look for it in unconventional terrestrial sources such as the emanating gas from the hot springs as well as monazite sands.

There are nearly three hundred thermal springs scattered all over India. Preliminary investigations on thermal springs[5], in Eastern India, carried out by Variable Energy Cyclotron Center, Department of Atomic Energy, Kolkata, reveal that quite a number of thermal springs emit natural gases containing helium of significant measure. Three distinct belts of thermal springs have so far been identified in different parts of India: (1) Eastern India: West Bengal, Jharkhand, Assam and Orissa[6] (2) West Coast of India: Ratnagiri, Thane, Colaba and Surat; and (3) Himalayan Belt[7,8]: Uttaranchal (Jamunotri) and Himachal Pradesh (Sutlej, Beas and Parvati Valleys).

With a mission to extract helium, we have set our attention on two hot spring sites, one at Bakreswar (West Bengal) and another at Tantloi (Jharkhand), 25 km away from each other. The springs are characterized by considerable gas discharges. The gas compositions of these springs are given in Table 1. The rate of gas outflow varies between 200 and 500 l/h. India also has a substantial stockpile of monazite sands from which helium can be derived as a byproduct. This particular mineral is abundantly available in the beaches of Kerala.

In order to extract helium from hot spring gases we have adopted the principle of cryo-condensation to enrich helium up to around 90.0 vol% from the raw gas. To re-

Nisith Kr. Das and P. Barat are in the Variable Energy Cyclotron Center; 1/AF, Bidhan Nagar, Kolkata 700 064, India and Prasanta Sen and Bikash Sinha are in the Saha Institute of Nuclear Physics; 1/AF, Bidhan Nagar, Kolkata 700 064, India
*For correspondence. (e-mail: nkd_vec@yahoo.com)

Table 1. Compositions of hot spring gases from Bakreswar and Tantloi

Gas	Bakreswar (vol%)	Tantloi (vol%)
Nitrogen	92.20	92.00
Helium	1.37	1.26
Argon	2.10	2.40
Oxygen	0.90	1.14
Methane	3.43	3.20

move the unwanted volatiles, especially nitrogen ~ 92.0 vol% which is the major constituent of the gas, we have made use of the widely differing boiling points between helium gas and nitrogen gas. This helium enrichment plant has been developed indigenously and is presently in operation with the available quantity of hot spring gas as the feed. The enrichment process primarily involves three stages, namely, drying, pre-enrichment and final enrichment. This paper describes the details of the plant and results obtained from plant operation and our experiences. To glean detailed information of process parameters of the enrichment plant leading to the end product, a Gaussian Process (GP) model has been introduced. This model specifies the relevance of the respective process parameters responsible for the enrichment and can predict the enriched helium for fixed process parameters with defined error bars.

Raw gas collection from hot springs

The scheme for raw gas collection as existing at field site Bakreswar is given in Figure 1. Since, the raw gas containing helium is upwelling with the hot waters and migrates to the surface through the water envelope from the earth's interior, it is naturally saturated with water vapour and the main contaminants are CO_2, CH_4, Ar, O_2 and N_2. Bubbling gas from spring vents is collected via inverted funnel F and passed through an online moisture trap WT and stored in the gas holders G1 & G2 (2 Nm^3 each). Subsequently, the gas enters into the chemical trap, T, containing glycol-amine solution in the ratio of 1 : 1 in order to strip off remnant moisture and CO_2 present in the raw gas. Finally, this gas is fed into the solid dessicant tower T_s, filled with molecular sieves 13X and then it is compressed into the gas cylinders Cy, by the recovery compressor C at a pressure 32 bar g. These cylinders are brought to the helium-enrichment plant at Kolkata for enrichment of helium. The chemical traps are periodically reactivated using tape heaters H and made ready for reuse[9].

Enrichment process

The enrichment plant consists of three distinct modules, a storage unit, a drier unit and a condensation module as shown in Figure 2. In the first stage, the feed gas available from the springs at a pressure of 32 bar g is put into a low pressure gas receiver R1 wherefrom it is transferred after compression at a pressure of about 150 bar g into the high pressure gas storage unit B1 of 50 Nm^3 capacity. Once the optimum filling pressure is reached in the B1, the plant is started to produce enriched helium till the pressure in B1 drops down to a minimum operating pressure of about 15 bar g. This helium-enrichment plant can process a maximum of 50 Nm^3/h of raw natural gas to yield 0.6 Nm^3/h of highly enriched helium (~ 90.0 vol%) and 10.0 vol% nitrogen.

Pretreatment of feed gas

Like all other cryogenic plants, this enrichment plant calls for absolutely dry feed gas free from CO_2 for smooth and uninterrupted functioning of the process. In order to ensure the complete elimination of moisture and CO_2, the raw gas is first passed through twin bed drier unit (D) containing molecular sieves-13X to eliminate CO_2 and activated alumina for removal of moisture. This adsorber unit has been designed for eight hour cycle beyond which the vessel is put to regeneration. During the regeneration mode, pure nitrogen vapour available from condensation module, heated to a temperature of about 150–160°C by electric heater, is allowed to pass in the reverse direction of the feed flow through the vessel. This in turn removes the adsorbed impurities like CO_2 and moisture from the adsorbents, making the column ready for the next run.

Principle of enrichment

Helium enrichment is achieved by making use of the widely differing boiling points between helium and other unwanted volatiles having higher boiling points present in the feed gas. The typical gas compositions considered for the design of the plant are given in Table 2.

The condensation module of the plant consists of two heat exchangers, Hx-I and Hx-II, one liquid nitrogen bath housing the Hx-II, two gas liquid separators S1 and S2 and buffer vapour column S3 as illustrated in Figure 2. The dry feed gas enters the condensation module at a flow rate 12 Nm^3/h and at a pressure of 15 bar g and then passes through the main heat exchanger (Hx-I) which is a brazed aluminium plate fin heat exchanger. Hx-I reduces the thermal duty (pre-cooling) of the incoming gas from 308 K to 106 K by counterflow of the return liquid nitrogen (LN_2) vapour from the gas liquid separator S3 and from the LN_2 bath of second heat exchanger Hx-II as shown in the flow diagram of Figure 2. At this temperature all the contaminants like Ar, CH_4 and O_2 get condensed out. The exit gas, (He + N_2), at a temperature of 93 K containing 11–12 vol% helium and 88–89 vol%

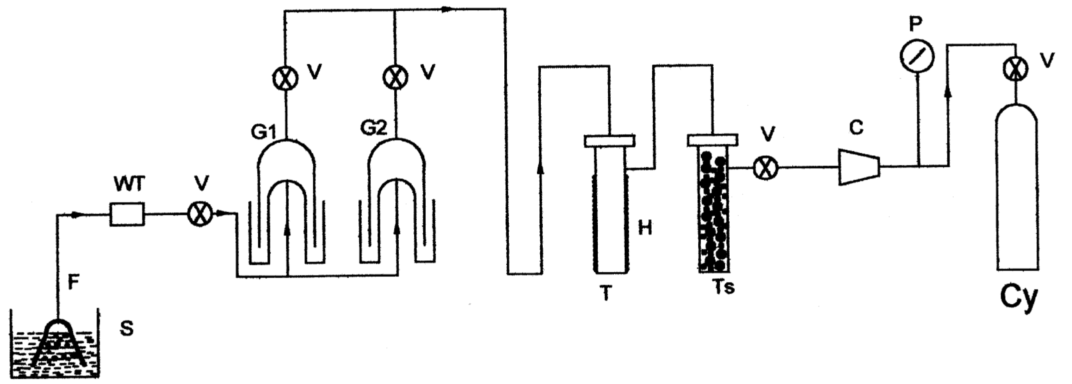

Figure 1. The scheme for raw gas collection from hot spring at the field site Bakreswar. S, Spring; F, GI Canopy; WT, Water Trap; V, Valve (Saunders); G1 and G2, Online gas holders; T, Glycol Amine trap; H, Tape heater; Ts, Solid desiccant; C, Compressor; P, Pressure gauge; Cy, Gas cylinder.

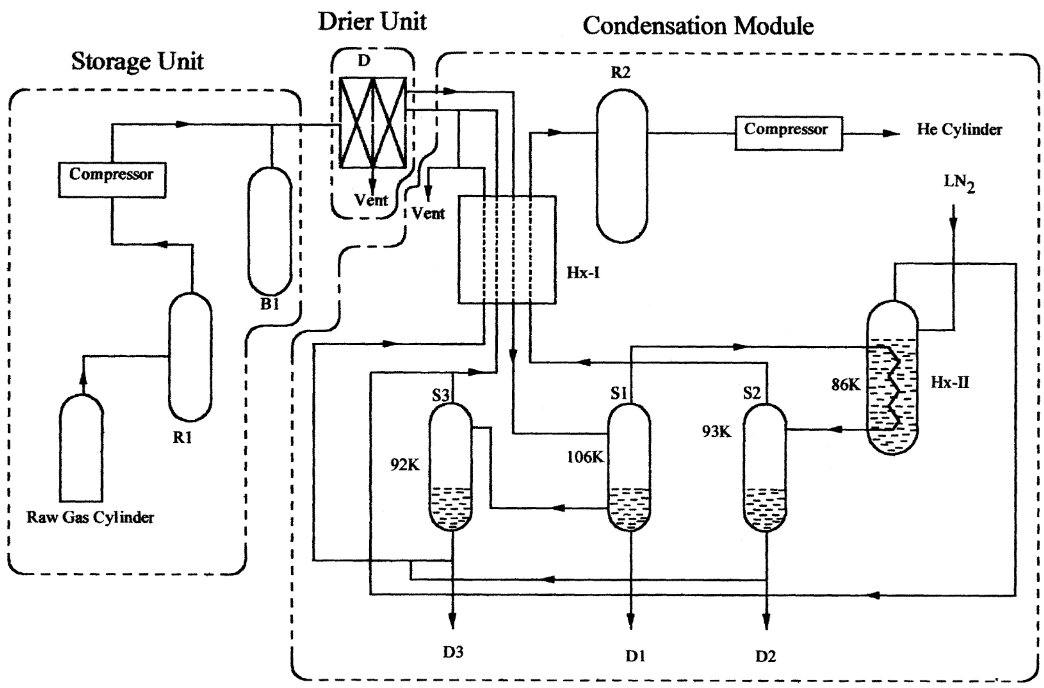

Figure 2. The schematic diagram of the enrichment plant showing three distinct modules; a storage unit, a drier unit and a condensation module.

Table 2. Typical gas compositions considered for design of the enrichment plant

Gas	Content (vol%)	Boiling point (K)
Nitrogen	90.0–94.0	77.36
Helium	0.5–2.0	4.22
Argon	1.5–2.6	87.29
Oxygen	0.4–1.3	90.18
Methane	2.1–3.9	111.70

nitrogen passes into the second coiled-tube heat exchanger Hx-II, immersed in a liquid nitrogen bath where the feed gas temperature is further lowered to 86 K. Most of the remaining nitrogen and traces of other gases become liquefied within this Hx-II and helium with small amount of nitrogen vapour is bled out to another gas–liquid separator S2. The gas chromatogram of the feed gas and the enriched helium (~ 90.00 vol%) is furnished in Figure 3. This product helium is then taken out from S2 and put into the receiver R2. Accumulated vapour from LN_2 bath of Hx-II is flashed into separator S3 wherefrom the cold gas is directed to the main heat exchanger Hx-I and is vented out. This cooled gas is used up to cool down and condense out part of the incoming gas. Perlite powder has been used as the insulation mate-

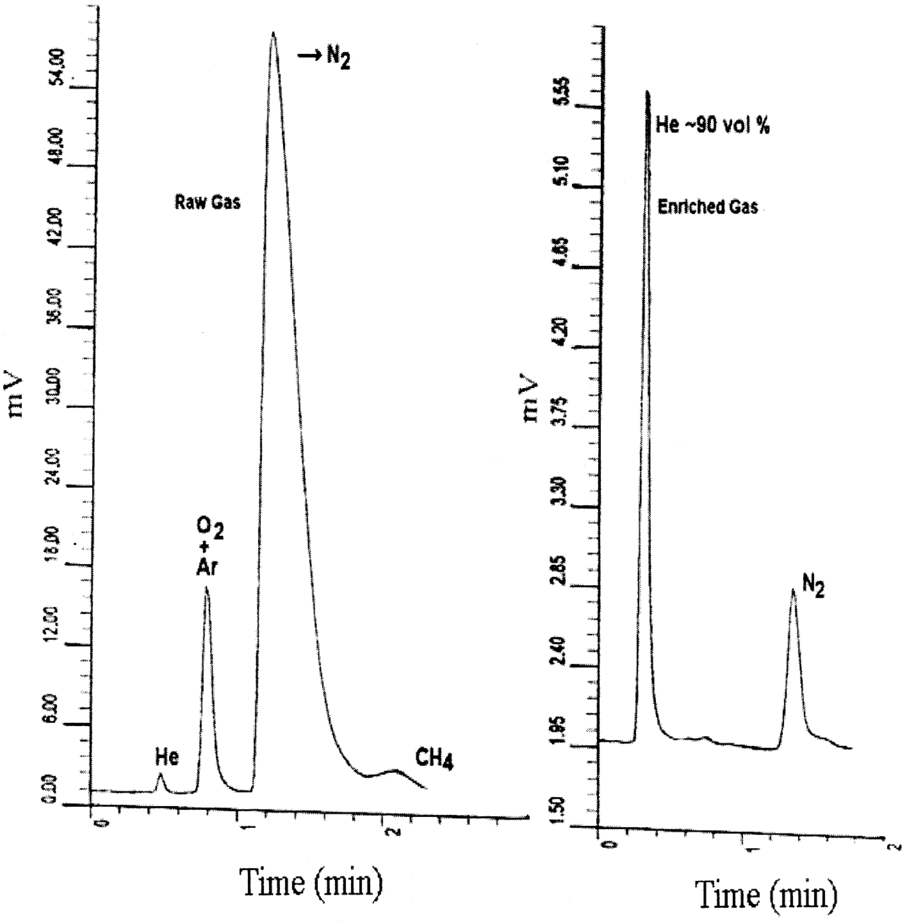

Figure 3. Gas chromatogram of feed gas from Bakreswar and enriched helium.

rial to contain the low temperatures within the condensation module and when the plant is shut down for longer periods, nitrogen atmosphere is maintained by isolating the pipelines and feeding the nitrogen from LN_2 storage container through a vaporizer.

As the basic principle adopted by us for design of the enrichment plant is based on cryogenic condensation technique, balancing the various parameters during fabrication of the plant is rather complex. We have utilized the approximate analytical relationships for different components to make the subsystems consistent with each other, however, it is observed that each process parameter has a direct effect on the level of enrichment. In order to understand the symbiotic associations of the various parameters, a Gaussian Process (GP) computer model has been implemented to study the relevance of these parameters.

Computer modelling of the enrichment plant

In modelling of complex systems, like the helium-enrichment plant, empirically, we do not know, *a priori*, the parameterized form of the input–output relationship. The Gaussian process model is a way of avoiding having to explicitly parameterize this relationship by invoking a parameterized probability model over the data instead[10,11]. Let the training data set consist of N input vectors $\{x_1, x_2, x_3 \dots, x_N\}$ each consisting of the input parameters of the plant and the corresponding set of known outputs (or 'targets') $\{t_1, t_2, t_3, \dots, t_N\}$, the degree of helium concentration. A prediction, t_{N+1}, can then be made at any new input value, x_{N+1}, based on these training data. For brevity, let x_N represent the set of input vectors and T_N be the vector of corresponding outputs. The approach of the Gaussian process model is as follows.

Let $P(T_N| x_N)$ be the joint probability distribution over the N output values in the training dataset, with a set of adjustable parameters called hyperparameters. These hyperparameters explicitly parameterize a probability distribution over the input–output function rather than the function itself. This is a probability distribution in an N-dimensional space. Training of the model is done by maximizing the probabilities of the hyperparameters. Once trained, the model predicts the most probable value of the output for a new set of inputs, together with a measure of uncertainty of the prediction. The degree of correlation achieved by given proximities of the input vectors is dictated by the hyperparameters. There is one of these hyperparameters for each input dimension. The

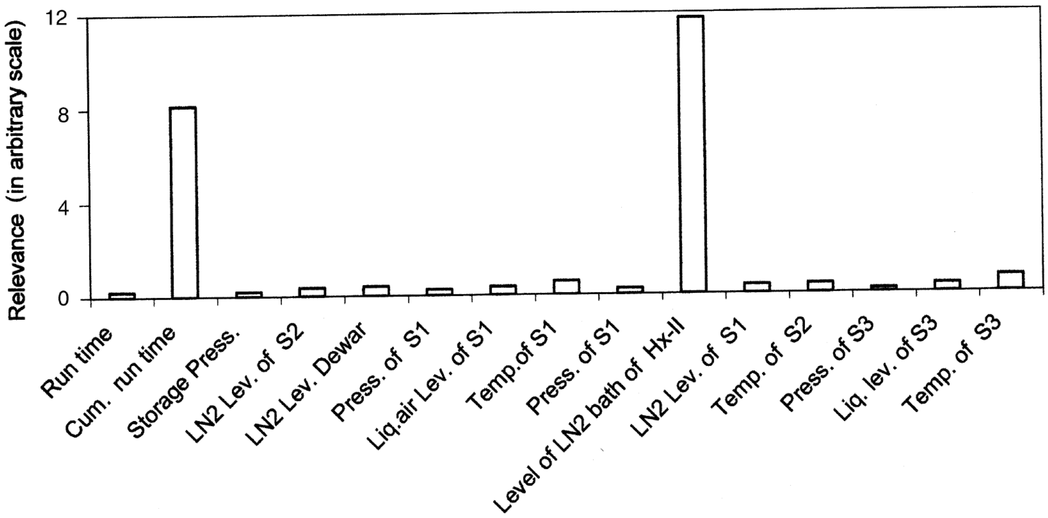

Figure 4. The relevance plot of 15 parameters of the process plant with cumulative time.

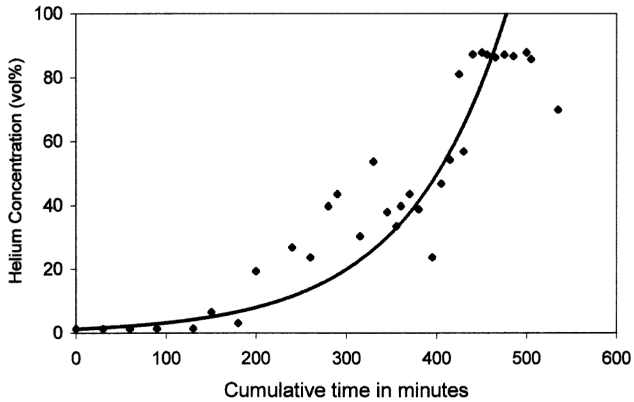

Figure 5. Plot of helium concentration against run time showing very good correlation.

relative size of the hyperparameters is a measure of relevance of each input dimension in determining the output. The model assesses the relevance of each input parameter for the prediction of the outputs from the associated values of the hyperparameters. This model also prohibits the process of over-fitting the training data, and finds the most generalized path to reach the output, taking care of error in the data of input parameters. In Figure 4 we show the relevance plot of 15 parameters of the process plant. Here it is evident that the cumulative running time of the plant and the liquid nitrogen level in the column of Hx-II are two immensely relevant parameters. From the basic idea of the cryogenic plant operation, it is quite justifiable that cumulative run time will be one of the most important variables, especially for the helium enrichment plant. This is also obvious from the plot of helium concentration against run time having very good correlation as shown in Figure 5. To study the relevance of only process variables, information of time has been removed in order to investigate the consequences. In Figure 6 we show the relevance of only the process parameters. This figure implicates that had the operation time been tacitly disregarded, the system parameters like liquid air level of S1, temperature of heat exchanger Hx-II bath and temperature of S3 turn out to be extremely relevant. This is because the temperature of S3 and liquid air level of S1 will assert the condensation of higher boiling impurity components (Ar, CH_4 and O_2) in the feed gas, while the temperature of Hx-II is crucial with respect to the liquefaction of nitrogen and thereby the level of helium enrichment. In the present study, the Gaussian probability model was run using 33 data sets of the different operating variables of the plant. In this procedure, 32 data sets are used for training and the remaining 33rd data set containing the helium percentage is predicted. This procedure is then repeated for each of the data sets. The results (predicted with 33 similar data sets) are plotted as the true values along the x-axis and the predicted one on the y-axis as shown in Figure 7. From this figure it is evident that the prediction at higher concentration is better than at the lower one. This again corroborates the fact that higher concentration of helium is achieved for longer time of plant operation as displayed in Figure 5. It is because the plant becomes thermally stabilized with respect to process parameters and the correlation with regard to the concentration of helium yield is more pronounced. However, the plant parameters do not stabilize towards the beginning of the operation and as such the helium concentration of the product is not appreciable. This has been reflected in Figure 7.

Conclusion and outlook

Utilizing the principle of cryogenic condensation, the enrichment of helium contained in the raw gas released from the hot springs at Bakreswar and nearby areas have been achieved. The plant is a non-conventional type and

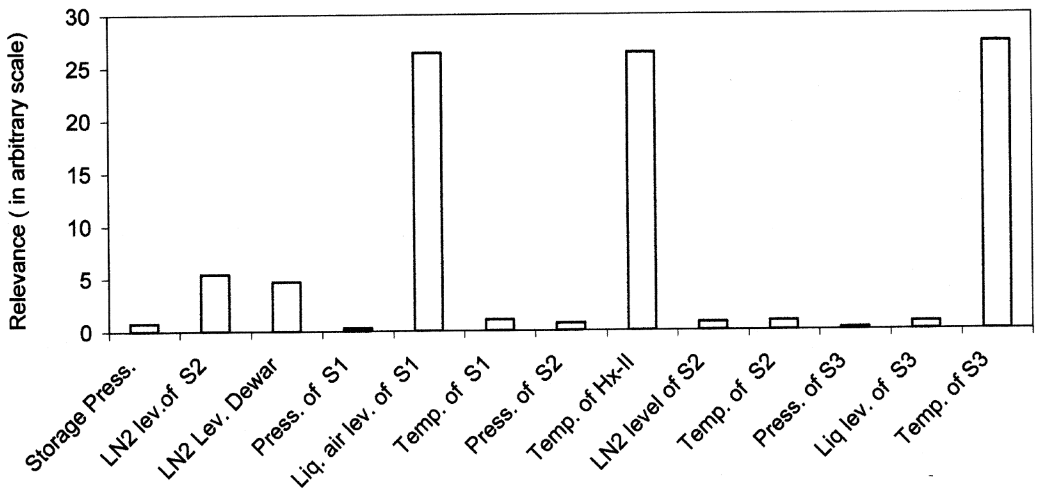

Figure 6. Plot of relevance with the process parameters without cumulative time.

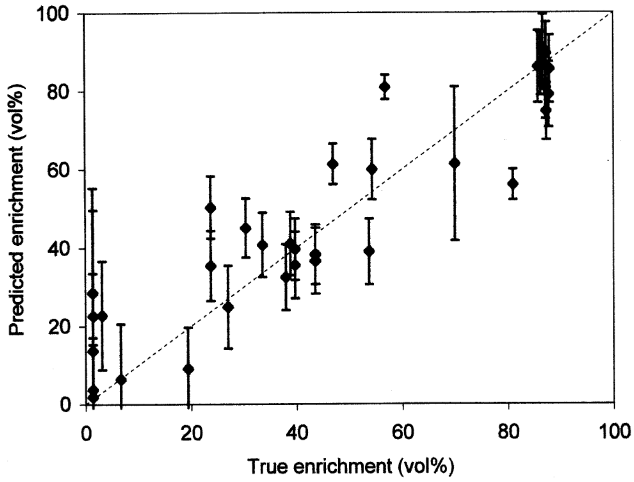

Figure 7. Predicted enrichment based on Gaussian process model plotted against the achieved true helium enrichment.

has been developed keeping an eye on our specific purpose of extracting helium from hydrothermal gases. The uniqueness of the plant lies on its capability of enriching helium from a concentration as low as 0.75 vol% to a strikingly high value of ~90.0 vol%. To the best of our knowledge, this plant is the first of its kind to establish the technology of helium enrichment in India although the process of helium extraction itself is in its early stages in our country. The plant process has been computer modelled using Gaussian process model to optimize and understand the relevance of the various process parameters.

With an eye to future activities, the infrastructure facilities at Tantloi for raw gas collection and pre-enriching the helium using membrane technology coupled with pressure swing adsorption (PSA) system at the sites has to be strengthened. The last-mentioned approach does not need any cryogenic facility and as such suits very well at Bakreswar and Tantloi sites. Helium being a strategic commodity, the setting-up of the pilot plant is quite a significant step, although the yield of pure helium (0.6 Nm^3/h in the present case) may appear to be small. Clearly, to meet the national needs for Grade-A helium only hot springs sources are not sufficient, one has to purify bulk helium from other probable sources like natural gas and monazite sands to Grade-A level to make the process commercially viable.

1. Mukhopadhya, M., *Cryogenics*, 1980, **20**, 244–246.
2. Van Sciver and Steven, W., *Helium Cryogenics*, Plenum Press, New York, 1986, pp. 7–9.
3. Monroe, A. G., Proceedings on Production and Uses of Industrial Gases, IPC Science and Technology Press, UK, 1974, pp. 123–125.
4. Jones, J. K. and Stacey, J. M., *Cryogenics*, 1974, **14**, 198.
5. Ghose, D., Das, N. K., Sinha, B. and Chatterjee, S. D., *Proc. Indian Natl. Sci. Acad.*, 1994, **A60**, 481–486.
6. Shankar *et al.*, *Geothermal Atlas of India*, GSI Spl. Publ. 1991, **19**.
7. Virk, H. S., Sharma, A. K. and Sharma Navjeet, *Curr. Sci.*, 2002, **82**, 1423–1424.
8. Singh, J. R., *Indian Miner.*, 1989, **43**, 7–18.
9. Ghose, D., Das, N. K. and Bhatacharya, D. G., *Indian J. Cryogenics*, 1993, **18**, 190–193.
10. Mackay, D. J. C., *Neural Comput.*, 1992, **4**, 415–447.
11. Mackay, D. J. C., *Network: Comput. Neural Syst.*, 1995, **6**, 469–505.

ACKNOWLEDGEMENTS. We acknowledge the contribution of Dr R. K. Bhandari, Associate Director, Variable Energy Cyclotron Center (VECC), Kolkata, during the development and operation of the plant. We thank the Department of Science and Technology and Department of Atomic Energy, Government of India, for funding the helium enrichment project. We thank the British Oxygen Limited, Kolkata for fabricating the helium enrichment plant.

Received 19 September 2002; revised accepted 5 March 2003